

Atmos. Chem. Phys. Discuss., referee comment RC2
<https://doi.org/10.5194/acp-2022-416-RC2>, 2022
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Comment on acp-2022-416

Anonymous Referee #1

Referee comment on "Airborne glyoxal measurements in the marine and continental atmosphere: comparison with TROPOMI observations and EMAC simulations" by Flora Kluge et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-416-RC2>, 2022

Kluge et al. present observation on airborne Limb and Nadir measurements of glyoxal in the troposphere from 72 flights during 8 missions on board aircraft HALO between 2014 and 2019 covering different environments conditions such anthropogenic, biogenic and pyrogenic emissions. At the same time these observations are compared to collocated glyoxal from TROPOMI measurements, which provide consistency overall agreement between data sets for most of environments with exception of biomass burning plumes over the oceans and anthropogenic plumes in the terrestrial or marine boundary layer. In addition, simulation with atmospheric-chemistry model EMAC are compared to glyoxal observations for different air masses showing underestimation from the model in comparison with the mini-DOAS observation.

This study provides a novel data set for glyoxal observations at global scale. Also, further discussion in the observed enhanced glyoxal levels that have been transported under biomass burning emissions but also over some highly polluted areas that is not present in any previous studies.

The topic of this work fits well within the scope of ACP, the main findings are extensively described, and the paper is well structured.

I recommend acceptance to ACP after addressing the comments below.

Major comments:

One of the main concerns in the CHOCHO retrieval is the impact of aerosols, specially for those emissions from biomass burning but also over highly polluted regions. Probably not accounting to aerosols could lead to overestimation of glyoxal from airborne and also satellite observation, and thus more discussion in this direction need to be introduced. Have you evaluate the impact of aerosols to the total error in your retrieval? How much is it?

Several glyoxal plumes are identify from biomass burning, but also anthropogenic emissions with four plumes corresponding to high amounts of glyoxal, and thus expected from these specific hot-spots. However, under non extreme events, glyoxal detection is low and probably model simulations could correspond more to real glyoxal amounts in average than to those specific scenarios, which could lead to a miss conclusions in this study.

Although, the authors mentioned that altitude criteria selection of flights is done, not clear reason why the difference between TROPOMI and mini DOAS come from, since both perform similar geometric observation and not difference should be expected between satellite and airborne above the altitude selection.

Finally, why the focus is only CHOCHO but not formaldehyde (HCHO), which is also possible to retrieve with the mini-DOAS system. In addition, it is well know than this is also emitted from biogenic, anthropogenic and pyrogenic emissions and behave similar to CHOCHO?

Specific comments:

P2, Line 25. 'potentially from sea', would you please clarify in which direction the differences occur

P2, Line 53. Glyoxal lifetime depends on environment conditions like urban or it is similar in all environments?

P5, Line 145, what is the effect in the glyoxal retrieval due to the low spectral resolution in the visible range?

P6, Table 1 and Table 2, why in the retrieval of glyoxal from mini DOAS system, different

cross-section is used than this used for the satellite retrieval, e.g. Pope and Fry (1997) instead of Mason et al. (2016), what is the impact of this cross-section in the validation?

P6, Table 2. Why CHOCHO retrievals are in different wavelength? Why not used same spectral range and how glyoxal depends on these changes? Could you quantify it?

P7, Line 162. Despite that the authors mention that not significantly effect is observed in the NO₂. Is the case for all type of environments? Because previous studies demonstrate that the cross-correlation between NO₂ and CHOCHO is significant under anthropogenic emissions.

P7, Line 170. As the authors pointed out that fitting window has been changed to a continues one in comparison to previous study by Kluge et al. (2020) yield to improvements in the spectral residuum and signal to noise ration, how much large are these improvements (e.g. residual and noise)? What is the relative difference in CHOCHO SCD between previous retrieval and the one used in this study?

P7, Line 178. What different ambient conditions? Is the retrieval of CHOCHO depending on the ambient conditions? If it is the case, why the CHOCHO retrieval should dependent the source producing it?

P7, Line 179. For off axis DOAS observations the impact of water vapor is significant and cross correlation is more evident for weak absorber such as glyoxal. However, in nadir observation the effect of water vapor in the glyoxal retrieval would be expected rather smaller. Can you quantify this effect in your retrieval and also have you evaluate the dependency of glyoxal retrieval on the water vapor cross-section used?

P7, Line 188. Would you please write to which location and altitude the reference spectrum correspond for the different flights?

P9, Line 240, Would you please clarify why VCD does not correspond to the total vertical column and only a fraction?

P10, Line 243. Would you please give a reference or clarification why the 8 km threshold is used for selection of for flight in the comparison?

P13, Line 338. How large is the spectral variability of Mini DOAS system regarding temperature changes in the spectrometer?

P13, Line 348. Would please describe how is applied the empirical correction in case of extreme NO₂ absorption?

P14, Line 354. Are aerosols account in the retrieval?

P17, Figure 3, The plumes 1.1, 2, etc., are not scaled? The text is a bit confuse, please make clear it.

P20, Line 495. Is the plume 1.2 also observed from TROPOMI? If yes, how this plume compare to these from Mini DOAS system? How was the evolution of the plume since the fire started? What is the age of the plume?

P21, Line 524. Why glyoxal from mediterranean Sea are large than from Amazon rainforest? Normally, glyoxal from biogenic emissions are expected higher than anthropogenic or any other source with exception from biomass burning. Does TROPOMI observe similar behavior between mediterranean region and Amazon rainforest?

P22, Line 540. Previous studies usually shown low glyoxal values over Europe and the medirranian Sea (Lerot et al. 2021, Alvarado, et al 2014, Chan Miller et al 2014), however in this study enhanced values are found over this region. How can be it explained? Could be related to the spatial resolution of satellite observations that low values are observed from Satellite for these regions?

P23, Line 594. Why do not use all the flights? Glyoxal is expected be found close to the surface for biogenic and anthropogenic emissions. To what altitude correspond those considered low flying aircraft? Do you expect glyoxal at 7 or 8 km altitude?

P24, Line 609. Although, to use large grid reduce the noise level in the satellite detection, a smaller grid could lead to more accurate comparison between mini DOAS system and TROPOMI. How the comparison looks using the smallest pixel size of TROPOMI versus mini DOAS system for large glyoxal plumes?

P24, Line 611. What is the delta time between observations (TROPOMI versus mini DOAS)? There is any criteria for it?

P24, Line 619. 3.2×10^{14} variability correspond to the standard deviation?

P27, Figure 8a. Please make more visible the legend for TROPOMI and mini-DOAS or move outside of figure.

P28, Figure 9. Although the good consistency between both data sets, the TROPOMI present more negative values than mini-DOAS. Do you know why is the case?

P29, Line 685. Has the missing of additional NO₂ cross-section impact in the mismatching between TROPOMI and mini-DOAS over these polluted region?

P29, Line 687. How is correct for aerosols in the TROPOMI and mini-DOAS retrievals?

P30, Line 731. Despite the large underestimation from the model, the variability of glyoxal from mini-DOAS is large, which also could lead to miss interpretation of the figure. For those days where the glyoxal variability is large. To what is it associated? Seems to be that TROPOMI (figure 9 vs figure 10) observations match better to the model than the mini-DOAS. How is it explained?

P34, Line 817-825. Despite that different plumes are observed and associate to specific events some of them present very high glyoxal amounts, however not for all fire events. Are those plumes observed similar spatially distributed? The amount of glyoxal depends on the precursor emitted from the fire or the type of vegetation? The altitude at which the plumes from fire are injected in the atmosphere play a role in the interpretation from fire emissions?