There is no clear outline presented in the manuscript, so I had to create my own (copied below) in order to grasp the manuscript completely. The manuscript includes two separate methodology sections (Section 2 and 6) and two separate theory sections (Section 4 and 5), and their arrangement and transitions left me frequently confused.	*We have included "An Outline of This Paper" immediately after the first paragraph, following the style suggested by R1. We will reformat this as AMT will ultimately decide. We believe that leads us to omit paragraphs Section 1.1, L120–L140, which are over-detailed superfluous.*
Additionally, there are many instances of parenthetical asides, notes, and comments (e.g., L362-365, L399-409, L422- 428, all of Section 6.4) that interrupt the flow of the manuscript and greatly impede its overall understandability.	*These instances and one other have been replaced by a named section of the Supplementary Material, e.g. "*See also SM for a Note on Initial Point." at L362. *I have attempted to make all such references minimally disruptive to the flow of the paper. Following AMT guidelines, they are not fundamental to advancing the arguments of the paper.*
The conversational tone of this manuscript additionally introduces confusion. For instance, L203 stats "We now move to…" and it's unclear if this means in the following paragraphs or in the next section. In L312 the phrase "Recall that…" is unclear.	*These are restated.:* *L203:* The next section provides motivation for and understanding of an alternate approach … *L312:* With this section, we illustrate tracer relationships that define our approach to EnRs and EFs in more detail and also in more difficult circumstances, e.g., where the MCE is difficult to estimate, for example because its range of applicability during continued flight sampling is not clear.
Also, the included figures are very difficult to understand, in part because their text, captions, and legends are frequently too small to read (esp. Figures 4, 8, and 9) and because full explanations of what are in the figures are found both within the figure captions themselves and within various portions of the manuscript body. Overall, these makes the manuscript difficult to follow and the presented scientific concepts and results difficult to understand.	*The figures have been largely redrafted to have larger text.* *Figures 4 and 9 have been redrafted to show labels more clearly. (An remaining error on some time markings will be corrected.)* *Explanations of Figure 4 are expanded:* **Figure 1.** (a) Timeline of sampling, for the period shown in Figure 3a, Montana, of CO_2+CO (blue, left axis) and the fire tracers CO and b_{scat} (red and green points, right axis). Orange-filled points were identified as clear plume points. Unfilled points were not, but might have some fire influence, especially near plume points. (b) scatter diagram of CO vs CO_2+CO with

	arrows showing the time progression of aircraft sampling of identified plume points. Colors provide a key to times shown in (a). (c) a similar diagram of b_{scat} vs CO_2+CO. Similar shapes of figures are noted in the text. (d) Timeline of sampling for the period shown in Figure 3b, Coastal Transect. (e) scatter diagram of CO vs CO_2+CO during the transect, like (b). (f) a similar diagram of b_{scat} vs CO_2+CO for the Coastal Transect. The black bars graphed in (a) and (d) are estimates of non-fire influenced C_{bkgd}, see text. They and the non-plume points suggest air-mass changes in CO_2+CO. *Figure10 has been made larger, and a large display in the published paper is recommended. When points representing different tracers overlap in the figure, this truly signals something about the excellent precision of the individual measurements, and we do not attempt to distinguish them. The figure caption has been expanded:* **Figure 10.** (Lower panel). Estimates of the 422 background $\hat{x}_i^0$ = CO_2+CO concentrations implied based on the 10 fire tracers indicated in the legend. Individual $\hat{x}_{ij}^0$ are shown by overlapping colored bars (–), with the median estimate indicated by a black bar. (Upper panel) Estimates of $C_{\text{burn}} = x_i - \hat{x}_i^0$ indicators of fuel carbon burned, in green line. A preliminary estimate of C_{burn} based on the consensus of tracer deviations (without variable EnR estimates) is also shown. Flight days are indicated by the days marked on the top axes, and individual plumes, separated by non-plume concentrations of longer than 10 minutes, are shown as vertical separator lines. A set of horizontal lines at ~400 ppm indicates selected intervals for optimizing numerics (see text, Section 6.3, item 7)..
I feel that there are two different manuscripts here, or at least one manuscript with a large appendix or supplement that includes the majority of the theory (Sections 4, 5, pages 12 –	*This is well-considered, but the authors find few other options. We have put much more into the Supplementary Material.*

19). The forthcoming paper (Chatfield and Andreae (2019) appears to be a useful companion to this manuscript, and it is referenced several times (e.g., L669-672), but it is unclear if the two papers are meant to be considered together or if they are stand-alone manuscripts. While I believe that this manuscript has significant scientific value and falls within the scope of AMT, and that the work described and methodology proposed (the MERET method) has substantial value, the current structure and length imposes a significant impediment on its understandability and impact. There were many times in which I was confused or lost, and so while I feel like I understand much of what was presented, I am not confident that the manuscript has successfully communicated all that the authors intended. As such, I feel that significant reorganization and clarification is needed before this can be recommended for publication.	*Consequently,* *(a) Material not strictly necessary has been moved to the material.* *(b) A table of contents has been included, following the reviewer's first comment and suggestion above.* *(c) The fact that the paper contains a development of plume theory is more prominent in the abstract:* A new theoretical development of plume theory for multiple tracers is developed after examining the aircraft samples *If the editors of AMT allow, we could change the title to:* Theory and Estimation of Emissions Relationships in Forest Firea Plumes: 1: Reducing Effect of Mixing Errors on Emission Factors *(d) The authors do not think that the theory could stand alone without showing that it leads to apparently good statistical estimates. and are unwilling to begin the whole AMT review process again if we suggest a division.*
The scientific value of understanding forest-fire plume properties, and in particular of quantifying the enhancement ratios (EnRs) for properties of interest via the MERET method, is very high and this manuscript is a significant contribution to the field. The descriptions of the relationships between EnRs, ERs, and EFs in Section 1 is informative, although it would be particularly valuable if additional descriptions of how EnRs "approximate emission ratios (ERs)" (L77) if they are sampled before atmospheric transformations can occur. What is the relation after transformations? This needs to be made clear in the introduction.	*Besides rewriting the paragraph, we have added a note to the Supplementary Material which clarifies this:* 'More on the relationships of EnRs, ERs, and EFs is found in the Supplementary Material (SM), "Note on EnRs and ERs". ' *The reviewer appears to want more information about when ERs can be larger or smaller than EnRs. This seemed appropriate for a note. A helpful suggestion!*

The interpretation of Figures 4b,c,e,f in Section 3 is extremely valuable, but I largely struggled with understanding what was being represented until the description of the different examples later in the manuscript (esp. Sections 4.2 and 5). Only on a second read-through was I able to follow the text and more completely understand what is presented in Figure 4.	*We thank the reviewer for this observation. We have rewritten the introductory paragraph:* This section provides some examples of C_{tot} and fire tracers. It illustrates the limitations of changes in C_{tot} along a sampling path as an indicator of fire influence, C_{burn}, for emissions estimation and the much greater similarities of the such changes of tracers that possess shorter transformation time-scales. These define our approach to EnRs and EFs. The relation of fire emissions to observed C_{tot} to C_{burn}, can be apparently simple or complex, depending on how the history of non-fire CO and CO_2 entrained into fire plume air parcels affects C_{tot}. We show this commonality of relationships will to motivate the theory of expanding plumes in Section 4. That theory will suggest a method worked out in Sections 5 and 6 to find the key variable, C_{bkgd}, that then provides C_{burn} and thus EnRs. *We have also edited several places succeeding paragraph, not described here..*
• L54: "Chatfield and Andreae (2017)" should be "Chatfield and Andreae (2019, in preparation)" • L66: "DCO_{tot}" should be "DC_{tot}". • Table 1: The line labeled "Proportional to carbon burned: define" is confusing. What does define mean here? Is this a typo? • Figure 2 refers to a slope of 32.60458 while the text (L299) refers to a slope of $33x10^{-3}$. This inconsistency is confusing. • The variable C_j used in L417-418 and other lines does not appear in the Table of Symbols (Table 2) and is only described on L418 • L425: "...the same plume. *provided* we..." is confusing	√ $\Delta C_{tot} = \Delta CO_2 + \Delta CO$ *Yes, a typo. Now* *Proportional to total burned material, as measured by C_{burn}* *Chose ppb/ppm rather than ppm/ppm* *Included.* *Changed. Remarks placed in the Supplement. After the equation (12) we now have:* For periods of expansion in which the entrained

• • • Figure 6 has an x-axis label of C_{tot} while the text (L469) refers to C_{burn} • L659: "However, we let the define the types..." seems to be missing a word. • I believe "Figure 9" on Line 733 should be "Figure 8"	concentrations are constant. See also SM: Note on Varying Entrainment *Changed.* However, we let the statistical technique define these types, *Changed*
• The variable C_j used in L417-418 and other lines does not appear in the Table of Symbols (Table 2) and is only described on L418 • L425: "...the same plume. *provided* we..." is confusing • • • Figure 6 has an x-axis label of C_{tot} while the text (L469) refers to C_{burn} • • L659: "However, we let the define the types..." seems to be missing a word. • I believe "Figure 9" on Line 733 should be "Figure 8" •	*This has been added. The variable is the constant of integration and is generally replaced by* $a_j = \exp(C_j)$ *Now in Supplementary Material, This now uses alpha and beta for different possible positions, values of i, and re-worded "α and β, in the same plume. These are supposed chosen so that we know that x^E and all the y_j^E remain coanstant.* *Both Ctot and Cburn are used. The x axis has Ctot , units, while the increment beyond the vertical axis at 380, shows Cburn. This is now indicated.* "However, we let the statistical technique define these types, and so apply basic clustering techniques." We also added a sentence soon afterward: "NMF and *k-means* clustering are shown to be equivalent in cases appropriate to our work (Ding et al., 2005)." *Yes, Figure 8, thank you!*

• The phrase "affine dependence" is used several times L522-523: The suggestion that the reader should make their own calculations in order to understand the linear responses is unhelpful. •	*True. "Linear" has several meaning in English. See Wikipedia. So we have now* "An affine dependence (linear polynomial relationship including an intercept). *Linear Transformations in linear algebra must omit the intercept, hence the unusual phrasing "linear polynomial." Chatfield has considered the ramifications of this dependence NMF linear transformations considerably*. *The reader is relieved of calculations now:* "Some similar calculations make it clear that the estimates respond in an appropriate averaging manner under varied assumptions." We simply emphasize the linearity of the analysis.
• (e.g., L145) and is unfamiliar to me. • In Section 1.2, there are many places where I get lost. For instance, the equation on L168 lacks a sufficient description and I'm unsure what the "aj <-- CO" and "aCO <-- (fire – added CO2 + CO)" terms mean. I feel a more complete explanation is needed. • The use of the variable x for Ctot in Section 1.2 and other places is confusing, especially when Ctot and x are used together (e.g., L153-158). •	*We have added an explanatory phrase: "*the slope $a_{j\leftarrow \mathrm{CO}}$ of the regression estimates of an EnR of the species with respect to CO, multiplied by an attempted very careful estimate of the slope $a_{\mathrm{CO}\leftarrow(\text{fire−added } C_{\mathrm{burn}})}$ EnR of CO with respect to fire-produced C_{burn} . The $a_{\mathrm{CO}\leftarrow(\text{fire−added } \mathrm{CO_2+CO})}$ was described using the Modified Combustion Efficiency,…"
L528-529: I do not understand what is meant by "provides safety against a variable and incompletely described background" or "The median is not affected by undetected changes in background..." •	*A good observation. We also needed to explain why we were concerned about this. I have changed this to* "This graph also suggests that if there are more than three tracers (we use 8), then the median of all the estimates, median ($\hat{x}_{ij}^{0}$), is robust against errors resulting if a tracer *j* has a variable or poorly described background resulting in $\hat{x}_{ij}^{0}$ at falling distinctly higher or lower than the others. We must be concerned about this since tracers can have occasionally important non-fire sources."

Figure 4 is extremely difficult to understand as there is almost no description in the caption itself; the descriptions and explanations are found within the text body. Specifically: • The text and images are very small ○ The label "b_{scat}" in Figure 4a,c is too small ○ The number labels in Figure 4b,c,e,f are too small • There are many individual components that are confusing	*We have put a lot of time to address this remark.* *All figures have been redrawn with larger lettering. See above for the wording of the section introduction and the expanded figure caption.*

Note on EnRs and ERs. (Section 1): Most often, ER < EnR, as attack by photooxidants reduces the emitted species, decreasing with the concentration of emitted fire product j, Δy_j in the the numerator $\mathrm{EnR}_j = \Delta y_j/\Delta x$. Aerosol properties like scattering coefficient may decrease as particles coagulate, so that the ration of the volume to the scattering-determining cross section decreases. Some EnRs describe species like ozone, not present in flame, or nitric acid and PAN, not present at high levels. A textbook understanding of organic compound oxidation and the literature suggest that formaldehyde experiences thermochemical production in the flame, decay by photochemical processes, processes that overwhelm significant continued production. For use of emission factors in modeling, the circumstances of emission need to be described clearly, and the ER must be a true result of emissions. The conversion of ERs to EFs requires a separate analysis of carbon in the wood biomass, factors like area burned, the wood distribution in the area, its carbon versus water content, etc. (Andreae and Merlet, 2001, Yokelson et al. 2007). This analysis can vary from fire to fire. Such analyses can also determine the N content of the biomass burned, and this is useful in understanding the emission of nitrogen compounds.

Note on CH4 Fil-in (Section 2): In order to provide a suitably complete dataset for SEAC4RS, we used the can samples to infer likely concentrations at one-minute intervals of key species, i.e., CH_4 for all flights and CO for the first few flights, using available can samples at slightly lower frequency. The **R** package for multiple imputations by chained equations (*mice()*) was employed, using the whole data period, but filling in observations with missing data. (Our assessment of the effect of imputation was informal and is reviewed again below.) It was highly desirable to include the imputed concentrations of methane, since it is commonly measured and appears to be a prominent signal of different types of "fire chemistry", i.e., enhanced emission of reduced species; methanol and acetone are often correlated with CH_4 and give support to this idea.

The use of imputation seemed justified by three observations: (1) Checks made when both LAS and GC data were available suggested agreement. In an early period, missing tunable-laser absorption spectrometry data for both CO and CH_4, some periods did not pass this test and all observations from this period were deleted. (2) The use of regression in both *mice()* and succeeding emission ratio analyses suggested that when observations were filled in, very little *information* was added, i.e., if the technique allowed missing observations, the results would be extremely similar. Specifically, CH_4 instances filled in using other variables do not contain any additional information. (We will describe evidence in the results later, in Section 7 and Figure 4.) (3) Comparisons of 10-second and 1-minute averages for the more detailed ARCTAS dataset (not reported here) suggested that the essential variability had been captured by 60-second data. We surmised that 30-second averages might have captured more. We are unsure how averaging affects difference-based methods.

Note on Volumes (Section 3): For example, the sequence of increasingly large boxes in Figure 5 emphasizes that the relative effect of entrained non-fire air is largest near the flames, but the absolute effect of entrained air on the composition of an observed parcel is often largest close to the parcel at its point of sampling. The following section gives a framework illustrating effects of emissions and entrainment on EnRs. The box volumes of parcels at different altitudes are similar to the mole amounts shown; to be consistent with adiabatic rise of parcels, volumes should be about ~11%-14% larger in linear dimension for most plume tops)

Note on Initial Point (Section 4.1): It is natural to ask where this time/molar-expansion integration should start, naming it as $\nu = \nu_1$. A reasonable start location is *when the fire plume parcels begin entraining predominantly environmental air*, not other fresh emissions. The plume that is characterized by this expansion-period analysis is then that mixture over space and time of all detailed variations in emissions before this transition. We remark that emissions from the very hottest flaming combustion in a fire front are likely mixed with neighboring fumes from less vigorous combustion. The hottest regions seem likely never to be directly sampled. Their relevance to all downwind processing and effects is only as part of a mixture. In our dataset, the very hottest burns, MCE > 0.97, were very rarely sampled. We speculate that values of x and y_{CO}, which represent the MCE during true flaming combustion, may typically be confined to a region very close to the fire, which is measurable in the laboratory but rarely in the field.

Note on Varying Entrainment: As a side note, in simple situations (e.g., observations in a plume with same environment but with decreasing dilution, from upwind to downwind), the equation reduces to

$$(y_{\alpha j} - y_{\beta j}) = a_j\,(x_\alpha - x_\beta) \tag{13}$$

for any two observational instances, α and β, in the same plume. These are supposed chosen so that we know that x^E and all the y_j^E remain constant. If there is different dilution in the next stage, say instances β and γ, a similar relation obtains, and the composite retains linearity. Questions regarding constants of integration and original concentrations at *a* can be resolved in the regression procedure, Section 5. More generally we need $|x^{E\alpha} - x^{E\beta}| \ll |x_\alpha - x_\beta|$ and $|y_j^{E\alpha} - y_j^{E\beta}| \ll |y_{\alpha j} - y_{\beta j}|$. Situations in which the entraining concentrations vary are described later in Figure 6d. All these observations invite a more general theoretical statement, one that is necessarily more complex and is appropriate for later work.

Note on Sensitivity to Number of Tracers Used

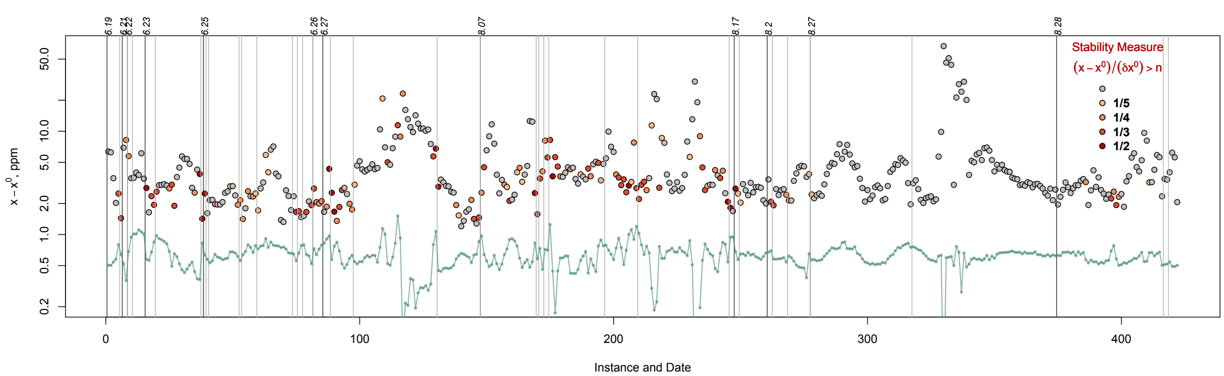

Figure S1: Estimate of variation in C_{burn} if x^0 is estimated by only three tracers (green dots:) and only five tracers (red dots). Green line repeats the pattern of $\hat{\boldsymbol{x}}_{\boldsymbol{i}}^{\boldsymbol{0}} = C_{\mathrm{bkgd}}$ shown with appropriate scale in Figure 8, for reference.

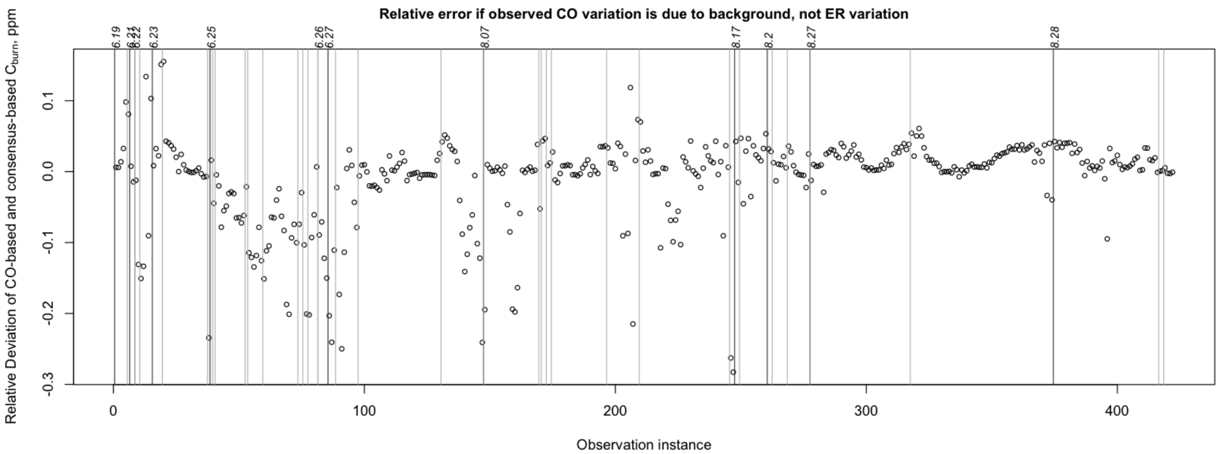

Figure S2 A heuristic measure of the stability of the situation.

The great variations in estimated $\hat{x}_i^0$ at a few points in Figure 9 can cause some concern (e.g. around sample numbered 8, 117, 130, 217m 231, 329, and 337) . Figure S2 suggests that sometimes these may be of concern, sometimes not. We constructed a measure of the temporal stability of the sampling situation by ratioing the changes of x0 to the amount of carbon burned, x-x0. The measure of change in ppm was

$$\text{Change Measure} = \left(\left|x_{i+1}^0 - x_i^0\right| + \left|x_i^0 - x_{i-1}^0\right|\right)/2$$

and to obtain a consistent measure of relative magnitude, an estimate of smoothed C_{burn} over the same span of i indices was also used,

$$\text{Magnitude Measure} = \left((x_{i-1} - x_{i-1}^0) + 2(x_i - x_i^0) + (x_{i+1} - x_{i+1}^0)\right)/4$$

This ratio of these gives our measure of the possible effect of relative change of actual C_{burn} during our the airborne measurements on the estimate of C_{burn} that the algorithm gives. Where the measurement crossed different plume boundaries (light gray lines in Figure 9), one-sided estimates consistent with these two values were calculated. We expect that use of absolute differences ratio may give a pessimistically high measure of potential influence; this was justified by a consideration of many different variations. The ratio Magnitude Measure / Change Measure can reach high values, where changes in x_i^0 can be 0.2, 0.25, 0.33, and 0.5 the amount of C_{burn}, as the colors of the points in Figure 8 show. Recall, however, that the neighboring x_i^0 estimates and C_{burn} estimates are derived independently, so the ratio does not correspond to traditional measures of high-frequency noise, only the stability and relation to identifiable processes on the ground. Figure S2 suggests that for some periods of high C_{burn}, variations of x_i^0 should matter little in calculations of C_{burn} or of the ER ratios derived from C_{burn}. In other periods, when C_{burn} is low, there can be reason for concern, even when sample-to-sample variability of x_i^0 is not especially high.

Note on Examples of Enhancement Ratios

Methods used to prepare these graphs of EnRs for the two periods of observation, Figure S3 and Figure S4, are described in Sections.7 and 9.

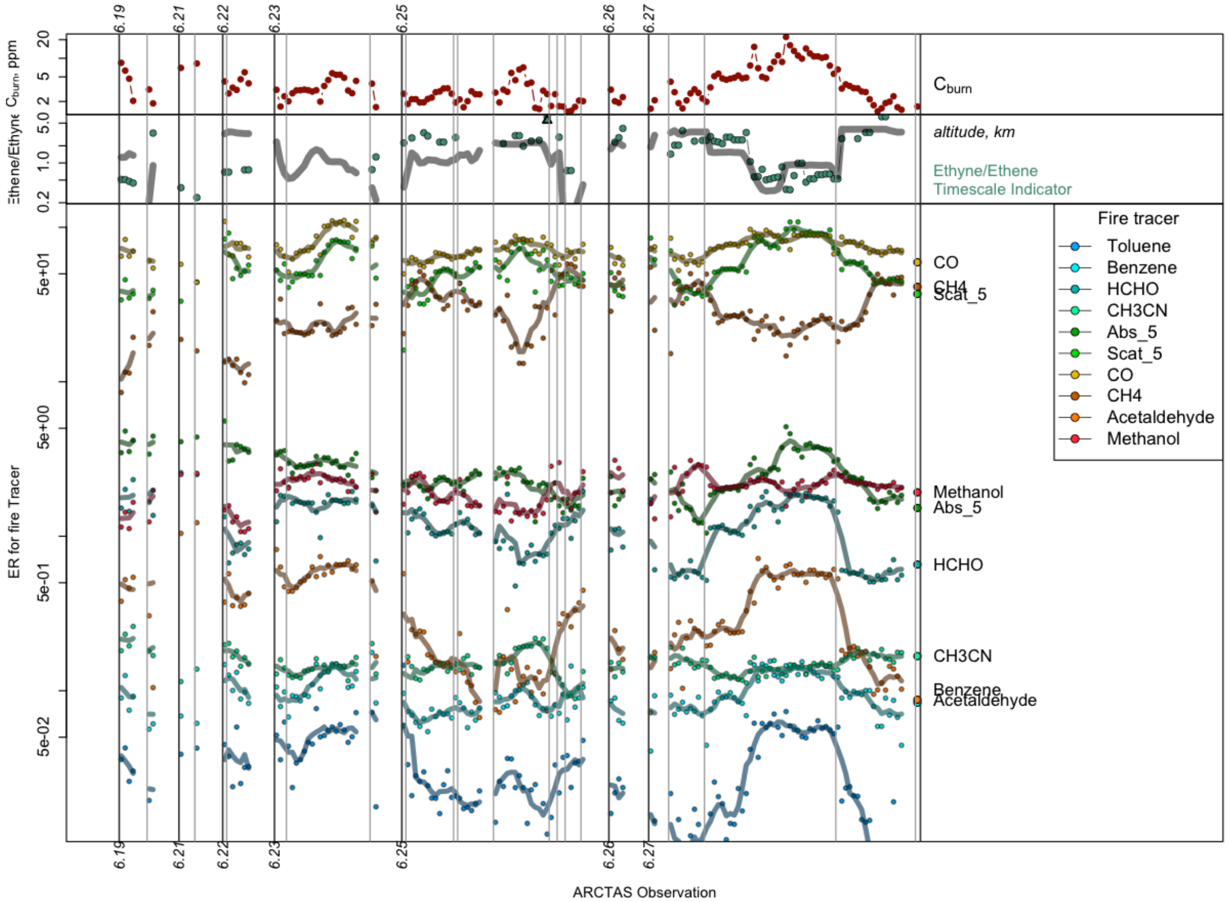

Figure S3. Estimates of C_{burn}, an ethyne/ethene photochemical transformation timescale, (Section 6.2), and enhancement ratios (EnRs) for each of the tracer compounds shown for all observations during the ARCTAS flights used in this analysis. Units for the EnRs are shown in Table 2. These diagrams were made using 10 tracers (including methane and methanol), not 8.

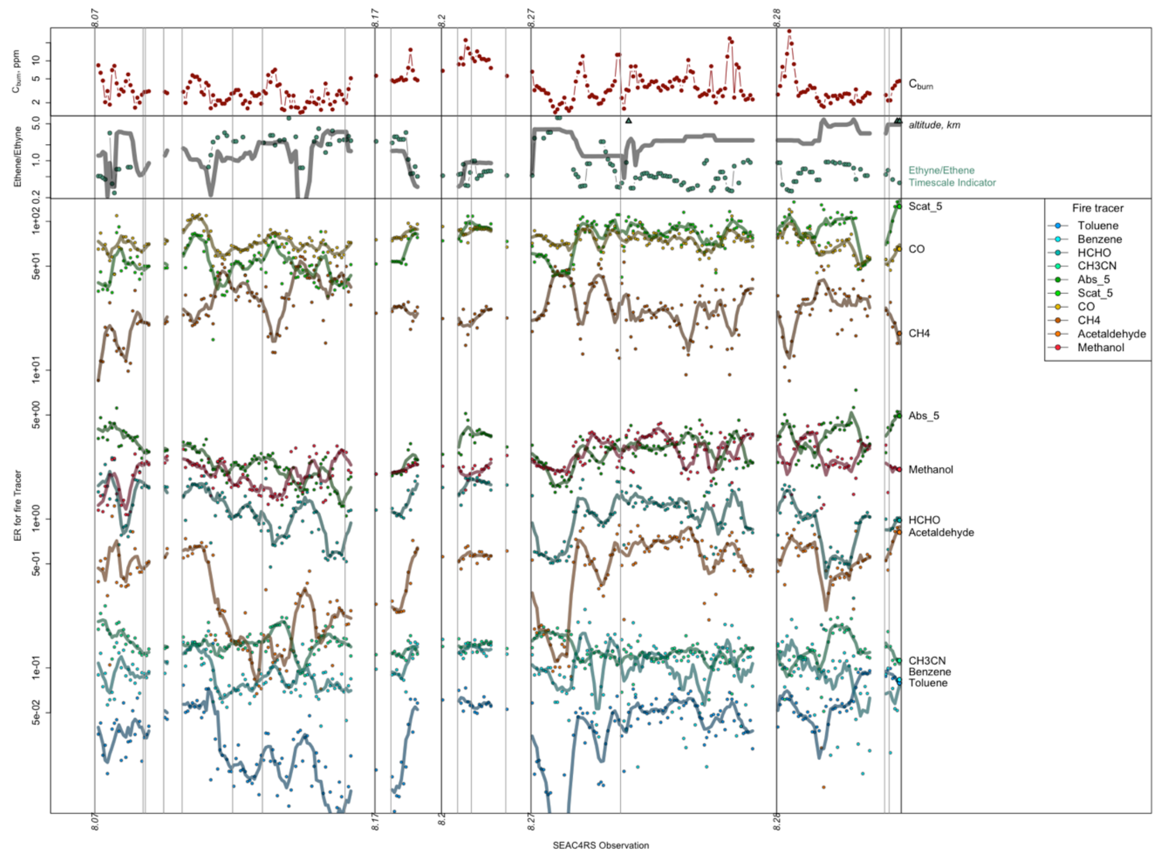

Figure S4. C_{burn} , an ethyne/ethene photochemical transformation timescale, (Section 6.2), and enhancement ratios (EnRs) for the SEAC4R observations. Units are given in Table 2.