Author reply to comments of anonymous Referee #2 on the manuscript

"Unexpected long-range transport of glyoxal and formaldehyde observed from the Copernicus Sentinel-5 Precursor satellite during the 2018 Canadian wildfires"

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We would like to thank the anonymous referee #2 for carefully reading our manuscript and for providing valuable comments, which helped to improve the quality of our manuscript. We have answered below point by point to each comment.

We noticed a small mistake in the discussion manuscript. For the AMF calculation in the CHO.CHO and HCHO satellite retrievals, we accidentally used wrong units in the profiles used for the simulations, which created some background offset between different days. In the revised manuscript, we corrected the CHO.CHO and HCHO VCDs. This introduces only relatively small changes in the magnitude of CHO.CHO and HCHO compared to the dataset shown in the discussion manuscript and thus does not affect the interpretation of the results.

Legend:

- **referee comments**
- **authors comments**

This paper describes TROPOMI satellite retrievals of glyoxal (CHO.CHO) and formaldehyde (HCHO) over Western Canada during the wildfire-intensive month of August 2018. Enhanced VCDs of ~14×10^{14} molec cm^{-2} CHO.CHO and ~50×10^{15} molec cm^{-2} HCHO are observed at wildfire locations and these enhancements appear to persist over long distances of up to 1500 km. FLEXPART tracer transport simulations using GFAS emission locations are able to reproduce the spatial distribution of enhancements if a lifetime of 20 hours or more is used.

My general suggestion for the paper is to articulate more clearly the usage of lifetimes in FLEXPART to avoid confusion. Since a full chemical transport model is not being used, (1) the model is not producing CHO.CHO and HCHO columns that can be directly compared to the observations and (2) the 'effective lifetime' does not represent the chemical/physical production/loss processes that are occurring within the large wildfire plume. Rather, the 'effective lifetime' in FLEXPART simply allows tracer particles to persist from their origin and continue to be transported. This provides a general spatial comparison to the observations. Hence the 'effective lifetime' here is a simple and useful computational proxy – but not a representation of – complex plume processes. The authors have clarified this in the Methods but it should be made more obvious to readers in other sections.

For example, the wording in the abstract suggests >24 hour lifetimes during night-time or at high latitudes (does this refer to Canadian latitudes?) and presents 20+ hours as the FLEXPART lifetime; these are referring to the different usages described above and can be confusing.

The authors discuss the far downwind CHO.CHO and HCHO observations and suggest continued production from precursors as the likely cause, and not a physical increase in the lifetimes. It is worth noting that British Columbia is a coastal province and the presence of chlorine-initiated oxidation adds to the skepticism of >20 hr lifetimes.

We thank the reviewer for the comment. We agree that this study does not try to determine the current chemical lifetimes of these species, and the simulations are only used to describe how they are physically transported from the source of production. The estimated lifetime over which these species are observed in the atmosphere corresponds to the assumption of a simple exponential first-order decay which is not representative for the complex chemistry in the plume. We have clarified in the text what we mean by this effective lifetime in our study to avoid confusion. However, we believe that FLEXPART simulations provide an important piece of information for understanding the behaviour of the plume until it is dispersed.
Regarding the latitudes in the manuscript, these refer to Canadian latitudes. The lifetimes of VOC depend on the season and these are connected to OH variability, and thus to photolysis as well. Globally, OH significantly decreases for latitudes larger than 45°N (Lelieveld et al., 2016), which corresponds to Canadian latitudes, and thus we expect longer lifetimes for VOCs over these latitudes.

This is a relevant paper for ACP. The paper is well written and the results are presented in an organized manner. The satellite retrievals of CHO.CHO and HCHO from the recently launched TROPOMI instrument are highly valuable and provide improved insight into Canadian wildfires as presented in this work. I recommend acceptance to ACP after addressing the above comments and the minor corrections below:
Thank you very much for your positive comments.

Line 05 – 'lifetimes'
Done

Line 24 – order of CHO and HCHO is awkwardly changed in this sentence
It has been rearranged in the revised manuscript.

Line 51 – remove comma after 'Spectroscopy'
Done

Line 57 – remove comma after '07'
Done

Line 73 – 'and/or'
Done

Line 81 – capitalize 'Precursor' and remove 'of'
Done

Line 85 – remove 'of'
Done

Line 86 – keep formatting of dates consistent (e.g. 13 October 2017 vs. August 07 2018 in Line 57 vs. 10th of August 2018 in Line 140, etc.)
The text has been modified in order to be consistent.

Line 89 – 13:30 LT
The manuscript has been modified accordingly.

Line 92 – again consider removing 'of'
Done

Line 94/95 – keep formatting of in-text citations consistent
The manuscript has been modified accordingly.

Line 106 – How many CHO.CHO peaks are within this range?
In the fitting range used in this study, five glyoxal absorption peaks can be found (see Figure 2A in the manuscript), including the strongest absorption band of glyoxal.

Line 110 – What is meant by a 'row-dependant' spectrum? Explain. Is it one background spectrum per line of latitude? The author states that a daily mean is used but if the background spectrum changes throughout the day, will this introduce significant error?
Here, a daily background spectrum is computed by averaging over the whole latitude range (50° S – 50° N) for each across-track viewing direction individually. Thus, one background spectrum is used per viewing direction (450 spectra). This approach is taken to minimise small across-track dependent differences of the TROPOMI measurements.
Ideally, a solar irradiance measurement would be used as a background spectrum in the analysis. However, for weak absorbers such as glyoxal and formaldehyde, use of a daily Earth shine background derived by averaging measurements over the Pacific has proven to reduce noise and offsets in the data. Day to day changes of this background spectrum are small and are therefore expected to introduce only small uncertainties. However, over longer time periods (weeks and months), instrumental drift may induce changes in trace gas columns if the background spectrum is not based on recent measurements. Use of a daily background spectrum resulted in a significant reduction of instrumental noise similar to that demonstrated in previous studies (Schönhardt et al. 2008, De Smedt et al., 2008, Anand et al., 2015, Alvarado, 2016).

Line 110 – '...as a background spectrum (Alvarado, 2016).'
Done

Table 1 – remove 'de' in title
Done

Line 127 – heading should state 'HCHO retrieval from TROPOMI measurements' to match Line 93
Done

Line 133 – 'consists'
Done

Line 161 – full citation in brackets
Done

Line 206 – remove comma
Done

Line 211 – 'HCHO'
Done

Line 220 – reword to '...which are discussed in detail in Section 3.2.'
The manuscript has been reworded accordingly.

Line 229 – reword to 'Figure 7 presents...'
The manuscript has been reworded accordingly.

Line 231 – 'However, on the 20th...'
Done

Line 233 – 'from'
Done

Line 252 – 'references'
Done

Figure 8 caption – '...for the 10th of August 2018.'
Done

Line 286 – misspelling of 'conclusions' in heading
This has been corrected in the manuscript.

References

Anand, J. S., Monks, P. S., and Leigh, R. J.: An improved retrieval of tropospheric NO2 from space over polluted regions using an Earth radiance reference, Atmos. Meas. Tech., 8, 1519–1535, 2015.

Alvarado, L. M. A., Richter, A., Vrekoussis, M., Wittrock, F., Hilboll, A., Schreier, S. F., and Burrows, J. P.: An improved glyoxal retrieval from OMI measurements, Atmos. Meas. Tech., 7, 4133–4150, 2014.

De Smedt, I., Müller, J.-F., Stavrakou, T., van der A, R., Eskes, H., and Van Roozendael, M.: Twelve years of global observations of formaldehyde in the troposphere using GOME and SCIAMACHY sensors, Atmos. Chem. Phys., 8, 4947–4963, 2008.

Lelieveld, J., Gromov, S., Pozzer, A., and Taraborrelli, D.: Global tropospheric hydroxyl distribution, budget and reactivity, Atmos. Chem. Phys., 16, 12477–12493, acp-16-12477-2016, 2016.

Schönhardt, A., Richter, A., Wittrock, F., Kirk, H., Oetjen, H., Roscoe, H. K., and Burrows, J. P.: Observations of iodine monoxide columns from satellite, Atmos. Chem. Phys., 8, 637–653, 2008.