Response to the Reviewers

We are submitting a response to the reviews as recommended by the editor. We thank the reviewers for the helpful comments which enabled us to improve the manuscript.

Reviewer 1

Lutsch et al. presented multiannual FTIR measurements of several gases (CO, HCN, and C2H6) at ten NDACC sites located both at high and middle latitudes and reported the analysis of the long-term variability of the corresponding column abundances. The analysis focused on wildfire pollution events and included identification and source attribution of such events using a tagged CO simulation with the GEOS-Chem global model. The analysis confirms that GEOS-Chem with GFAS emissions is capable of adequately simulating the impact of biomass burning on CO column abundances in the Arctic. The presented data of multi-year FTIR measurements can be of interest to the scientific community. However, the manuscript has major flaws explained in my comments below. If the major issues are sufficiently addressed, the manuscript will likely require an additional review focusing on minor issues.

Comment 1.1 — I find that the manuscript lacks sufficiently novel findings. The main results of the analysis, at least as they are formulated in the abstract and conclusions, are descriptive and rather trivial. In particular, the fact that biomass burning plumes from fires in North America and Northern Asia can be transported into the Arctic, leading to strong enhancements in ambient concentrations of CO and other species, has been known long ago. Furthermore, the manuscript makes the impression that virtually all the established experimental facts (or their close analogs) discussed by the authors have been reported in the scientific literature previously. The authors should have tried to emphasize any new findings and to formulate a clear scientific message (or messages).

Reply: The authors have revised the manuscript to emphasize the focus of our findings and the novelty of our results. We quantify the impact of more than 60 wildfire events, globally, on 6 Arctic sites and 4 mid-latitude sites using FTIR measurements of multiple trace gas species. Previous studies have only considered individual wildfire events and at fewer sites. In addition to replicating published emissions ratios, we provide, for the first time, a comprehensive set of emission ratios across multiple wildfire events and trace gas species. The reported emission ratios agree with published values, demonstrating the ability of FTIR to capture wildfire plumes, as well as providing much needed longer-term estimates of emissions ratios and their variability. Furthermore, we identify and characterize the contribution of biomass burning to CO measurements specifically at each FTIR site.

Comment 1.2 — The manuscript would benefit if they could look deeper into the origin and nature of some episodes by addressing, for example, the following questions: How far the plumes were transported before they reached the FTIR sites? What is the typical age of the major plumes at the high-latitude sites? Are there any specific meteorological conditions that favor long-range transport of the biomass burning plumes into the Arctic? Are the emission factors specified in GFAS for HCN and C2H6 consistent with the measurements?

Reply: The authors thank the reviewer for their suggestions. We have revised the manuscript to include travel time calculations for each of the identified events and the typical age of the major plumes. The travel time calculations also enabled the calculation of the emission ratios from the calculated enhancement ratios from FTIR measurements. The emission ratios were compared to previous literature values.

Comment 1.3 — 2. The manuscript is poorly structured, unnecessarily long, and, consequently, is difficult to read. My suggestions in this respect are that the presentation of the results (in particular, in Section 3.1) has to be structurally separated from their discussion/interpretation. The analysis methods described in Sections 3.2 - 3.5 just before the corresponding results should be introduced and explained in Section 2 ("Methods"). The discussion of volume mixing ratio profiles and averaging kernels, which does not result in any significant findings, should be shortened, while the corresponding figures (Figs. 5-8) provided as Supplement. The content of Section 3.5, where GEOS-Chem tagged CO simulation is validated against the FTIR CO measurements, should be provided before the GEOS-Chem simulation is first used in the analysis (that is, before Section 3.3).

Reply: The authors have shortened and restructured the manuscript to improve its clarity. The presentation of the results has been explicitly separated from the discussion. The methods of Sections 3.2-3.5 were moved to Section 2. Section 3.5, the GEOS-Chem versus FTIR comparisons were moved as suggested. We have moved parts of the discussion related to the retrieved FTIR profiles and averaging kernels to appendix materials.

Comment 1.4 — The methods used in the analysis are questionable and need to be better justified or otherwise revised. Specifically, the enhancements of CO due to wildfire pollution events are identified with respect to the fit to FTIR data according to Eq. 3. However, this fit describes not only the "ambient concentration" of CO as apparently assumed by authors but also takes into account the contribution of biomass burning to the observed CO columns. Accordingly, the real wildfire pollution events are likely much stronger and longer than those identified in the manuscript, as, in fact, confirmed by Fig. 12. Further, while estimating the enhancement ratios for HCN and C2H6 with respect to CO, the authors do not take into account "the background" concentrations of these species. However, the simulation results shown in Fig. 12 indicate that the background concentrations can constitute more than half of the CO columns during the selected events. Therefore, the enhancement ratios reported in Table 5 can be quite different from the actual enhancement ratios in biomass burning emissions, and the corresponding discussion on page 14 is mostly irrelevant. Finally, I doubt that the lengthy source attribution procedure described in Section 3.3 is really necessary, especially taking into account the uncertainty of the simulation data. Would not the source attribution estimates be more reliable if they were obtained simply by averaging the model data over the whole event identified using the FTIR measurements?

Reply: The authors agree that Equation 3 does not only account for the ambient concentrations of each species, but also includes a contribution from the biomass burning sources. However, the contribution of the biomass burning sources to the fitted function is representative of an increase in the seasonal background of CO due to the accumulation of CO from biomass burning sources during the wildfire season as opposed to anomalous enhancements. These anomalous enhancements captured in the CO times series (as the result of a smoke plume passing over the site) are assumed to result in increased CO columns well above the seasonal background driven by biomass burning emissions. A statement on this was added to L234-237 of the revised manuscript.

The contribution of wildfire pollution may indeed be longer and stronger than that detected in the FTIR measurements, which may be limited as result of non-clear sky conditions and therefore may not capture the full duration of a wildfire plume. Furthermore, the long fire-affected periods observed in Figures 12-16 (Figures 7-10 of the revised manuscript) do not in reality correspond to the plumes observed in the FTIR measurements. The relative contribution of the the biomass burning sources have a cumulative influence, with increasing contribution to the CO column during the fire season. The detected events in the FTIR measurements are representative of an individual isolated event within the large domain of the tagged CO source regions.

The enhancement ratio does not take into consideration the background concentrations as doing so can introduce errors as a result of mixing of air masses during transport as well as the the uncertainty in calculating the background or ambient concentrations themselves. The enhancement ratio is defined as the slope or rate of change of the species of interest with respect to CO. Since, the enhancement ratio is calculated as the slope of the correlated fire-affected measurements, assuming a constant background over the fire-affected period and correcting for the background will not have an influence on the calculated slope. This assumption has been used extensively in past studies examining the enhancement ratios derived from FTIR measurements (e.g. Paton-Walsh et al., 2010, Vigouroux et al., 2012, Viatte et al., 2013, Viatte et al., 2015, Lutsch et al., 2016, Lutsch et al., 2019) and this has been noted in the revised manuscript on lines 260-264.

The source attribution discussion of Section 3.3 has been revised based on the reviewer's suggestion and added to the revised manuscript on L285-287.

Comment 1.5 — Abstract. It is not quite clear what the authors mean by saying about "ambient concentrations" in view of my previous comment. Furthermore, the manuscript discusses the column abundances, not the concentrations.

Reply: The authors have revised this statement to refer to the "seasonal background" used in this work and to also more accurately describe the method in response to the previous comment.

Comment 1.6 — P. 2, lines 15-28. Several major research directions (the Arctic climate, black carbon, boreal wildfires, and emissions wildfires) are discussed in just one paragraph. I suggest splitting this paragraph into smaller but logically linked paragraphs.

Reply: The authors have made this revision in the revised manuscript.

Comment 1.7 — Page 2, lines 34-37. I see a contradiction here: if reactive species are short-lived, then why long-range transport is a reason to measure their concentrations?

Reply: Short-lived reactive species that are measured using FTIR (e.g. C2H2, CH3OH, H2CO, HCOOH and NH3) may have lifetimes that range from days to weeks which may be long enough to undergo long-range transport to the Arctic (Viatte et al., 2015, Lutsch et al., 2016, Lutsch et al., 2019). Additionally, these species may have secondary sources within a smoke plume, while the injection of a smoke plume into the free troposphere may further extend their lifetime. Measuring these species by a network of FTIR instruments can provide insight into the potential for them to undergo long-range transport. A statement on this was added to L36-40 of the revised manuscript.

Comment 1.8 — Page 7, the last paragraph. It is not quite clear how the time series were obtained. Are the data shown in Figs. 2-4 are averages over all the measurements available for a given week over the period of sixteen years? Or, are they weekly-means averaged over the sixteen years? There is a similar question regarding the calculation of sigma.

Reply: The time series are the averages (and standard deviations) of all the measurements over the sixteen year period available for the given week of the year. The figure captions have been revised to clarify this.

Comment 1.9 — Page 8, lines 210-215 and 223-226. I suggest the authors should make a distinction between the facts evident in Fig. 2 and the assumptions which are not directly supported by the measurements discussed in Sect. 3.1.1. Section 3.1.2. How the a priori profiles were determined? Why the a priori profile standard deviation is shown only for the Thule site. What does the deviation of the retrieved profile from the a priori one signifies in the context of this study?

Reply: The authors have revised these statements (L333-334 and L347-348, respectively) where the contribution of wildfire contribution to CO is shown and discussed.

The a priori profiles are determined from a 40 year average (1980-2020) of the Whole Atmosphere Community Climate Model (WACCM). A statement on this was added to L370-374. A standard deviation was only shown for Thule as a monthly a priori profile was used, while all other sites used a single a priori profile.

Deviations of the retrieved profiles from the a priori are the result of the measurement and these profiles correspond to the column abundances presented here as stated on L375-L376.

Comment 1.10 — Page 12, line 354. The correlation coefficient value of 0.5 normally indicates a very weak correlation. How would the results of the analysis change if the threshold value of the correlation coefficient were chosen to be larger or smaller than 0.5?

Reply: We have revised the methods to use a higher cut-off value of r=0.6 to improve the reliability of the estimated emission ratios. In many cases, a strong correlation (r>0.8) was observed. It was found that a r∼0.6 generally corresponded to a fire-affected period based on a qualitative assessment of the GEOS-Chem time series. Decreasing the threshold of r below 0.5 for both HCN and C2H6 enhancement ratios resulted in a number false detections when no wildfire enhancement was observed in the GEOS-Chem time series. Increasing the threshold above 0.6 resulted in a number of missed detections, where all species were enhanced but may not be particularly well correlated.

Comment 1.11 — Page 12, lines 364-365. Why are the authors sure that AOD simultaneously enhanced with CO AERONET measurements provides evidence for wildfire emissions. Cannot these enhancements be due to strong anthropogenic pollution?

Reply: The authors agree that enhanced AOD and CO may also be indicative of anthropogenic pollution events; however, the AERONET data was used in conjunction with the GEOS-Chem time series to determine if the AOD was of wildfire origin. A statement on this was added on L237-246 of the revised manuscript.

Comment 1.12 — Page 15, line 443, and Fig.11. Can the authors explain why the oxidation of CH4 tends to have a maximum in winter when OH concentration in the northern hemisphere is minimal?

Reply: CO produced by CH4 oxidation follows the same seasonal cycle of CO. It is produced at lower-latitudes and is transported pole-ward where it accumulates during the winter months due to the lack of OH.