

***Interactive comment on* “Emissions Relationships in Western Forest Fire Plumes: I. Reducing the Effect of Mixing Errors on Emission Factors” by Robert B. Chatfield et al.**

Robert B. Chatfield et al.

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It is difficult to understand how this review came to be written. We the authors sympathize with the reviewer’s difficulties, as expressed. The paper has analysis, theory, statistical technique, and some resultant Enrichment Ratios. However, bearing in mind the seven months since the paper’s submission, and the 4-5 months since the first review came in, the reviewer is requested to spend perhaps 5%, maybe 1%, of the number of hours expended we expended on this very, very full response. Apparently the reviewer has not even read at the first reviewers courteous and very full and helpful review, and has not attempted wording to counter the positive aspects of that review.

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Perhaps the time allotted to get in "just some review" seemed now very short. True, we are all very distracted for the last months. Shouldn't our sheltering time now give this reviewer even more opportunity to attempt some helpful and constructive comments. Following is a text-only version of the response, taken from a table. Figures attached show the very full responses as the original table. The attachment provides this tabular view as well as revised supplementary material which address the matter of creating a direct main paper.

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 These instances and one other have been replaced by a named section of the Supplementary Material, e.g. "See also SM for a Note on Initial Point."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 CO₂+CO (blue, left axis) and the fire tracers CO and bscat (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 CO₂+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 bscat vs CO₂+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 CO₂+CO during the transect, like (b). (f) a similar diagram of bscat vs CO₂+CO for the Coastal Transect. The black bars graphed in (a) and (d) are estimates of non-fire influenced C_{bk}, see text. They and the non-plume points suggest air-mass changes in CO₂+CO. Figure 10 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 $\hat{C}_i = \text{CO}_2 + \text{CO}$ concentrations implied based on the 10 fire tracers indicated in the legend. Individual \hat{C}_{ij} are shown by overlapping colored bars (–), with the median estimate indicated by a black bar. (Upper panel) Estimates of $C_{\text{burn}} = x_{i-x} \hat{C}_i$ 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

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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 – 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. This is well-considered, but the authors find few other options. We have put much more into the Supplementary Material. 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 Fire 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

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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 CO₂ 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: “DC C_{tot} ” should be “DC C_{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" $\int \dot{V} dC_{tot} = \int \dot{V} dC_{CO_2} + \int \dot{V} dC_{CO}$ 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 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

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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 $a_j = \exp(\alpha_j C_j)$

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 $x \in E$ and all the $y_j \in E$ remain constant. Both C_{tot} and C_{burn} are used. The x axis has C_{tot} , units, while the increment beyond the vertical axis at 380, shows C_{burn} . 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

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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 “ $a_j \leftarrow CO$ ” and “ $a_{CO} \leftarrow (fire - added\ CO_2 + CO)$ ” 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). We have added an explanatory phrase: “the slope $a_j (j \leftarrow CO)$ of the regression estimates of an EnR of the species with respect to CO , multiplied by an attempted very careful estimate of the slope $a_{CO} (CO \leftarrow (fire-added\ C_{burn}))$ EnR of CO with respect to fire-produced C_{burn} . The $a_{CO} (CO \leftarrow (fire-added\ CO_2+CO))$ 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, $median(x \hat{C}_{ij}^0)$, is robust against errors resulting if a tracer j has a variable or poorly described background resulting in $x \hat{C}_{ij}^0$ 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 o The label “bscat” in Figure 4a,c is too small o 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.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2019-235, 2019.

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<p>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.</p>	<p><i>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.</i></p>
<p>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.</p>	<p><i>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.</i></p>
<p>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.</p>	<p><i>These are restated:</i> <i>L203:</i> The next section provides motivation for and understanding of an alternate approach ... <i>L312:</i> 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.</p>
<p>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.</p>	<p><i>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:</i></p> <p>Figure 1. (a) Timeline of sampling, for the period shown in Figure 3a, Montana, of CO₂+CO (blue, left axis) and the fire tracers CO and <i>b</i>_{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 CO₂+CO with</p>

Fig. 1.

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	<p>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.</p> <p><i>Figure 10 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:</i></p> <p>Figure 10. (Lower panel). Estimates of the 422 background $\hat{x}_i^0 = \text{CO}_2+\text{CO}$ concentrations implied based on the 10 fire tracers indicated in the legend. Individual \hat{x}_i^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).</p>
<p>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 –</p>	<p><i>This is well-considered, but the authors find few other options. We have put much more into the Supplementary Material.</i></p>

Fig. 2.

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<p>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.</p> <p>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.</p>	<p><i>Consequently,</i></p> <p><i>(a) Material not strictly necessary has been moved to the material.</i></p> <p><i>(b) A table of contents has been included, following the reviewer's first comment and suggestion above.</i></p> <p><i>(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</i></p> <p><i>If the editors of AMT allow, we could change the title to: Theory and Estimation of Emissions Relationships in Forest Fire Plumes: 1: Reducing Effect of Mixing Errors on Emission Factors</i></p> <p><i>(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.</i></p>
<p>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.</p>	<p><i>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!</i></p>

Fig. 3.

<p>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.</p>	<p><i>We thank the reviewer for this observation. We have rewritten the introductory paragraph:</i></p> <p>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 CO₂ 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_{bgd}, that then provides C_{burn} and thus EnRs.</p> <p><i>We have also edited several places succeeding paragraph, not described here.</i></p>
<ul style="list-style-type: none"> • 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 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. <i>provided we...</i>” is confusing 	<p>√</p> $\Delta C_{\text{tot}} = \Delta \text{CO}_2 + \Delta \text{CO}$ <p><i>Yes, a typo. Now</i></p> <p><i>Proportional to total burned material, as measured by C_{burn}</i></p> <p><i>Chose ppb/ppm rather than ppm/ppm</i></p> <p><i>Included.</i></p> <p><i>Changed. Remarks placed in the Supplement. After the equation (12) we now have: For periods of expansion in which the entrained</i></p>

Fig. 4.

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Fig. 5.

<ul style="list-style-type: none"> 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. 	<p><i>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.</i></p> <p><i>The reader is relieved of calculations now:</i></p> <p>“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.</p>
<ul style="list-style-type: none"> (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 “$a_j \leftarrow CO$” and “$a_{CO \leftarrow (fire - added CO_2 + CO)}$” 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). 	<p><i>We have added an explanatory phrase: “the slope $a_{j \leftarrow CO}$ of the regression estimates of an EnR of the species with respect to CO, multiplied by an attempted very careful estimate of the slope $a_{CO \leftarrow (fire-added C_{burn})}$ EnR of CO with respect to fire-produced C_{burn}. The $a_{CO \leftarrow (fire-added CO_2 + CO)}$ was described using the Modified Combustion Efficiency,…”</i></p>
<p>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...”</p>	<p><i>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.”</i></p>

Fig. 6.

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<p>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:</p> <ul style="list-style-type: none"> • The text and images are very small <ul style="list-style-type: none"> ○ 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 	<p><i>We have put a lot of time to address this remark.</i></p> <p><i>All figures have been redrawn with larger lettering. See above for the wording of the section introduction and the expanded figure caption.</i></p>

Fig. 7.