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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Stratospheric pollution from Canadian forest fires" *by* Hugh C. Pumphrey et al.

Anonymous Referee #1

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The paper details multiple analyses of the lower to middle stratospheric after the Pacific Northwest Event biomass burning event in August 2017. Using data from Aura MLS, the polluted airmass is tracked around the world in the months after the injection. The authors analyze the composition of the polluted airmass and attempt to track it to its specific source. Overall, the work is sound, though more elaboration on the analyses and results are needed in various locations in the paper and the authors should provide more quantitative results on species other than CO.

General Comments

In Section 3.1 (Pg. 02, Lns. 044–51) the authors describe a technique (used previously in Pumphrey et al. (2011)) to distinguish enhanced amounts of a gas species versus

background levels. This scheme uses the mean and standard deviation in an iterative fashion to flag values that stand apart from the main distribution. This appears to be somewhat of a convoluted (and admittedly subjective) process. Have the authors thought about using simple statistical metrics more apt at detecting outliers such as the median and median absolute deviation, or is the frequency of enhancement so high as to be bimodal (in which case a completely different method should be performed)?

Are the data used in Figure 2 poleward of 25N or 15N? The caption appears to have a typo.

I assume the oscillatory appearance of enhanced CO in Figure 3 over North America is a byproduct of the MLS sampling (and not any transport behavior), but that it does reveal the meridional extent of the plume. If true, it might be worth mentioning in the figure caption as a clarification.

What is the semi-transparent red circle at roughly 70N and 125W in Figure 3?

Pg. 04, Ln. 059: ". . . the polluted airmass is divided into two parts on three occasions: at approximately 9 days, 16 days and 41 days after 12 August 2017"
What about the split at 30 days?

I was wondering what the causes of the different splits could be and whether they might play a role in the correlation between CO and H2O. For example, the split around 9 days occurs over the Atlantic and appears to be associated with some lifting. Hurricane / Tropical Storm Gert was active around that time and location, is it possible that it played a role in this split and ascent? Similarly, the split around 30 days occurs at the border of the Tibetan plateau during the Asian Monsoon (the eastward path goes around the northern side while the westward path goes south above the plateau towards lower latitudes and is also associated with some lifting).

The split at 41 days is interesting because it is difficult to tell if it really is a split. Trying to follow along between Figs. 3 and 4, the paths are disjointed. It does appear that the airmass moving East over northern Japan becomes the so-called "slower part" in Fig. 4. However, the "faster part" appears about 15 degrees North of this airmass and then subsequently moves over Alaska, Northern Canada, Quebec, before reappearing over Europe. Could the disconnect be an observation gap in MLS observations or the technique for determining "enhancement" not being sensitive enough at that time? Could the split have anything to do with Typhoon Talim that was in that area at that time (and also possibly affecting the amount of H2O)? Could it possibly not have been a split and instead been new material injected at that time, which might explain the small bump/deviation in Figure 5b around late September? Does a plot similar to Fig. 4, but using latitude instead of longitude, help visualize this at all?

Figure 5 says it shows all data poleward of 27.5N. Is there a reason this is different from Fig. 2?

Pg. 05, Ln. 068: "The seasonal behaviour of CO is fitted with a mean value and annual and semiannual sinusoids as shown in Fig. 5(a); this fit ignores the data for a period of 99 days after the PNE."
Over what total date range is this fit to the seasonal cycle determined?

Pg. 05, Ln. 071: ". . . we fit the decay with an exponential . . ."
While often popular, the decay is almost never linear in log space and allowing for some degree of curvature would be more precise. Unfortunately, this makes a simple

calculation of the decay constant impossible and likely contributes to the wide range of possibilities.

Pg. 05, Ln. 075: ". . . presumably because the plume has a small horizontal extent and lies in between the MLS orbit tracks."
Or was material was still being injected / lifted?

Pg. 06, Ln. 088: "These points are clearly not connected with the PNE so we have removed them from Fig. 6."
What were the criteria for separation and what are the origins of these removed data? Also, the authors state that these are removed from Fig. 6, but it definitely still appears to contain data that likely have nothing to do with the polluted airmass.

Pg. 07, Ln. 093: "The algorithm used fails to detect the enhancement at this time because of the very large background variability at lower latitudes."
Would a different detection method such as the median and median absolute deviation help with this?

Pg. 07, Ln. 095: "After about 50 days the water vapour in the polluted airmass exceeds the background value to a greater extent than is the case for CO. After 63 days, the polluted airmass can only be identified in the water vapour data; this is presumably due to the short chemical lifetime of CO in the stratosphere."
Might a figure showing a time-series of H2O like Fig. 5 be useful here?

If HCl is an product that is considered enhanced as an artifact, then why is it included in Figure 8 (and the only one of those products included)?

The authors state that the computed slopes of correlation are within the expected ranges. Can the authors show the expected ranges either in Table 2 or Figure 8?

Pg. 12, Ln. 161: "This suggests that over these first few days the polluted airmass is closer to 215 hPa than to 147 hPa."
Can the vertical resolution of MLS be playing a role here? Also, this might suggest that the airmass was rising in a way that the model was not properly accounting for (e.g., self-lofting of the black carbon in the plume).

Pg. 13, Ln. 190: "The injected mass is 620$\pm$80Gg, considerably smaller than the value of 2400$\pm$300Gg that we obtained by the simple method discussed in section 3.1. This is qualitatively reasonable, given that the MCMC approach only considers CO that is observable by MLS near the start of the event, while the simple method of section 3.1 includes CO that was injected at too low an altitude for MLS to observe it and subsequently ascended to observable altitudes."
I would assume that some of this discrepancy is also likely because the initial MLS measurements do not cover the full horizontal extent of the spread of the polluted airmass at this time. I believe this is what the authors are trying to say but it could be made more explicit. Additionally, regarding the comment of altitudes of injection, do the MLS observations not reach the tropopause?

Pg. 14, Ln. 207: "The mass of CO injected into the lower stratosphere by the Black Saturday fire was, when estimated by the technique of section 3.1, 1.3 Tg – just over half of the mass injected by the PNE."
Is this result from an analysis first performed in this work or does it derive from another source? If the latter, it should be referenced. If the former, then more elaboration on the analysis needs to be included than simply the result.

Pg. 14, Ln. 209: "The 2019/20 ANY fire injected a mass of CO considerably larger even than the PNE: initial esimates **[there's a typo here]**, again using the method of section 3.1, place this mass in the range 8–10 Tg."
In the same line as my previous comment, any additional analysis needs to elaborate more on at least the temporal and spatial extent of the data used. Also, while I understand the desire to compare the PNE to the more recent and even larger ANY event, the authors already claim to have a paper in preparation so I find it quite unusual to throw in preliminary results of that analysis in this paper.

Pg. 15, Ln. 232: "The location of these dots suggests that the polluted airmass observed by MLS is probably associated with Anvils 2, 3 and 4 and is less likely to be associated with Anvil 5. This in turn suggests that the emissions come from the plateau complex of fires (west of Nazko) and not from the Elephant Hill fires (north of Ashcroft)."
The analysis performed here is with MLS data that is highly localized. The analysis supports the stated origin of the polluted airmass observed by MLS, but it does not rule out the other locations as potential sources that injected polluted air into regions not immediately observed by MLS.

The entire discussion about comparing black carbon and nuclear war on page 15 (lines 240 to 247) seems very out of place in this paper.

Pg. 15, Ln. 251: "The polluted airmass was much wetter than the surrounding air in the stratosphere . . ."
How much wetter? There do not appear to be any quantitative results of H2O in this paper.

Why does the paper discuss other biomass burning products (i.e., HCN, CH_3CN, CH_3OH, and CH_3Cl) and perform correlation analyses and yet only computes an injection mass for CO? This should be performed or the title changed to something centered on CO.

It would be good to include some more results (both qualitative and quantitative) in the conclusion.

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