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Comment

Interactive comment on “Field measurements of trace gases emitted by prescribed fires in southeastern US pine forests using an open-path FTIR system” by S. K. Akagi et al.

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Interactive comment on “Field measurements of trace gases emitted by prescribed fires in southeastern US pine forests using an open-path FTIR system” by S. K. Akagi et al.

We thank both Referees for their excellent comments, which we have used to strengthen the paper as detailed below.

Anonymous Referee #1

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R1.1. The authors analyze three prescribed fires using an open-path FTIR placed strategically downwind prior to the fires being lit, and compare their observations during the burn to those of two close-cell FTIRs deployed to the same prescribed fires, one on the ground and one airborne. The authors clearly detail their findings, both qualitatively and quantitatively, and use their results to comment on the implications for both short term and 8-hour exposure to firefighters working on firelines. This paper connects to previous papers from this group, and even to papers already published from these prescribed fires, but the findings presented here still merit publication as they include the first open-path measurements of prescribed fires in this manner.

Quantifying emissions from evolving fires is clearly a complex problem, and the authors have described this complexity in a way that sometimes feels almost as meandering as the smoke itself. At times, the writing style feels somewhat incoherent and drifting. The worst example of this is the first paragraph of Section 3.2, which contains an overwhelming amount of information on EFs for a single paragraph, and seriously lacks direction. Please clean up this paragraph by separating it into specific topics, and then clearly and individually express these in individual, connected paragraphs.

A1.1. Thank you for this suggestion. Section 3.2 (now Section 3.3) has been heavily reorganized and broken down into subsections to follow a logical and progressive explanation of results as detailed next.

The entirety of Section 3.3 is focused on comparing OP-FTIR data with data from other platforms, namely, the LAFTIR and AFTIR. Paragraph 1 compares MCEs and general EF trends observed between AFTIR, fire-averaged OP-FTIR, and LAFTIR (Fig. 5). Paragraph 2 now breaks down the fire-averaged OP-FTIR data into “early” and “late” phase EF, and compares these emission factors with data from the other platforms (Fig. 6). Trends in MCE, EF, and discrepancies are then discussed in that order.

R1.2. Page 18503, lines 25-27: “Methane and methanol EF appear to follow a decreasing “step-wise” trend from smoldering dominant to flaming-dominant platforms,

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correlating with low to high MCE.” First, the order of the bars in Fig. 6 is confusing to me. In normal left-to-right reading, I would have probably placed the early fire information to the left, and progressed to the right (i.e, LAFTIR, OP-FTIR “early”, OP-FTIR “late”, AFTIR).

A1.2. We have taken this suggestion to place early fire information to the left and we have reordered the bars in Figure 5 to show: 1) AFTIR, 2) OP-FTIR, and 3) LAFTIR. The bars in Figure 6 have been reordered to show: 1) AFTIR, 2) OP-FTIR (early), 3) OP-FTIR (late), and 4) LAFTIR. This order was loosely based on MCE and the type of smoke sampled from the beginning of the fire (AFTIR) to the end of the fire (LAFTIR).

R1.3. Second, I disagree that the methane EF appears to follow a decreasing “step-wise” trend from smoldering (late) to flaming (early). Beyond the fact that it seems awkward to follow “late” to “early” as a progression – better to suggest that it follows an increasing “step-wise” trend from early to late – this isn’t true for Block 22b for methane, which actually has a lower “late” EF for methane than “early”. And contrary to the next statement “Trends are not so straightforward for EF(C₂H₄): : :” there *is* a consistent decrease in the EF for C₂H₂ between “early” and “late” for C₂H₂, even though the authors suggest there is not. Please clarify this.

A1.3. The reviewer is correct in that the trends mentioned for methane were not observed on Block 22B, where OP-FTIR “early” was equal to, and not less than, OP-FTIR “late”. The text has been revised to limit our discussion of CH₄ and CH₃OH trends, as these trends were not observed across all fires and all smoldering species.

R1. Technical comments:

R1.4. Page 18503, line 5 – there should be a comma after “samples”, not a semicolon.

A1.4. Thank you, this has been changed.

R1.5. Fig. 1 caption: Be consistent with the “DD Mon” or “Mon DD” date type. (2 Nov or Nov

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2: : : not both.)

A1.5. “Nov 2” has been changed to “2 Nov”.

R1.6. Fig. 2: I can’t tell the difference between the lines for Primary and Secondary in the legend. They look identical. And I assume they refer to roads, but that is also not made clear here, nor is this in the text.

A1.6. Thank you for pointing this out. We agree that the lines for Primary and Secondary appear the same in the burn block image and have removed “Secondary” from the legend in Fig. 2.

R1.7. Fig. 6: “The OP-FTIR EF have been broken down into “late” (blue) and “early” (orange) as shown in Fig. 5.” Technically, the colors blue and orange are not “as shown in Fig. 5”. Those are the primary differences between Fig. 6 and Fig. 5. Reword this.

A1.7. Thank you for this catch, this was a typo and “Fig. 5” should be “Fig. 3”.

Anonymous Referee #2

General comments:

The paper "Field measurements of trace gases emitted by prescribed fires in south-eastern U.S. pine forests using an open-path FTIR system" by Akagi et al. presents a set of open-path FTIR measurements of fire chemistry that are part of a comprehensive measurement campaign of three prescribed fires. The data are valuable for understanding fire chemistry and the impacts on human health, and will be useful for comparison to future field campaigns and for modeling studies. The paper should undergo some reorganization, clarification, and redundancy reduction before publication. The discussion of trends in emission factors of some compounds between the early and late periods is not robust (highlighted in specific comments), and should be revised and the statistical significance more clearly shown. With these adjustments, the paper makes a strong enough contribution to the field to merit publication.

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Specific comments:

R2.1. 18490 L15 - clarify the meaning of actively in "actively located individual residual smoldering combustion fuel elements"

A2.1. By "actively located", we mean to say that RSC sources were located based on the observation of a visible smoke plume that was usually whitish colored. We have eliminated "actively located" in the abstract and elaborated on its meaning in the text where this phrase first appears on P18492, L10.

R2.2. L25 - Clarify what is meant by platform differences. This is a significant point in the abstract, but the meaning is unclear.

A2.2. We now clarify: "... the different fuels sampled also likely contributed to differences in emission factors observed between measurement platforms."

R2.3. L27 - Delete "also" from this sentence.

A2.3. Done

R2.4. P18491 L12 – The "beneficial role(s) that fire plays in fire-adapted ecosystems" should be stated explicitly in this sentence.

A2.4. The beneficial roles of fire in fire-adapted ecosystems are elaborated on in the next sentence. These two sentences were combined to improve flow.

R2.5. P18492 L10 - Define "actively located".

A2.5. As mentioned above, "actively located" was elaborated here in the text.

R2.6. P18494 L1 - How often were the ambient emission spectra collected? Was the frequency high enough to account for the effect of changes in the ambient temperature over the measurement procedure?

A2.6. Ambient emission spectra were collected either 2 times a day on 30 Oct and 1 Nov (in the morning and after lunch) or just 1 time on 1 Nov. This is likely sufficient,

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given the change in absolute temperature over the course of the day was quite small.

R2.7. P18495 L12 - The acronym for excess mixing ratios (EMRs) should be defined in section 2.1 where the procedure is described, not in the beginning of 2.3.

A2.7. The definition of EMR has been moved to Section 2.1

R2.8. P18495 L25 - What could the potential consequences of this be for the accuracy of your results? "For NH₃ and CH₃COOH, for unknown reasons, there was a large positive intercept in the plots versus CO so the intercept was not forced."

A2.8. Thank you, we have added to this sentence so it now reads: "For NH₃ and CH₃COOH, for unknown reasons, there was a large positive intercept in the plots versus CO and the intercept was not forced, but the slope was still well-constrained and provides our best ER estimate."

R2.9. P18496 L8 - Define "SNR".

A2.9. Done

R2.10. P18498 Sec2.5 - The first paragraph of the "Three-pronged sampling approach" section is almost entirely of results and should be moved to section 3.

A2.10. The body of this section has been moved to the Results section (Section 3.1).

R2.11. P18498,L25 - P18499,L3 - The point of these sentences should be clarified. The justification is not clear for why the stated expectation was not met. This belongs in the results section.

A2.11. We now clarify this in the text.

R2.12. Sec2.5.1-.3 - These whole subsections could be moved to the results sections. Or, the information about the fire, strategy, and meteorology could be retained in section 2, but results regarding the MCE should be moved to the results. I suggest the former.

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A2.12. Sections 2.5.1-2.5.3 have been moved to the Results and are now Sections 3.1.1-3.1.3.

R2.13. P18501, L9 - The EF results in Table 1 should be presented more in the text before the discussion that occurs following L9. It seems the contents of Sec 3.1 and 3.2 are mixed here. If discussing the difference between early and late, Table 2 should be introduced earlier. The 3.1 section needs to be much more clearly organized to clearly present the results of the study (MCE, EF, and ER) and to discuss the early/late differences. The heading "initial emissions" does not match the content of this paragraph.

A2.13. The reviewer is right in that Sections 3.1 and 3.2 discuss similar concepts but these ideas are essentially blended together into one large paragraph that lacks focus. Section 3.1 (now Section 3.2) has been reorganized to only focus on OP-FTIR initial emissions presented in Table 1, and only the OP-FTIR early/late columns in Table 2. This includes a comparison between the early/late emissions differences. The focus of Section 3.2 (now Section 3.3) is to present EF from different measurement platforms (rest of Table 2) and to provide a comparison of EF between these platforms (AFTIR, LAFTIR, and how these compare to OP-FTIR).

R2.14. P18501, L15 - explain what is meant by other platforms.

A2.14. "...including airborne (AFTIR) and other ground-based (LAFTIR) platforms." was added after "from other platforms".

R2.15. P18502, L9-12 - The reason that the different instruments should sample different fuel types was not clear to me in the preceding text. Please explicitly state this in this section or with the information in section 2.5. It is unclear whether the discussion of measurement losses is the explanation for the trend presented in L12, or if the fuel type argument is a different one. Please clarify. This paragraph should be split into two or more based on topic.

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A2.15. We have added some info on the vegetation sampled by each technique to the new Section 3.1 to clarify that each technique sampled not only different emissions with different MCE, but also different types of fuel.

R2.16. P18503, L14 - What is meant by "significant" here? If not statistically significant, please reword to indicate the specific meaning.

A2.16. The sentence has been reworded

R2.17. P18503, L21 - Is this statement supported: "we see higher EF for some smoldering compounds like methane and methanol late in the fire associated with lower MCE"? The trend for CH₄ EF between early and late are not consistent for each site. Are the differences statistically significant for CH₄ and methanol? Any modifications to the text should also be reflected in the introduction and conclusion.

A2.17. The Reviewer is correct and there are no consistent trends observed for CH₄ and CH₃OH "early" and "late". We have added this to the text.

R2.18. P18504, L5 - Please postulate why the "late" OP-FTIR samples deviate from the apparent EF vs MCE trend observed for other cases. The deviant late OP-FTIR measurements do not support this statement: "consistent relationship between the EF obtained and the flaming to smoldering ratio each instrument can sample."

A2.18. Fires are highly variable natural phenomena and so strict adherence to trends is not always observed or expected. Figure 7 has been removed to limit our discussion of "early"/"late" trends, which were not consistent across species and fires.

R2.19. P18504, L16 - Your definition of MCE is not the same as dCO/dCO₂. Are you presenting the dCO/dCO₂ in parenthesis or after the commas. Be explicit, consistent, and clear. Could just present MCE?

A2.19. This sentence has been reworded:

"The fire-averaged MCE and dCO/dCO₂, respectively, from Wooster et al. (0.913 ±

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0.026 and 0.095)...”

R2.20. P18505, L2 - Which emission factors from this work are within the natural variability? All the species listed in table 1 except for NH_3 ? How is the natural variability of an EF determined? Please add relevant citation.

A2.20. The emission factors referred to here are the limited species reported by Wooster et al., and include: CO , CO_2 , HCHO , and CH_4 . This sentence has been revised to specify this: “Emission factors from this work are all within the natural variability of EF (computed as the $1-\sigma$ standard deviation of fire-averaged EFs) reported by Wooster et al. (2011) (i.e. CO , CO_2 , CH_4 , HCHO), except for NH_3 .”

R2.21. P18507, L25 - I find the terminology normalized excess mixing ratios ($dX/d\text{CO}$) confusing because it is equivalent to your defined molar emission ratios (dX/dY), which were not referred to as normalized excess mixing ratios. Shouldn't this published $dX/d\text{CO}$ be referred to as a published molar emission ratio (ER) or (PER)?

A2.21. We use the term “normalized excess mixing ratio (NEMR)” as an umbrella term that encompasses the molar emission ratio. The molar emission ratio is a special and specific NEMR that is always calculated from measurements of initial emissions, and thus, it is given a unique term to denote this. The term NEMR is important to retain since we often discuss downwind/aged emissions, and comparison of NEMRs collected at different points of an aging plume can provide insight regarding species degradation/formation. We retain the terminology presented in the paper to be consistent with previous publications and other works in the literature, however, we clarify this at L25.

R2.22. Sec 3.4. I would consider moving the first three paragraphs of this section to the end of the introduction.

A2.22. The first three paragraphs have been moved to the introduction.

R2.23. Conclusion - Should be shortened by removing redundant portions that restate

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results.

A2.23. We have shortened the conclusion.

R2.24. Table 1. Define "NMOC" in table header or footnote. The table heading should be changed to more clearly reflect which data are MCE and which are EF. You could have the MCE in bold for example and make that clear in the table heading. The heading should either consistently use acronyms MCE and EF or the full names, not this mix. Same comments for Table 2.

A2.24. These changes have been made in Tables 1 and 2.

R2.25. Figure 6. The standard deviation is very difficult to see in this figure. Please change the scale to make it clear which differences (especially between early and late OP-FTIR) are significant.

A2.25. Figure 6 has been redone to make the std deviations more visible by adjusting the location of the y-axis break. Also, the font size has been increased.

R2.26. Figure 7. The correlation coefficient or R2 should be shown for the fit to support that

stated strong negative correlation.

A2.26. Figure 7 has been removed.

R2.27. Table A2, Table4 and P18507-8 text. Please be consistent and either use 'estimated'

or 'calculated', but not both.

A2.27. Thank you, 'estimated' will be used throughout.

R.2.28. Table S1, supplemental file Why are some numeric cells italicized? If there is a meaning

please define, otherwise format uniformly.

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A2.28. Thank you, there was meaning anymore and and the formatting is now uniform.

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