Author reply to comments of anonymous Referee #1 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 #1 for carefully reading our manuscript and 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 presents satellite-derived observations of glyoxal and formaldehyde from the TROPOMI instrument, over British Columbia, Canada. Elevated column densities were associated with fire hot-spots and observed over distances of up to 1500 km. Based on comparisons with FLEXPART simulations with different lifetimes, effective lifetimes of >20 hours are required to explain the observations. The authors indicate that the effective lifetimes are in contrast to the shorter expected lifetimes of these species.

My main concern with the paper is with the references to the lifetimes of glyoxal and formaldehyde. The paper does not provide adequate evidence to support the determination of atmospheric lifetimes, mainly because chemistry and deposition are not considered (and as the authors state, not within the scope of this paper). The observations of glyoxal and formaldehyde enhancements downwind of the fire hot spots are likely due to formation (and loss) processes (as the authors also note) and thus, reference to lifetimes accounting only for transport time is not appropriate.

We agree that full chemical simulations along the trajectory would enhance our understanding of the chemical transformation taking place as the fire emissions are transported. However, we consider the focus of this study was slightly different. We present simultaneous observations of CHO.CHO, HCHO, NO_2 and CO in plumes coming from wildfires. The FLEXPART simulations describe how the air masses are physically transported from the source of production, in this case, the fires. They are coupled with estimates of the lifetime of a theoretical tracer species travelling in the transported air mass. Our initial assumption, that the formaldehyde and glyoxal were produced in the fire and then transported and chemically removed, primarily by photolysis and reaction with hydroxyl radicals, clearly does not explain the observed formaldehyde and glyoxal temporal evolution. We consider that FLEXPART simulations provide an important piece of information to help us understand the behaviour of air mass plumes, as they are transported. To avoid confusion we have clarified in the text the objectives of the modelling and our use of the term "effective lifetime" in this study.

This is a relevant paper for ACP and would be of interest to ACP readers. The paper is comprehensive, well written with clear study objectives, logically presented and articulated conclusions. The satellite-derived observations of glyoxal and formaldehyde far downwind of the fire sources are quite interesting and can stand on their own without comparison to 'expected' lifetimes.
I recommend acceptance to ACP after addressing the above comments and a few minor comments below.

Thank you very much for your positive comments.

L42: biomass burning includes wildfires – what is meant by indicating both?

The sentence has been removed. What we intended to express here is that pyrogenic emissions include wildfires and agricultural fires.

L49: transported to 'those' regions – please clarify or reword
Done

Intro – break into paragraphs for easier reading
Done

L117 – any comment on the uncertainty associated with using an aerosol profile to depict the glyoxal profile?
Quantification of uncertainty associated with the assumed profile is difficult at it depends on several factors such as the geometry of observation, the presence of clouds, the altitude of aerosols, the surface albedo, etc. For this study, we consider that the most accurate approach is assuming a vertical distribution of glyoxal similar to the one measured for the aerosols. This is

because no significant contribution from other sources is expected. If there is any contribution from layers close to the ground, it is shielded by the aerosol layers and difficult to detect by satellite under the conditions of the measurements in our case study. This is because the measurement sensitivity decreases below the aerosol layer as most photons are scattered back to the satellite before they can reach these altitudes (Leitao et al., 2010). Here, a sensitivity study has been conducted assuming glyoxal profiles at different altitudes and evaluating the impact on the glyoxal AMFs. Figure 1A shows glyoxal profiles with maximum concentrations at different altitudes. Figure 1B shows the AMFs dependence with SZA for different profiles. All AMFs behave quite similar, however, for layers at higher altitude the AMFs are larger than those for a layer closer to the ground. Relative differences between AMFs were also computed using as reference the profile with maximum concentration at 2 km. The AMFs vary between 15% and 30% for small SZA but larger deviations are found for large SZA, especially for profiles with a maximum at high altitude. In general, uncertainty associated with the assumed vertical profile is one of the most significant sources of error in DOAS retrievals and can lead to uncertainties between 10 and 30% (Boersma et al., 2004; Lerot et al., 2010).

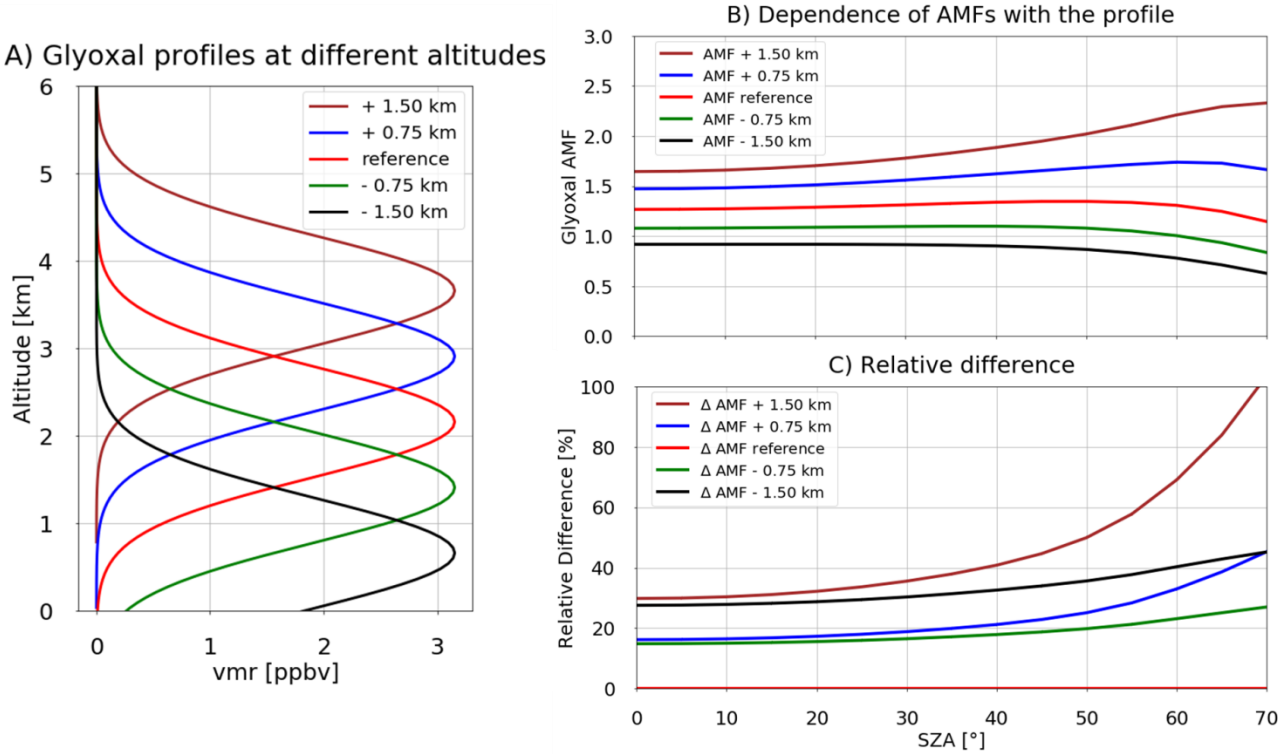

Figure 1: A) Glyoxal profiles peaking at different altitudes. B) Glyoxal AMFs computed using the profiles of A). C) Relative difference of AMFs for profiles at difference altitude against the AMF for the reference profile.

L122 – what is meant by a homogeneous distribution? The same profile is used over the geographic

region studied?

Yes is the short answer. We assume that the aerosols are distributed homogeneously in the whole region. For each day, the mean aerosol profile is computed as the average of all aerosol profiles measured in the region after removing cloud-contaminated pixels, and this profile is then used in the retrieval of the trace gas data.

L131 – how much reduction in noise? Can this be quantified?
The random noise in the large fitting range is about 4 times smaller than the corresponding value obtained using a smaller fit window. In the figure below, a comparison of the variation of formaldehyde slant column densities over the equatorial Pacific is shown. In this area, HCHO is mainly produced by methane oxidation and therefore assumed to be homogeneously distributed. Variations in the retrieved HCHO columns are thus taken as indication of retrieval uncertainty. The scatter obtained using a large fitting window corresponds to about 4.5×10^{15} molec.cm^{-2}, while the fitting window used by Vrekoussis et al., (2010) leads to a variability of about 1.6×10^{16} molec.cm^{-2}.

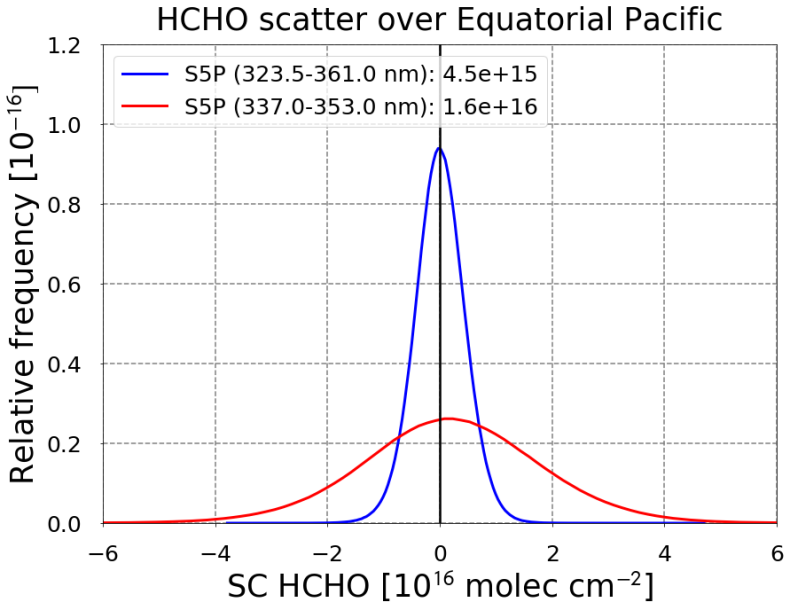

Figure 2: Distribution of S5P HCHO differential slant column densities over a clean equatorial area ocean region (5° S – 5° N, 150° – 210°) for August 2018.

L132 – 'strong absorption of the latter'; of the latter not appropriate in this sentence, confusing. Remove 'of the latter' and clarify.
The manuscript has been modified accordingly.

L150 – confusing sentence regarding lifetimes.....
The text has been modified in order to be clearer.

L156 – 'exact emissions'; what is meant by this? emission type (pollutant?) or emission rate?
Here, we meant "emission rate", which has been clarified in the revised manuscript.

L161 – reference should be in brackets
Done

References

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Lerot, C., Stavrakou, T., De Smedt, I., Müller, J.-F., and Van Roozendael, M.: Glyoxal vertical columns from GOME-2 backscattered light measurements and comparisons with a global model, Atmos. Chem. Phys., 10, 12 059–12 072, doi:10.5194/acp-10-12059-2010, 2010.

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