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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Detection and Attribution of Wildfire Pollution in the Arctic and Northern Mid-latitudes using a Network of FTIR Spectrometers and GEOS-Chem" *by* Erik Lutsch et al.

Anonymous Referee #1

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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.

Major comments

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). 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?

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 pro-

vided before the GEOS-Chem simulation is first used in the analysis (that is, before Section 3.3).

3. 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?

Specific comments

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.

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.

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?

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.

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?

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?

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?

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?

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