Interactive comment on “Greenhouse gas emissions from laboratory-scale fires in wildland fuels depend on fire spread mode and phase of combustion” by N.C. Surawski et al.

N.C. Surawski et al.

Correspondence to: Nic Surawski (Nicholas.Surawski@csiro.au)

Our responses to the first reviewer’s comments are detailed below.

Overall comment: The reviewer states that: “The sampling approach is not validated, the math is described in a misleading and inconsistent manner, and there is no practical application of the results even if the experiment had been done correctly.”

Response: The revised version of the manuscript comprehensively addresses the three areas of the manuscript requiring improvement; namely, validating the sampling approach, discussing our emission factor calculations in a more transparent fashion and demonstrating practical application of our results.

Major comment 1 on measurement approach: There is an important place for lab measurements in fire research. For instance, smoke data can be obtained with instruments that might not be field worthy. However, when working close to a fire, elucidation of the impact of fire behavior on emissions is only valid if it can be shown that the sampling is representative of the overall lab fire emissions for all the behavior types considered. In other words, it needs to be shown that the smoke is well mixed so that data acquired at the sampling point do not reflect a fire-behavior impact on the height at which emissions from different processes are released. As an example, Christian et al., (2004) show that temperature and mixing ratios are constant across the stack at the level where sampling occurs for their lab fires. (Prior to that test, they published results based on an optical path that spanned the whole stack.) The good mixing Christian et al confirmed was due largely to a torus surrounding the base of the stack that promotes turbulent mixing. In contrast, wind tunnels are designed to eliminate turbulence, which discourages good mixing. In fact, Christian et al considered wind tunnel measurements, but found that wind tunnel fires produced a strong vertical temperature gradient with hot gases (flaming emissions) mostly at the top of the wind tunnel and cooler...
gases (smoldering emissions) lower. Thus, the CO/CO₂ ratio depends strongly on the point-
sampling height selected. This separation of process-specific emissions likely varies strongly
by fire spread mode. In other words, the author’s CO/CO₂ data could be reproducible, but not
be representative of fire behavior effects if the emissions are not well mixed and flaming
emissions have greater tendency to rise above their one fixed sampling point for some spread
modes. Without evidence that this artifact does not occur the data are not of value.

Response: The same comment was made by the second reviewer as well. We have added a
new section to the discussion section of this article called “Representativeness of combustion
wind tunnel emissions measurements” (section 4.1 in revised version of manuscript) and a
new Table of supporting data (Table 3 in revised manuscript) which together provide further
analysis supporting our measurement approach. In this new section, we calculate the reaction
Damköhler number (\(Da\)) which is the ratio of the flow time scale to the chemical reaction
time scale (Law, 2006). We calculate \(Da\) at two flame heights and axial positions within the
flow with \(Da\) exceeding \(10^6\) in all cases. Therefore, for the species we measure in this
experimental effort, the timescale required for chemical reaction is very short relative to the
flow timescale in our combustion wind tunnel. Therefore, the chemical reactions are at
equilibrium (or are “frozen”) by the time our sampling manifold is reached and furthermore
do not depend on sampling height.

The new section in the discussion (section 4.1) reads: “Since emissions sampling was
conducted at a single fixed height above the wind tunnel floor (see section 2.1), further
analysis needs to be conducted to ensure the representativeness of measurements. If chemical
reactions were still occurring at the axial position of sampling, and if those reactions had a
dependence on sampling height, then the emissions measurements obtained would not be
representative of the entire plume. Here we calculate the reaction Damköhler number (\(Da\))
(Law, 2006, p. 189) which characterises the ratio of the flow time scale (\(\tau_F\)) to the chemical
reaction time scale (\(\tau_C\)). The reaction Damköhler number is given by:

\[
Da = \frac{\text{Characteristic flow time}}{\text{Characteristic reaction time}}
\]  

(11)
= \frac{\tau_F}{\tau_C} \\
= \frac{\kappa L}{U'}

where \( \tau_F \) is given by the characteristic length scale \((L)\) divided by the characteristic velocity \((\overline{U})\) (Law, 2006) and \( \tau_C \) is the reciprocal of the reaction rate \((k)\). We choose \( L \) as the axial distance from the flame position to the sampling manifold (either 3.6 or 8.4 m), \( \overline{U} \) as the mean wind speed employed during testing \((1.5 \text{ m s}^{-1})\) with \( k \) given by the lumped kinetic scheme of Ranzi et al. (2008), which describes the production of \( \text{CO}_2, \text{CH}_4 \) and \( \text{CO} \) (plus other carbon compounds) from biomass pyrolysis. We calculate \( k \) at two heights within the flame, with maximum temperatures at the flame base being based on those recorded by thermocouples on the CSIRO Pyrotron floor, whilst flame tip temperatures are based on measurements made in eucalypt shrubs by Wotton et al. (2012). Calculation of the reaction Damköhler number enables us to assess how close the relevant chemical reactions are to equilibrium at two flame heights and axial positions within the flow, with the results of this calculation being shown in Table 3.

We see that the reaction Damköhler number depends on vertical position within the flame, with smaller \( Da \) being observed at the flame tip \((\text{i.e. } 3.0 \times 10^6)\) compared to the flame base \((1.8 \times 10^8--2.9 \times 10^8)\). There is also variation in the \( Da \) observed with different fire spread modes which is due to differences in the maximum flame base temperature and the influence it has on reaction kinetics. Whilst we see variation in \( Da \) with respect to fire spread mode and vertical position within the flame, all of the \( Da \) exceed \( 10^6 \) \((\text{rounded to the nearest order of magnitude})\) which does not change the conclusion that the reactions are
near equilibrium or "frozen" (Jenkins et al., 1993). Hence, we can conclude from this analysis that our emissions sampling is representative of the entire plume since the timescale required for the relevant chemical reactions to occur is very short relative to the flow timescale.

Table 3 in the revised manuscript reads:

**Table 3. Calculation of the reaction Damkohler number \((D_a)\) for several axial positions and flame heights within the flame.**

<table>
<thead>
<tr>
<th>Fire spread mode</th>
<th>(T_{\text{flame tip}} ) (K)</th>
<th>(T_{\text{flame base}} ) (K)</th>
<th>(\tau_F ) (s)</th>
<th>(\tau_{C_{\text{flame tip}}} )</th>
<th>(\tau_{C_{\text{flame base}}} )</th>
<th>(D_{a \text{flame tip}} )</th>
<th>(D_{a \text{flame base}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heading</td>
<td>540</td>
<td>1170</td>
<td>5.6</td>
<td>8.0 \times 10^{-7}</td>
<td>2.2 \times 10^{-6}</td>
<td>7.0 \times 10^{6}</td>
<td>2.6 \times 10^{6}</td>
</tr>
<tr>
<td>Heading</td>
<td>540</td>
<td>1170</td>
<td>4.4</td>
<td>8.0 \times 10^{-7}</td>
<td>2.2 \times 10^{-6}</td>
<td>3.0 \times 10^{6}</td>
<td>1.1 \times 10^{6}</td>
</tr>
<tr>
<td>Flanking</td>
<td>540</td>
<td>1050</td>
<td>6.6</td>
<td>8.0 \times 10^{-7}</td>
<td>3.1 \times 10^{-6}</td>
<td>7.0 \times 10^{6}</td>
<td>1.8 \times 10^{6}</td>
</tr>
<tr>
<td>Flanking</td>
<td>540</td>
<td>1050</td>
<td>2.4</td>
<td>8.0 \times 10^{-7}</td>
<td>0.1 \times 10^{-6}</td>
<td>3.0 \times 10^{6}</td>
<td>7.7 \times 10^{7}</td>
</tr>
<tr>
<td>Backing</td>
<td>540</td>
<td>1220</td>
<td>5.6</td>
<td>8.0 \times 10^{-7}</td>
<td>1.9 \times 10^{-6}</td>
<td>7.0 \times 10^{6}</td>
<td>2.9 \times 10^{6}</td>
</tr>
<tr>
<td>Backing</td>
<td>540</td>
<td>1220</td>
<td>2.4</td>
<td>8.0 \times 10^{-7}</td>
<td>1.9 \times 10^{-6}</td>
<td>3.0 \times 10^{6}</td>
<td>1.3 \times 10^{6}</td>
</tr>
</tbody>
</table>

**Major comments 2a-d on EF’s.**

**Comment 2a:** The reviewer states that: “Emission factors (EF) are meant to be used with fuel consumption data and fuel consumption data explicitly doesn’t count unburned carbon that remains on the site”.

**Response 2a:** One factor that the reviewer has neglected to consider in their comment is that burnt fuel carbon does not necessarily have to be emitted to the atmosphere, even though most of it is. As we detail in later in this set of responses (i.e. major comment 2), burnt carbon could be present in the post-fire combustion residues as black carbon, ash or partially charred/combusted fuel.

Based on our literature research we conducted, Andreae and Merlet (2001) suggest that best practices in fire research **should** consider burnt carbon present in the post-fire residue. For example, Andreae and Merlet suggest: “Calculation of this parameter (i.e. emissions factors) requires knowledge of the carbon content of the biomass burned **and the carbon budget of the fire**; both parameters are difficult to establish in the field as opposed to laboratory experiments where they are readily determined.”
It is generally common practice in atmospheric chemistry research to only consider carbon emitted to the atmosphere and to neglect carbon remaining in the post-fire combustion residue that has been burnt. Since this was a laboratory based study we considered the complete carbon budget of the fire, which as Andreae and Merlet suggest is simpler to do in a laboratory, rather than field, setting. Considering that we have taken this additional factor into account does not indicate that we have done anything it wrong, it merely suggests that we have considered the complete carbon budget of the fire as recommended by Andreae and Merlet.

Comment 2b: Following on from this point, the reviewer then suggests: “The authors are confused about this and make misleading statements about emission factors in other work. Further, they express EF both in the normal g/kg and as unspecified percentages.” Related to this point, the reviewer then states: “Further, they express EF both in the normal g/kg and as unspecified percentages.”

Response 2b: This comment was also made by the second reviewer (please see major comments 2a-b on EF’s). In this article we have reported emissions factors two ways; namely: 1) as a percentage of the burnt carbon or nitrogen, or 2) on a per unit dry fuel consumed basis. We have modified the sentence on page 23133 (line 17) to make it clear that when we report emission factors as a percentage, it is a percentage of the total carbon or nitrogen burnt and not some “unspecified percentage” as claimed by both reviewers. Furthermore, we have furnished this revised sentence with several references to indicate that reporting emission factors this way has occurred widely in the emissions literature since the method was developed by Radke et al. in 1988. This revised sentence now reads: “A carbon mass balance approach developed by Radke et al. (1988), and applied (for example) by Lobert et al. (1990), Hurst et al. (1994a), Hurst et al. (1994b), and more recently by Meyer et al. (2012), was used to calculate emissions factors for different carbon- and nitrogen-based pollutants on a per unit element burnt basis.”

Comment 2c: The reviewer then states: “The authors are correct that some burned C is converted to charcoal and this is a source of a small error in some standard carbon balance approaches. However charcoal yields are generally small and should not be confused with remnants of unburned carbon. For instance, Kuhlbusch et al. (1996) noted: “The ratio of black carbon produced to the carbon exposed to the fire in this field study (0.6–1.5%) was somewhat lower than in experimental fires under laboratory conditions (1.0–1.8%) which
may be due to less complete combustion.” Some of their black carbon was in the emitted particles and some in the ash, with the ash portion representing the error in the carbon mass balance method due to C in the residue. When charcoal yields are high, as in the case of purposeful charcoal production, a method to adjust the CMB for this has already been published (Bertschi et al., 2003).”

Response 2c: Kuhlbusch report black carbon production percentages of 1.0-1.8% (relative to total carbon exposed) based on laboratory testing; however, based on preliminary $^{13}$C NMR results conducted by the authors (which we reserve for presentation in a future publication) we think this percentage varies from 3% for heading fires to 7.5% for backing fires. This percentage is calculated by ascribing aryl structures from the NMR spectrum as being aromatic in nature and relatively resistant to degradation. Recent field work conducted by Volkova et al. (2014) on carbon emissions from prescribed burning and wildfire has noted increased charring of combustion residues from lower intensity fires (such as those conducted in this study) compared to wildfire. Thus, there is empirical evidence to support our results suggesting greater black carbon production relative to Kuhlbusch et al. More importantly though, Kuhlbusch et al. report on a number of carbon possibilities post-fire with black carbon representing only a portion of the carbon forms present. There will also be partially charred/combusted material, ash and also some unburnt (but nonetheless thermally exposed and altered) leaf, bark and twig remnants. Thus, the reviewer is incorrect in suggesting that post-fire carbon is composed merely of black carbon and an unburnt carbon pool.

Comment 2d: The reviewer also makes the suggestion that our combustion factors are small and that only non-carbon containing elements can be significant in the post-fire combustion residue.

Response 2d: Kuhlbusch et al. (1996) report backing fire carbon volatilisation percentages of 72% for the FP 4/2 fire and 78.2% for the FP 4/1 fire which is in excellent agreement with ours (74.8%). For heading fires Kuhlbusch et al. report carbon volatilisation percentages between 85.4% (KPE/1 fire) and 95.5% (KP3/3) which is, once again, very similar to ours (88.3%). Therefore, our carbon volatilisation percentages are in agreement with the Kuhlbusch et al study. It should also be noted that combustion factors near 100% could occur in extreme wildfire situations; however, in our experimental fires (please see Table 1) the Byram fireline intensity is more indicative of a prescribed fire situation. As a result, we would expect combustion factors less than 100% as indicated by our results. Furthermore,
our results and those of Kuhlbusch et al suggest that it is possible to get 30% of total fuel carbon deposited in the post-fire combustion residue despite claims being made to the contrary by the reviewer.

**Major comment 3 on application of results:** A serious problem is that real fires present a mix of fire spread modes (as the authors themselves state) and in any case there is no way to operationally monitor fire spread modes for all the fires of importance, especially since the majority of global biomass burning goes undetected from space (Yokelson et al., 2011). Even if single spread modes were applicable to real fires, and they could be routine measured, many other factors effect emissions interactively such as fuel geometry, moisture, RH, etc.; and wind effects on the ability of a fire to propagate are probably far more important than subtle emissions differences. I.e. wind has other impacts such as aiding fire spread in dispersed fuel, making fire control more difficult, and possibly enabling ignition of live fuels that might not burn otherwise. Wind interacts with fire induced convection in complex ways. None of variables can be operationally monitored in complex fire environment and realistic replication of some complex fuel beds including live, moist, or large fuels etc. is probably not feasible. If the numerous variables could be controlled one at time there are likely still non-linear interactions between driving variables.

**Response:** We agree with the reviewer that most global fires cannot be managed; however, in the section of the manuscript where we apply our results (section 4.5 of the revised manuscript) we are considering prescribed fire where there is explicit choice (i.e. selected before the burn) regarding the range of variables that the reviewer discusses in their comment; such as: wind speed, fuel moisture as well as the ignition pattern. This is operationally achieved by carefully selecting the ignition timing to correspond with fire weather conditions that are appropriate for achieving the objectives of the burn. Furthermore, the ignition pattern selected is based on a judicious choice regarding the moisture, load and contiguity of fuels, the prevailing wind speed and direction as well as topography and the presence of firebreaks. In our current article, we argue that mitigation of greenhouse gas emissions could become part of the overall prescribed burn design; of which we assess the potential of by applying single fire spread modes over a landscape. Whilst we agree that a single or universal fire spread mode cannot be achieved in a prescribed fire situation; in practice, a variety of ignition patterns are commonly employed in such operations that enable the fire spread modes we considered (i.e. heading, flanking and backing) to predominate in different fuel, weather and
topographical conditions. We add a sentence to the 1st paragraph in section 4.5 (page 21 of revised manuscript) stating that ignition patterns exist which enable a single fire spread mode to predominate. This new sentence reads: “Whilst it would not be possible to apply a single fire spread mode to a forested landscape in a prescribed fire situation, ignition patterns are practised in Victoria which enable a single fire spread mode to predominate (Tolhurst and Cheney, 1999), such as the three investigated in this study.

Other miscellaneous comments: Real fires burn with a mix of smoldering and flaming that is further not operationally available. Both main hypotheses are already in literature. Keene et al showed fire spread mode impacts MCE and countless papers have already shown that CH₄ correlates with MCE.

Response: The unique aspect of our study was outlined on page 23128 where we state “In this study, we re-examine the burning methodology of Keene et al. in a controlled laboratory study with an explicit experimental design combined with statistical testing of results. As such, examining the hypothesis that greenhouse gas emissions could depend on fire spread mode is the major focus of this article.” The only greenhouse gas species measured in the study of Keene et al. was CO₂ (which we stated on page 23128 line 14 of the original manuscript) which motivated us to revisit their burning methodology (i.e. heading, flanking and backing) to assess its impact on other greenhouse gas species. This is the basis of our original contribution in this article; not the other foci suggested by the reviewer.

Comments below were put in a Page, Line format by the reviewer.

Comment 1: 2, 4: diameter?

Response: Change made.

Comment 2: 2, 14: twice as much CO as what?

Response: We’ve added to some more detail to the end of this sentence to make it clear that heading fires produced twice as CO as flanking and backing fires.

Comment 3: 4, 1: Actually there are an infinite number of possible angles, they are normally mixed, plus any real fire has multiple wind directions.

Response: We emphasise that we are referring to the three “mutually independent” fire spread modes in this article, whereas the reviewer is referring to an infinity of directions.
obtained by linear combinations of the fires spread modes we considered. To avoid confusion though, we have changed the word “different” in this sentence to “main” to account for the possibility of having many fire spread modes.

Comment 4: 4, 20-24: There is no way to operationally monitor fire spread modes and in fact the majority of global fires go completely un-detected, plus no single fire spread mode applies to a whole fire.

Response: For wildfire this may be true, but this is not the case for a prescribed fire where monitoring the fire spread mode is an explicit consideration in the conduct of such a burn (Tolhurst and Cheney, 1999). (please see our response to major comment 3 on representativeness of experiments).

Comment 5: 6, 1: all gas sampling at one height – no evidence well mixed for all fire types

Response: We have added a new section to the manuscript (section 4.1 in the revised version of the manuscript) which addresses this comment. Please see our response to reviewer 1’s major comment on our sampling design (major comment 1).

Comment 6: 7, 15: windspeed of 1.5 m/s or ~5 km/h kind of low

Response: This wind speed is one that is relevant for prescribed fire.

Comment 7: Pages 9-11: un-needed lengthy discussion of old math, plus a misprint in eqn 7

Response: These points were also raised by reviewer 2. Considering that both reviewers questioned the reporting of emission factors on a per unit element burnt basis (as a percentage), we thought it would be good practice to methodically work through our methods of calculation including relevant references to make our calculations transparent to the readers of this paper. The typographical error in equation (7) has been corrected.

Comment 8: 14, 21-24: “Fire spread mode had a statistically significant effect on CO₂ (p<0.0001), CO (p<0.0001) and carbon residue emissions (p<0.0001) but did not have a statistically significant effect on CH₄ (p = 0.269) or N₂O emissions (p = 0.261).” Something went wrong here because fire spread mode effects MCE and CH₄ is strongly correlated with MCE and the authors claim N₂O is strongly correlated with CH₄.

Response: The reviewer is incorrect in suggesting that something has “gone wrong” with our MCE versus CH₄ results. Below we have included a plot of CH₄ emissions factors which shows a statistically significant relationship with MCE (p < 0.0001, R²=0.68). The source of
Confusion for the reviewer is, perhaps, that we don’t get a statistically significant relationship between CH₄ emission factors and fire spread mode due to the observed variability in our data set.

Also, the reviewer is mistaken in stating that we claimed a correlation between CH₄ and N₂O emissions. Whilst on page 23139 lines 3-18 we say (in a general way) that CH₄ and N₂O emissions are increased during smouldering combustion, but we do not claim that they are correlated.

Comment 9: 14, 23: “carbon residue emissions”?

Response: Changed to carbon residue production.

Comment 10: 15, 17-18: On the same page the authors first claim that CH₄ increases during smoldering and N₂O doesn’t, then a few lines below they make opposite claim.

Response: The source of confusion for the reviewer here is that lines 3-18 (page 14) discuss the results numerically, whereas formal testing of results for statistical significance occurs on lines 19-27. What appears like a trend numerically may not pass the test as being statistically significant. To alleviate this confusion we have added two sentences on page 14.

We add the first sentence in line 8 stating: “In this paragraph we discuss the numerical trends found, whilst the next paragraph discusses testing of our results for statistical significance.”

The second sentence is added in line 25 stating: “Whilst the non-significant result for CH₄ may appear to contradict the trends discussed in the previous paragraph, the CH₄ results are more variable which prevents a statistically significant result from being found.”

Comment 11: Page 17: In general: The EF has to be multiplied by fuel consumption to get emissions!

Response: We are aware of that, but for equivalence of the two methods in reporting total emissions we need to multiply emissions estimates (obtained from an emissions factor
reported per unit dry fuel consumed) by $\sum \frac{C_{\text{emit}}}{C_{\text{fuel}}}$ at some point as we correctly suggested on page 17.

Comment 12: 17, 11-12: Wrong, the widely used CMB approach assumes that burned fuel carbon (except for charcoal) is emitted to the atmosphere.

Response: We disagree with the reviewer. This is merely the assumption we want to relax by considering the fraction of burnt carbon that is emitted to the atmosphere by explicitly multiplying by $\sum \frac{C_{\text{emit}}}{C_{\text{fuel}}}$. For example, in one of the references provided by the reviewer (Bertschi et al., 2003) they state in paragraph [21] (2nd sentence): “we assume that all the burned carbon is volatilized ...”. As discussed in our response to the major comment on EF’s by the 1st reviewer (please see major comments 2a-d) burnt carbon is not completely emitted to the atmosphere. Once again, to be in a position to estimate one needs to consider the complete carbon budget of the fire (i.e. emitted to the atmosphere and burnt and remaining in the post-fire residue) as suggested by Andreae and Merlet (2001). In addition, charcoal (although a small fraction of total fire exposed carbon but larger in our results) is not the only carbon form present in the post-fire combustion residue not emitted to the atmosphere. In our results, we show that a significant fraction (between 10-30%) of total (fire exposed) carbon is left in the post-fire combustion residue in a variety of forms including: black carbon, ash, partially charred/combusted material and some thermally exposed/alkaline fuel.

Comment 13: 17, 17: If fuel carbon remains on site and is not counted as fuel consumption then the authors approach will incorrectly estimate carbon emissions.

Response: We disagree with the reviewer. To be counted as fuel consumption the fuel has to be burnt and emitted to the atmosphere. Burnt carbon remaining on site does not get counted as consumed because it is not emitted to the atmosphere.

Furthermore, it is apparent to the authors of this paper (based on comments 12 and 13) that there is some confusion regarding the difference between the terms burnt and consumed. We
have added two sentences in the first paragraph of section 4.2 (page 17 lines 20-24 in the revised manuscript) stating: “For our purposes, we define ‘burnt’ as fuel that has been thermally altered as a result of exposure to fire and either emitted to the atmosphere or left in the post-fire residue. We define ‘consumed’ as that component of the fuel that is emitted to the atmosphere as a result of exposure to fire.

Comment 14: 18, 6: How can EF be expressed as a percent?
Response: This comment was also made by the second reviewer. We described this in section 2.4.1 of the original manuscript (Calculation of emissions factors). As stated earlier under the major comment on EFs (please see major comments 2a-d) we added a sentence showing that reporting emission factors per unit of element burnt has been done widely since the 1980’s in the atmospheric chemistry literature.

Comment 15: 18, 12-14: Here the authors explain perfectly why their work has no realistic application, real fires, they state, have mixed spread modes.
Response: We addressed this earlier under the major comment on Application of our results (please see major comment 3). In prescribed fire situations in Australia, the ignition location (and hence the fire spread mode) is a controllable parameter (Tolhurst and Cheney, 1999).

Comment 16: Table 1: Does not label the fire spread modes?
Response: We added fire spread mode and fuel moisture content as two extra columns to this table in the revised manuscript.

Comment 17: Table 2: The footnote discusses comparisons that are not in the table
Response: We have removed the footnote to Table 2 as its contents appear elsewhere in the manuscript.

References


Tolhurst, K. G. and Cheney, N. P.: Synopsis of the knowledge used in prescribed burning in Victoria, Department of Natural Resources and Environment, East Melbourne, Victoria, 1999.
