# 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.**

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Our responses to the second reviewer's comments are detailed below.

**Major comment 1 on sampling methodology:** The reviewer states: "the experimental setup of plume sampling (only at one point) is probably not representative of the average emission composition as result of in-homogeneity of the plume in tunnel effluent as result low turbulence and temperature gradient."

**Response:** 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:

$$Da = \frac{\text{Characteristic flow time}}{\text{Characteristic reaction time}} = \frac{\tau_F}{\tau_C} = \frac{kL}{\bar{U}}, \quad (11)$$

where $\tau_F$ is given by the characteristic length scale ($L$) divided by the characteristic velocity ($\bar{U}$) (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), $\bar{U}$ as the mean wind speed employed during testing (1.5 m $s^{-1}$) with $k$ given by the lumped kinetic scheme of Ranzi et al. (2008), which describes the production of $CO_2$, $CH_4$ and 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 (i.e. 3.0 x $10^6$) compared to the flame base (1.8 x $10^8$–2.9 x $10^8$). 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 $10^6$ (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 Damköhler number $(Da)$ for several axial positions and flame heights within the flame.

| Fire spread mode | $T_{\text{flame tip}}$ (K) | $T_{\text{flame base}}$ (K) | $\tau_F$ (s) | $\tau_{C_{\text{flame tip}}}$ | $\tau_{C_{\text{flame base}}}$ | $Da_{\text{flame tip}}$ | $Da_{\text{flame base}}$ |
|---|---|---|---|---|---|---|---|
| Heading | 540 | 1170 | 5.6 | $8.0\times10^{-7}$ | $2.2\times10^{-8}$ | $7.0\times10^{6}$ | $2.6\times10^{8}$ |
| Heading | 540 | 1170 | 2.4 | $8.0\times10^{-7}$ | $2.2\times10^{-8}$ | $3.0\times10^{6}$ | $1.1\times10^{8}$ |
| Flanking | 540 | 1050 | 5.6 | $8.0\times10^{-7}$ | $3.1\times10^{-8}$ | $7.0\times10^{6}$ | $1.8\times10^{8}$ |
| Flanking | 540 | 1050 | 2.4 | $8.0\times10^{-7}$ | $3.1\times10^{-8}$ | $3.0\times10^{6}$ | $7.7\times10^{7}$ |
| Backing | 540 | 1220 | 5.6 | $8.0\times10^{-7}$ | $1.9\times10^{-8}$ | $7.0\times10^{6}$ | $2.9\times10^{8}$ |
| Backing | 540 | 1220 | 2.4 | $8.0\times10^{-7}$ | $1.9\times10^{-8}$ | $3.0\times10^{6}$ | $1.3\times10^{8}$ |

**Major comments 2a-b on EF's.**

**Comment 2a:** The reviewer states that "the data treatment and presented formulation is given in a very confusing way with a unnecessary long discussion of equations for Emission Ratios and Emission Factors that in several cases are inaccurate, using unclear symbology."

**Response 2a:** As discussed in detail in the next paragraph, given that we report emission factors in two equally valid ways (either as a percentage of the burnt carbon or nitrogen and on a mass per unit of dry fuel consumed basis) we thought it would be good scientific practice to clearly describe and cite the calculation methods used. We thought this would add transparency to our analysis, but regrettably, the reviewers have correctly identified one typographical error in both equations (3) and (7) which we have now corrected. With these two errors rectified we believe that sufficient detail (and no more) has been provided for readers to understand our methods. As for unclear symbology, we have used the nomenclature present in the papers we have cited which involve terms commonly used in

wildfire emissions science. As such equations (2-4) are based on Hurst et al. (1994b), (5-6) on Yokelson et al. (1999), whilst (7) is based on Andreae and Merlet (2001).

**Comment 2b:** The reviewer then states: "Emission Factors are given as a fraction of burned/fired carbon, as a fraction (g/Kg) of burned biomass and in Section 4.2 as an un-specified percentage of something."

**Response 2b:** 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 (please see also major comments 2a-d by reviewer 1). 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."

**Major comment 3 on representativeness of experiments:** My doubts are that these laboratory experiments with quite uniform and low litter sizes and humidity conditions, can be representative of prescribed fires that are done in less extreme dried conditions with winds that produce a mixture of processes (heading/flank/back).

**Response:** We selected the fuel load, fuel moisture content and wind speed to obtain Byram fireline intensities which are representative of that which occurs during many prescribed burning operations. We have now added a sentence at the end of section 2.2 (last sentence) explaining why these various parameters were chosen. This new sentence reads: "Altogether, the selection of fuel loads, fuel moisture content and wind speed were selected to achieve Byram fireline intensities (Byram, 1959) (which is the product of the lower heating value of the fuel, fuel consumed and the forward rate of spread) indicative of those during prescribed burning conditions in temperate eucalypt forest in Australia (i.e. approximately < 500 kW $m^{-1}$ (Cheney, 1981) or approximately < 345 kW $m^{-1}$ (McArthur, 1962))"

The comment the reviewer makes about having a mix of fire spread modes was also raised by the first reviewer (please see major comment 3 by reviewer 1 on application of results). 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 21of 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.

**Comment 1:** Line 25, page 23129- develop experiments positioning the tube at different heights above the floor of combustion to access the homogeneity of the plume.

**Response:** The new section we added to the revised manuscript (section 4.1: Representativeness of combustion wind tunnel emissions measurements) has addressed this comment.

**Comment 2:** Line 13, page 23130- removal of fragmented material will not produce a combustible less representative of natural conditions?

**Response:** The comment made by the reviewer is correct but adding a duff layer to the fuel bed would have added an extra level of complexity that we did not want in our first set of emissions experiments. It was beyond the scope of the current set of experiments to include another fuel stratum in our experiments.

**Comment 3:** Line 1-2, page 23131- To dry the combustible to this low humidity is representative of conditions of burning in prescribed fires? Usually prescribed fires are taken during periods of lower fire hazard, therefore more humid.

**Response:** Prescribed burns in Victoria are usually conducted between 9-16% (Tolhurst and Cheney, 1999) but in the current work we dried the fuel to give Byram fireline intensities indicative of those at the higher-end of prescribed fire. We have added a sentence at the end of section 2.2 to explain why the fuel moisture was dried to such a low level (please see major comment 3 by reviewer 2 on the representativeness of results).

**Comment 4:** What means dilution with zero air? Is it normal external air, with usual $CO_2$ content, or air without $CO_2$? Clarify. If it is air with normal ambient $CO_2$ (and $CH_4$, etc) which is the imprecision resulting from the subtraction for conditions when burning is producing less emissions (in the end of experiments)?

**Response:** We have used air consisting of 20.5% $O_2$ in $N_2$. Hence there are no additional sources of carbon that need to be accounted for when correcting for the dilution ratio. We have modified line 7 on page 23132 to add this compositional information on what we mean by zero air.

**Comment 5:** Lines 13-15, page 23132- Unclear

**Response:** All we are saying is that the initial dilution ratio applied was increased during the heading fire experiments, but this did not happen for backing and flanking fires. We thought it was written clearly so have not modified this sentence.

**Comment 6:** Pages 23132-23133- I think that this discussion about ER is probably not necessary. It is only a methodology to calculate emission factors from concentration measurements. The associated figure 4 is also not very enlightening. Is it for heading, flanking or backfires?

**Response:** We have decided to keep the discussion on emission ratios as a choice needs to be made about which reference gas to use for calculating emission factors. The associated figure (i.e. Figure 4) is enlightening as it indicates that $CO_2$, CO and $CH_4$ would all be good choices as a reference gas for calculating emissions factors based on the quality of the linear fits. The caption for Figure 4 has been modified to make it clearer that the results from all 18 experimental fires appear in each panel.

The new caption for Figure 4 reads (with a modified first sentence in the caption): "Linear fits of excess mixing ratios for all 18 experimental fires (not corrected for the overall dilution ratio) using either $CO_2$, CO or $CH_4$ as a reference gas.

**Comment 7:** Equation 2- This equation is not exact. With basis in in concentration molar ratios (ppm) the values for NMHC should take into account that all hydrocarbons have more than a C atom. Also molar ratio for PC is not well defined.

**Response:** The fact that non-methane hydrocarbons have more than one carbon atom is addressed explicitly by the parameter *n* in the next equation (i.e. equation 3). Whilst the emissions factor for particulate carbon is not well defined it nonetheless contributes to the carbon being emitted to the atmosphere and hence should be in the equation.

**Comment 8:** Equation 3- lacks a delta before $CO_2$

**Response:** Change made and thanks for spotting this typographical error.

**Comment 9:** Lines 16-18, page 23134. To adapt equation 3 to $N_2O$ it needs also to substitute in for the ratio between $N_2O$ and $CO_2$ number of atoms in the molecule (that is- 2). The consequent emission factor is in fraction of N emission per N present in the combustible burned? Clarify.

**Response:** There is no need to do this (as described in Hurst et al. (1994b) and Meyer et al. (2012)) as the molar nitrogen-to-carbon ratio (which we divide equation 3 by) takes into account the fact that $N_2O$ has two nitrogen atoms.

To alleviate this potential confusion for readers, the sentence on page 10 of the revised manuscript (lines 15-17) has been modified to read: "To estimate emissions factors for $N_2O$, the excess mixing ratio for $N_2O$ is substituted into the numerator of equation (3) and is then divided by the molar nitrogen-to-carbon ratio of the fuel to account for the fact that every mole of $N_2O$ has two moles of N.

**Comment 10:** Equation 5- to use the same symbol EF for this and equation 3 is confusing. Fc needs to be in fraction in the equation and not in % as it is suggested. In the equation there is confusion between molecules and atoms of carbon.

**Response:** We've made many changes to the manuscript (please see major comment 2 by both reviewers and comments 16 and 17 by reviewer 2) articulating how we've used two methods of reporting emissions factors in this paper. The context of which of the two approaches and where it's used is now clear in the revised manuscript.

We've changed the part of line 4 page 23135 which refers to $F_C$ to read "$F_C$ is the fractional fuel carbon content (measured before burning: 0.516)." Given our nomenclature follows that

of Yokelson et al. (1999) we don't see any confusion between molecules and atoms of carbon in equation (5) as suggested by the reviewer.

**Comment 11:** Equation 6- The symbol NCj is used to specify the same than the symbol n in equation 3. Equation 6 is unnecessary to explain the evaluation methodology.

**Response:** We believe that equation (6) is necessary to explain how the calculations are performed as it shows, explicitly, that $CO_2$ is selected as the reference gas for calculations; which is not the only choice. For example, Figure 4 in our manuscript shows that CO or $CH_4$ would have been acceptable choices for the reference gas as well. Besides, as stated in the previous response, our nomenclature follows that of Yokelson et al. (1999) who reported this equation in their paper and we think it is necessary for transparently communicating the methods we chose.

**Comment 12:** Equation 7- What is the meaning of $EFN_2O/CO_2$?

**Response:** This should read $ER_{N2O}/_{CO2}$. This typographical error has been corrected.

**Comment 13:** Table1- No specification about which data corresponds to which fire process (heading/flanking/back). Define Byram fire line intensity.

**Response:** We added fire spread mode and fuel moisture content as two extra columns to this table. We have also defined Byram fireline intensity in the caption to Table 1.

**Comment 14:** Figure 5- The colors for lines representing flanking and backfires are difficult to discriminate.

**Response:** We have halved the font size of each experimental fire to make it more readable.

**Comment 15:** Lines 19-25, page 23138- I did not understand this discussion. As far as I understood from the experimental part, the humidity of the combustible was always the same. So no influence of humidity variability on emissions could be detected because there was no humidity variability.

**Response:** It is the moisture content of the fuel and not the atmospheric humidity which we were trying to control. If we had significant variation in the fuel moisture content this would have added extra (unwanted) variability to our data. As stated on lines 1-2 page 23131 (and from column two in the revised version of Table 1) we achieved fuel moisture contents between 4.6-6.8% after oven drying. As a result, there was still some variability in fuel moisture for different experimental fires. Therefore, fuel moisture was not a "fixed" quantity

as suggested by the reviewer. The fact that there was still some residual variation in fuel moisture content motivated us to statistically test for whether this subtle variation in fuel moisture content influenced emissions factors. On lines 19-25 page 23138 we merely state that the covariate (i.e. fuel moisture content) did not affect emission factors with a p value of 0.60 (a highly non-significant result). In summary, we thought it would be better scientific practice to measure fuel moisture content for every burn and to statistically test for its impact on emissions factors, rather than to assume it was not a source of variation.

**Comment 16:** Section 4.1 is confusing because it is not clear which definition of EF is being discussed at each moment.

**Response:** We have added three extra sentences to the start of section 4.2 (in the revised manuscript) to remedy any potential confusion as to which emissions factor reporting method we are referring to. These three new sentences read: "In this section, we discuss a comparison between the two methods for reporting emission factors which are both based on a carbon mass balance approach (see section 2.4.1). As such, we switch interchangeably between reporting on a per unit element burnt basis (i.e. either carbon or nitrogen) or a per unit dry fuel consumed basis. The relevant equation number or associated units are provided to make it clear which emissions factor reporting method we are using."

As stated in the last of these three sentences we provide equation numbers or units to make it clear which method of reporting emission factors we are using.

**Comment 17:** Section 4.2- I could not understand and follow most of this discussion that now uses Emission Factors in percentages, mixed with the previous definitions of EFs.

**Response:** Several changes to this section have been made to make it clearer which emissions factor reporting method we are using. In all four paragraphs of section 4.3 (in revised version of the manuscript), when the term "emissions factors" appears we have added extra detail in parentheses following this term to indicate which emissions factor reporting method we are using. In addition, on line 1 page 23143 the emissions factors (per unit element burnt) have been changed from ratios to percentages to provide a consistent method of reporting them throughout the manuscript.

## References

Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, Global Biogeochemical Cycles, 15, 955–966, doi:10.1029/2000gb001382, 2001.

Byram, G. M.: Combustion of forest fuels, in: Forest Fire Control and Use, edited by Davis, K., chap. 4, pp. 61–89, McGraw-Hill, New York, 1959.

Cheney, N. P.: Fire behaviour, in: Fire and the Australian biota, edited by Gill, A. M., Groves, R. H., and Noble, I. R., pp. 151–176, The Australian Academy of Science, Canberra, 1981.

Hurst, D. F., Griffith, D. W. T., Carras, J. N., Williams, D. J., and Fraser, P. J.: Measurements of trace gases emitted by Australian savanna fires during the 1990 dry season, Journal of Atmospheric Chemistry, 18, 33–56, doi:10.1007/bf00694373, 1994a.

Hurst, D. F., Griffith, D. W. T., and Cook, G. D.: Trace gas emissions from biomass burning in tropical Australian savannas, Journal of Geophysical Research-Atmospheres, 99, 16 441–16 456, doi:10.1029/94jd00670, 1994b.

Jenkins, B. M., Kennedy, I. M., Turn, S. Q., Williams, R. B., Hall, S. G., Teague, S. V., Chang, D. P. Y., and Raabe, O. G.: Wind-tunnel modeling of atmospheric emissions from agricultural burning – influence of operating configuration on flame structure and particle-emission factor for a spreading type fire, Environmental Science & Technology, 27, 1763–1775, doi:10.1021/es00046a002, 1993.

Law, C. K.: Combustion Physics, Cambridge University Press, New York, 2006.

Lobert, J. M., Scharffe, D. H., Hao, W. M., and Crutzen, P. J.: Importance of biomass burning in the atmospheric budgets of nitrogen-containing gases, Nature, 346, 552–554, doi:10.1038/346552a0, 1990.

McArthur, A.: Control burning in eucalpyt forests, Forestry and Timber Bureau No. 80, Commonwealth of Australia, Canberra, 1962.

Meyer, C. P., Cook, G. D., Reisen, F., Smith, T. E. L., Tattaris, M., Russell-Smith, J., Maier, S. W., Yates, C. P., and Wooster, M. J.: Direct measurements of the seasonality of emission factors from savanna fires in northern Australia, Journal of Geophysical Research-Atmospheres, 117, doi:10.1029/2012jd017671, 2012.

Radke, L., Hegg, D., Lyons, J., Brock, C., and Hobbs, P.: Airborne measurements on smokes from biomass burning, in: Aerosols and climate, edited by Hobbs, P. and Patrick McCormick, M., pp. 411–422, A. Deepak Publishing, Hampton, Virginia, 1988.

Ranzi, E., Cuoci, A., Faravelli, T., Frassoldati, A., Migliavacca, G., Pierucci, S., and Sommariva, S.: Chemical Kinetics of Biomass Pyrolysis, Energy & Fuels, 22, 4292–4300, doi:10.1021/ef800551t, 2008.

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.

Wotton, B. M., Gould, J. S., McCaw, W. L., Cheney, N. P., and Taylor, S. W.: Flame temperature and residence time of fires in dry eucalypt forest, international Journal Of Wildland Fire, 21, 270–281, doi:10.1071/WF10127, 2012.

Yokelson, R. J., Goode, J. G., Ward, D. E., Susott, R. A., Babbitt, R. E., Wade, D. D., Bertschi, I., Griffith, D. W. T., and Hao, W. M.: Emissions of formaldehyde, acetic acid, methanol, and other trace gases from biomass fires in North Carolina measured by airborne Fourier transform infrared spectroscopy, Journal of Geophysical Research-Atmospheres, 104, 30 109–30 125, doi:10.1029/1999jd900817, 1999.