

Response to the Reviewers

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

Reviewer 1

The authors have made numerous changes in response to my comments, and I find that the manuscript has been much improved, with most of my concerns having been sufficiently addressed. However, I believe that further revisions (mostly minor) are needed before the manuscript can be ready for publication in ACP.

My major concern was that the reviewed manuscript lacked sufficiently novel findings. Although the manuscript has not been considerably improved with this particular respect, I appreciate that the authors have explained the novel points of their study more clearly and that they reported the emission ratios for C₂H₆ and HCN with respect to CO based on the analysis of observations of multiple wildfire events. I believe that the overview of the multi-year FTIR measurements at several sites and the estimates of the emission ratios can be of interest to the ACP audience. However, it is disappointing that despite the declared focus on the emission ratios, the revised manuscript does not provide any illustrations of the relationships between CO and C₂H₆ (HCN) concentrations. Without such illustrations, it is difficult to judge whether the emission ratio estimates reported in Table 4 are sufficiently reliable and robust. Hence, I suggest that the authors provide two additional figures (maybe as supplementary material) illustrating the relationships between CO and C₂H₆, as well as between CO and HCN for selected events for one of the sites of each type. The authors might wish to consider also my other comments and suggestions provided in the attached pdf file of their manuscript. Furthermore, the manuscript may need fairly extensive copy-editing.

Reply: The authors have included two additional figures in the appendix to illustrate the linear regressions used to determine the enhancement ratios of HCN and C₂H₆. The authors have also implemented the typographical edits suggested by the reviewer.

Comment 1.1 — L85 - This is a confusing statement in light of the above mentioned studies by Viatte (2013) and Yurganov (2004, 2005). Please revise or remove it.

Reply: The authors have removed this statement based on the reviewer's suggestions.

Comment 1.2 — L255, Eq. 2: Lefer et al. (1994) determined the enhancement factors in two different ways, one of which involves the background concentrations, and another is based on the estimation of the slope of the linear regression. I understand that the authors followed the second method, but this should be clearly explained right after Eq.2.

Reply: The authors have made this clarification after Eq. 2 in the revised manuscript.

Comment 1.3 — L263-264: I believe this is not true, because "background" (e.g. anthropogenic) concentrations of the different species can also correlate. In fact, the authors themselves recognize this possibility in Sect. 4.1 while discussing an overestimation of EnhR for C₂H₆ at Toronto. Accordingly, I suggest that the authors expand the discussion of the impact of the anthropogenic emissions on their estimates of EnhRs.

Reply: The authors agree that the anthropogenic concentrations of the different species may also correlate. However, the authors refer to "background" columns of each species as the column amount in the absence of any enhancement. The background concentration will therefore follow the seasonal cycle of the species. Given the relatively short duration of the wildfire events identified, the background concentrations are assumed to be constant. Under this assumption, the background amounts does not influence the slope of the linear regression (i.e. the enhancement ratio). The authors agree, that in the presence of an enhancement due to anthropogenic sources could influence the slope of the linear regression. This is stated in the manuscript in Section 4.1 as the reviewer has noted. However, this influence would only occur due to mixing of the smoke plume with an anthropogenic enhancement (as stated on L483-485), and not to due to a constant background. Furthermore, the relative contribution of biomass burning and anthropogenic sources can not be determined from FTIR measurements.

Comment 1.4 — L284: Isn't it a lower confidence for the enhancement ratio estimates?

Reply: The correlation criteria was applied to the enhancement ratios of C₂H₆ and HCN to detect wildfire events. However, for Ny Alesund, Poker Flat and Rikubetsu, the enhancement ratios were not calculated as stated on L283-285, resulting in a lower confidence that the observed enhancement was due to wildfire emissions.

Comment 1.5 — L349: Can the atmospheric lifetime of HCN be really so short (just days)? If so, would not it invalidate the estimates of the emission ratios?

Reply: The authors have corrected this statement to refer to "weeks", instead of "days" as reported by the cited studies.

Comment 1.6 — Table 1: These and other similar numbers are difficult to read. I suggest using other units (e.g. mg g⁻¹). The units should be indicated anyway.

Reply: The authors have included the units in the table caption. The authors have chosen to report the values in these units to maintain consistency with the commonly reported units used in the past studies that were cited in the table.