

This manuscript is one of a set of publications, of which the second appears to be in preparation (Chatfield and Andreae, 2019). The authors take over 400 samples from the recent ARCTAS and SEAC4RS aircraft campaigns and develop a complex methodology for estimating Enhancement Ratios (EnRs) and their relationship to emission ratios (ERs) and emission factors (EFs), as well as a means for separating the background and burned carbon content in measurements of carbon emissions from biomass burning plumes. The primary work here is in the development of the Mixed Effects Regression Emission Technique (MERET), which can be used to disentangle total carbon levels in a plume from the carbon existing in background air and carbon from the biomass burning itself. The manuscript starts with a review of previous work quantifying EnRs, ERs, and EFs including a thorough description of the history of EF estimation efforts. It then moves to a description of the methodology in which data are selected and a description of their properties. Then follows two separate theory sections (one on the theory of plume expansion and one on EnR regression relationships), and then another methodology section on determining the background carbon levels and the description of the MERET method. Next, results are presented, and two examples are given. The manuscript closes with conclusions and questions for future research.

This is a very long manuscript that alternates between analysis and theory. Both parts have significant scientific value, but the manuscript itself is difficult to follow due to its length, the complexity of both the theory and analysis sections, and the way in which the manuscript is presented. 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. 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. The conversational tone of this manuscript additionally introduces confusion. For instance, L203 states “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. 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.

1. Intro
 - 1.1. Importance of Previous Work
 - 1.2. Development of EF Estimation to Date
2. Methodology: defining an indicator dataset
3. Observed behavior of C_{tot} in fire plumes – Properties of tracers
4. Theory: expanding plume for several species
 - 4.1. A general relationship
 - 4.2. Examples showing robustness of computations of idealized C_{tot}
5. Theory: A regression relationship for EnRs

6. Methodology:
 - 6.1. Finding the CO₂ + CO background
 - 6.2. Practicalities: variable EnRs
 - 6.3. Summary of the MERET method
 - 6.4. Remark: Number of independent samples
7. Results: Estimation of xi0 and C_{burn}
8. Estimates of emissions ratios: Two MERET examples
 - 8.1. MERET results for our two examples
 - 8.2. Table of several significant emissions
9. Conclusions
 - 9.1. Questions for future research
10. Team List
11. Acknowledgements
12. Author contributions

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 – 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.

Independent of these issues, the following is a review of the content of the manuscript itself.

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.

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.

SPECIFIC COMMENTS

There are many places where there are typos and undefined variables being used in the manuscript:

- L54: “Chatfield and Andreae (2017)” should be “Chatfield and Andreae (2019, in preparation)”
- L66: “ $\Delta\text{CO}_{\text{tot}}$ ” should be “ $\Delta\text{C}_{\text{tot}}$ ”.
- 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 33×10^{-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
- 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”

The phrase “affine dependence” is used several times (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 “ $a_j \leftarrow \text{CO}$ ” and “ $a_{\text{CO}} \leftarrow (\text{fire} - \text{added CO}_2 + \text{CO})$ ” terms mean. I feel a more complete explanation is needed.

The use of the variable x for C_{tot} in Section 1.2 and other places is confusing, especially when C_{tot} and x are used together (e.g., L153-158).

L522-523: The suggestion that the reader should make their own calculations in order to understand the linear responses is unhelpful.

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...”

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

- It is nearly impossible to see the arrows in 4b,c,e,f
- It is unclear without locating the matching description in the text what:
 - the color lines are on the top of Figure 4a
 - What the difference is between the blue circles, red circles, and orange dots are in Figures 4a,d
 - What the blue dashes are in Figures 4a,d
 - What the colors indicate in Figures 4b,c,e,f
 - Which of the three plotted variables are on which line in Figures 4a,d

The text and color labels in Figures 8 and 9 are similarly difficult to see and understand.