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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Revisiting the contribution of land transport and shipping emissions to tropospheric ozone" *by* Mariano Mertens et al.

Anonymous Referee #1

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Title: Revisiting the contribution of land transport and shipping emissions to tropospheric ozone.

Overview: This paper estimates contributions to ozone using a tagging methodology. They focus on land transportation and shipping, which are important sectors. They compare their results to comparable studies from the past and attempt to distinguish between perturbation and "contributions." The methods are generally clear and the results are well presented. There are several points of interpretation and extension of this work to conclusions that go beyond what the work supports. The main problem in this paper is cooption of terms that this reviewer believes are inappropriate. Much of

this is framing, but has important implications that need to be better fleshed out.

The field has historically estimated "contribution" in many ways including perturbation, source apportionment tagging (e.g., CAMx OSAT/APCA and CMAQ ISAM), renormalized sensitivities (e.g., DDM or adjoint). Yet this paper argues that "only tagging estimates the contribution of emissions." Note that many tagging techniques (OSAT/APCA and ISAM) have sensitivity-based metrics to account for relative importance (e.g., Sillman-ratio threshold). One goal of the relative importance approaches is to make a "contribution" that is meaningfully consistent with sensitivity because of its usefulness to policy makers. These relative importance factors are omitted in the technique applied in this paper. Why is this combinatorial tagging the only approach that can estimate "contribution"? If combinatorial tagging is somehow more appropriate, then why not include all reactants? The ad absurdum argument would then say that a large fraction of all ozone is simply natural due to molecular oxygen required for the formation of RO2. Thus, the formulation already assumes that limiting factors are important. Why is the limiting factor not important between NOx and VOC in "contribution"?

The IPCC AR5 WG1 Chapter 8 defined radiative forcing as "an instantaneous change in net (down minus up) radiative flux (shortwave plus longwave; in W m–2) due to an imposed change." AR5s definition is generally consistent with previous definitions (e.g., Seinfeld and Pandis 2006; Jacob 1999). Contribution as defined as the combinatorial tagging is not consistent with an imposed change. First, there is no imposed change. In fact, removing those emissions (tra or shp) would not impose a change of similar magnitude. Thus, the idea that transport or shipping contributes to RF proportionally to combinatorial tagging is conceptually flawed.

The authors assert that this technique is useful in understanding changes in emissions (particularly section 4.1). The current state of practice uses an emission reduction matrix to explore sensitivities at multiple emissions reductions (20, 40, 60, 80%) of both NOx and VOC. How is tagging this technique more useful than the iterative NOx/VOC matrix?

Finally, I have concern about the methodology as described in Eq 2. Apportionment based on fraction of NOy and NMHC concerns me. See Page,Line comments.

Much of this critique is specific to the interpretation and assertions of unique value. The methods and results are internally consistent. I am skeptical of the species family approach as described. The biggest issue is that the article attempts to fully own the term "contribution", applies combinatorial tagging to RF in an odd way that needs to be clearly distinguished from traditional RF, and implies regulatory value that is likely already met. Most of these comments can be addressed by revising the interpretation.

Page,Line Comments:

1,3: recommend "complementary" because the dynamics of "competition"

1,5-7: The regions are not clear in the abstract. Consider adding "ocean" to each region to be consistent with text and clarify.

1,20: This is a narrow definition of the word contribution and I have seen no argument that combinatorial tagging is the only way to define contribution.

2,15: It is not important to know "contribution" as defined by combinatorial tagging to define mitigation strategies. In fact, knowing sensitivity is fundamentally more important to mitigation since the mitigation intends to impose a change.

3,4: F should be f

4,23-5,2: If implemented as discussed, this approach assumes two things that are fundamentally at odds with our understanding of atmospheric chemistry. - First, it assumes that all NOy (NOx + NOz) is equally available for ozone production. This is problematic because NOy photochemical lifetime is much longer than NOx. As a result, this Eq 2 will attribute ozone production to NOx and NOz proportionally. That would lead to ozone being attributed to HNO3ˆtag in the mid to upper troposphere. Unless NOy is being defined differently than the field convention, this is troubling. - Second, and less concerning, NMHC are not all equally reactive nor do they have equal RO2

yields. Assuming concentration fractions are proportional to combinatorial contribution is not consistent with the chemical mechanism.

5,23: Februar[y]

6,12: Is the seasonality of non-traffic reasonable and expected?

6,24: Why is July most comparable? What did those studies look at?

7,3: Reword or edit grammar

7,8: This assumes that contribution == tagging, which the authors need to further consider.

9,1-4: Are these ratios of partial column or average ratios?

9,7: contribution...

10,2: consider replacing "almost" with "closest to".

10,7-12: Are not mitigation strategies more aligned with sensitivities?

10,20-31: See discussion of sensitivity matrix, which is the current approach for developing mitigation.

11,20: "[global] land transport." This section is tricky because the production may come from upwind sources. Try to be more explicit.

13,24: Be more specific than "some".

13,25: trough -> through?

13,25: is the author referring to engineering simplifications in the CTM?

13,28-29: CAMx OSAT/APCA[camx.com] and CMAQ ISAM [doi: 10.5194/gmd-8-99-2015] are a couple of examples of similar complexity to this scheme.

14,22-24: One interpretation is that the radiative forcing in this paper is an overestimate

due to the lack of realism in the tagging compared to an actual imposed response.

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