Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2022-438-RC1, 2022

Comment on acp-2022-438

Anonymous Referee #1

Referee comment on "An improved representation of fire non-methane organic gases (NMOGs) in models: emissions to reactivity" by Therese S. Carter et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-438-RC1, 2022

The authors have pushed forward a critical topic in atmospheric science, a full accounting of carbon emissions from important combustion sources. They have demonstrated successfully that they have made a substantial improvement to the GEOS-Chem model using recent datasets better describing speciated NMOG emission factors from fires. Armed with better emissions and some improved chemical mechanisms, the authors have evaluated the model with available measurements and then used the model to offer new insights into how fire-based NMOG emissions are distributed across the globe, and where their impacts might be felt. The paper is written well, and the study is mostly designed soundly. I have some concerns though about the model development choice to exclude fast-reacting species. I also have some presentation-relevant suggestions that follow.

Specific Comments:

1) Lines 284-292: I appreciate the authors including this detailed discussion to reconcile the new emission factor updates with the source datasets and lab measurements at a high level before introducing the CTM complexities. However, I failed to understand some key points. The new model is capturing 72% of the NMOG carbon mass, and I think the reference point for that percentage is the total of the EFs from the same dataset (i.e. Andreae 2019)? If the point of the study is that understanding and accounting for all NMOG is critical, then why purposefully leave any NMOG carbon mass out? Why not lump it with existing model species or add 1 or 2 new lumped species to capture that reactivity? The authors argue that so many of the fast-reacting species can be ignored because they will be oxidized before transporting out of the model domain, but, as the authors also state, their products (which could be longer-lived fragments) could be quite important. If the authors are sure that species won't be transported, then why not modify the chemical mechanism so that this mass can be added as non-transported species but still contribute to oxidation products that already exist in the model? As an aside, can the authors please state the contribution of the 'unspeciated or unidentified NMOG' from Coggon et al. (2019) to give a better idea of how large the gap still may be?

2) Lines 292-299: Again, why leave over half the reactivity out of the model if you already have some emission factors to go on, even if they are not global EFs. Is there a discussion somewhere in the manuscript for what is required for an EF to be global? Do you just need data for more tree and grass species? Why not run a version of GEOS-Chem with all of the red species for the simulations targeted to the U.S. measurement campaigns?

3) Perhaps some of my confusion would be resolved if the authors systematically distinguished between local-scale reactivity, which would be better constrained by Coggon et al. (2019), and downwind (or globally relevant) reactivity, which is what is being improved in this study. If the authors agree, then consider articulating this difference when reporting how much of the reactivity the model can explain in different contexts. Should we expect global models to explain all the reactivity seen in the lab? Are the ATTO cOHR measurements local or at-scale (ties into part of comment 4)?

4) Line 420-424 and Fig. 5: The color scheme is confusing. Is the red boxplot in the left panel driven by GFED or FINN emissions? If the former, then please add the FINN boxplot to the left panel. Please also add a supporting figure that shows the contributions of species (or species categories) to the GFED and FINN results so that one can see explicitly why the model update doesn't seem to matter so much. An alternative interpretation could be that both FINN, GFED, and GEOS-Chem are missing emissions of the faster-reacting species, and that these species are playing a significant role in the obs data in addition to the species that are scaled up in the FINN inventory (i.e. maybe those species are scaled up too high). A missing piece of the analysis here is the transport time from the fire to the ATTO site. How much of those fast-reacting species are still going to be around?

5) Could the authors provide a table summarizing the land-use types that are used to parameterize fire EFs in GEOS-Chem. Only three types are highlighted in the SI (Savannah, Temperate Forest, and Agriculture). What about Boreal, Tropical, etc.? It would be better to see emissions for all of the types available in GEOS-Chem. I think this would help readers connect with statements about western US EFs versus global EFs. Fig. 10 is especially hard to interpret without some context about how much variability there is now in speciated EFs across the world.

6) Figure S8 is interesting, and I think there is a lot more to discuss than what is provided so far. The discussion of this figure is limited to pointing out differences between the tropical and boreal case decay-rates. But is that because of latitude, or some kind of marine chemistry at play? Would you find the same fast loss for a boreal fire closer to the ocean? Is the orange trend emissions or is it concentration? Why is there so much scatter in the trends over land? Are these just for surface concentrations? If so, maybe column sums would be more appropriate. How do you determine how the trends follow the fire plume? It would be instructive to see how the species contributions to NMOG reactivity change along this axis, and see what notable differences there are between tropical, temperate and boreal fires. Are there differences when emissions are injected into the free troposphere vs. when they are trapped at lower altitudes? This aspect of the study could be very helpful when planning or interpreting future aircraft campaigns.

Minor Comments/Typos:

Line 34: semicolon should be a comma.

Line 47: Reads like ATTO was in the US along with FIREX-AQ and DC3. Recommend rewriting.

Line 51: exceeding à greater than

Line 68: particulate matter with diameter smaller than 2.5 microns

Lines 128-130: recommend moving 'from fires' to line 130: "…and measured OHR from fires and that furans…"

Line 198: "multiplied by its concentration as follows:"

Fig. S4: It would help to have a table that defines all of the chemical species names here. Some names are more ambiguous than others, but I think readers who aren't steeped in chemical mechanism modeling would find a key useful.

Fig. S4: Why are furans not in these figures? Can they be added? Or if they are already present, which species do they fall under?

Fig. 1a: This is a nice figure! While the gray box as defined is sort of interesting from a model development stand-point, particularly for folks used to the GEOS-Chem or global model context, I think there could be a higher-impact option. Since the take-home point (I think) is to separate fast-reacting from slow-reacting species, can you instead add regions illustrating the timescales for local, regional, continental, and hemispheric transport? Pretty much all readers should have an intuitive grasp of this, but I think it would be useful to picture it directly behind the quantified OH lifetimes.

Fig. 1a: Isoprene is blue. Does this mean that the standard model didn't have it? I'm

confused about whether blue means that it is a brand-new model species, or whether it is just now getting fire emissions for the first time.

Fig. 1b: The standard definition of the VOC/IVOC cutoff is halfway between 1e6 and 1e7 in log space, although I agree that Ahern et al. (2019) went to 1e6, as you write. Consider moving the guiding line. Also please add 298 K (I assume) to the y axis label.

Line 274: I am not yet convinced by Fig. 1 that transport is not important for all the red species. I am missing the OH concentration assumed for this axis. How consistent is that with what is seen in wildfire plumes?

Line 296: Think more about how to refer to the "minor species". They don't sound minor based on their contribution to reactivity. "minor species by mass" or just "fast-reacting species" (?)

Line 298: "all of these fractions" is vague. I think the NMOG% are relative to the EF datasets and the OH reactivity percentages are relative to Coggon et al. (2019), no?

Fig. 9: I recommend reconceptualizing this figure into a format that can be better-used as a reference. Pie charts are notoriously difficult for humans to interpret, and the large number of colors, and varying sizes of the circles make that even tougher. What about a three-panel figure of bar charts? Panel 1 could be stacked bars showing the contribution of fire and non-fire cOHR over the globe and land. Panel 2 could be fractional stacked bars showing the break-down of CO, NO2, Other, and total NMOG for both cases. Panel 3 could then be fractional stacked bars showing the contribution of each NMOG species to total NMOG. There are a lot of species, so maybe this panel could stretch horizontally across the bottom.

Fig. 10: Do you have the data from the standard model configuration to depict before and after for each of these regions? I recommend adding it if you do. Second, another dataset to consider adding is a measure of acres burned for each of these regions. This way you can depict both the potency of fires in each region as you are now and the magnitude of the signal. For example, I wonder about the abundance of reactivity exported from Asia versus Alaska and how much of the data here is adjusted low by the huge land area of Asia (I assume these data are averaged in space and time?). Finally, please grab a new color-scale. I'm not colorblind, and the purples somewhat ran together for me. Mixing the green with these reddish colors might also be problematic, but I'm not sure.

Line 548: recommend changing generally to potentially. Unless I'm mistaken, the model still hasn't been evaluated successfully in Africa for cOHR.

Line 566-567: Recommend rephrasing the comment about phenol. The model results predict phenol is important just in the boreal regions, but it also missed the phenol measurements during FIREX-AQ. I agree that it’s hard to say how much of that might be non-fire anthropogenic sources, but I encourage you not to discount phenol in the mid-latitudes or even tropics just yet. There could also be secondary production that the mechanism is missing.