Response to Referee #1

We would like to thank anonymous referee #1 for the positive comments and suggestions. Below we address the referee's specific comments:

Specific comments

Comment (1) Introduction. I suggest adding a note in the introduction (e.g. at the end of Page 6316 Line 16) to explain that the paper is part 2 of a companion paper (Paton-Walsh et al., 2014), and briefly comment what part 1 and part 2 cover.

Reply (1) An explanation that this is Part 2 of a companion paper has been included in the revised introduction as suggested, with a brief description of what Parts 1 & 2 cover.

Comment (2) Page 6316 Line 24. mercury-cadmium-telluride (MCT) detector

Reply (2) This has been corrected as suggested in the revised manuscript.

Comment (3) Page 6320 Lines 5-9. It is not clear to me how different are the retrieval parameters from part 1 and part 2 of the work. Please clarify.

Reply (3) There are three differences: the spectral resolution (0.5 cm^{-1} here, 0.96 cm^{-1} in Part 1); the field-of-view (20 mrads here, 22 mrads in Part 1); and the apodisation function (triangle here, Hamming in Part 1). The comparison with Part 1 is not included in lines 5–9 and has been included in the revised manuscript for clarification of the differences between Part 1 and Part 2.

Comment (4) Page 6322 Line 2. Figure 4d is mentioned before Figures 4a-c. The Authors should rearrange Figure 4 to cite the images sequentially.

Reply (4) The text in the revised manuscript has been edited to refer to Figure 4 before any subfigures are specifically mentioned.

Comment (5) Page 6323 Line 11. "Figure 4c", same comment as above.

Reply (5) As for Reply (4)

Comment (6) Page 6324 Line 22. It is not clear what "this moves slowly" refers to. Please clarify.

Reply (6) The revised manuscript has been edited to state "the fire moves slowly".

Comment (7) Page 6325 Lines 25-26. Why did Paton-Walsh et al (2014) (part 1 of the paper) find C_2H_4 better correlated to CO2, whereas this paper finds C_2H_4 better correlated to CO? My understanding is that both papers use the same (or very similar) dataset.

Reply (7) Parts 1 and 2 use the same analysis techniques, but on two very different datasets. Part 1 is an analysis of data collected at Australian temperate forest fires, whilst Part 2 is an analysis of data collected at Australian tropical savanna fires. C_2H_4 is a product of the pyrolysis stage of combustion (Lobert & Warnatz, 1993). This occurs close in space and time to the CO_2-emissions-dominated flaming stage of combustion, which explains high correlation with CO_2. C_2H_4 is also a product of incomplete combustion during the smouldering stage of combustion (Lobert & Warnatz, 1993), which explains why it might also be highly correlated with CO. There was very little difference in the correlation between C_2H_4 and CO, and C_2H_4 and CO_2 for the fires measured in this paper. Across all 21 fires C_2H_4 was found to have a better mean correlation with CO. This was not the case in Part 1, where C_2H_4 was found to have a better mean correlation with CO_2. We suggest that this difference is real and due to differences in the vegetation (fuel) types and combustion efficiency of the fires.

Comment (8) Page 6328 Line 2. Missing parenthesis? (based on MCE<90

Reply (8) This has been corrected in the revised manuscript

Comment (9) Page 6328 Line 14. Drivers of variations in emission ratios *and* emission factors

Reply (9) This has been corrected as suggested, in the revised manuscript.

Comment (10) Page 6333 Line 14. I think it is Paton-Walsh et al (201*4*)

Reply (10) This has been corrected as suggested, in the revised manuscript.

Comment (11) Summary and Conclusions. I feel this section is mostly a summary of their work and lacks some conclusions...

Reply (11) The revised manuscript addresses this comment by adding to the conclusions and separating the summary from the concluding comments, these are currently intermixed.

Figures and Tables

Comment (12) Table 2. Table refers to *Y * compounds, whereas the text refers to *X * compounds. Please be consistent. What are the units of the emission ratios, ppm/ppm? Also, the caption of the table is long. The table would be much clearer is some of the text is moved to footnotes below the table.

Reply (12) The manuscript has been corrected so that all references to emission ratios use *X*. Emission ratios are not usually referred to using units. To be consistent with Part 1 (Paton-Walsh et al. 2014) and other emissions literature, we will keep this as unit-less. Some of the text from the Table caption has been moved to table footnotes.

Comment (13) Table 3. Table refers to *Y * compounds, whereas the text refers to *X * compounds. Please be consistent. What are the units of the emission ratios, ppm/ppm?

Reply (13) As for Reply (12)

Comment (14) Table 5. The caption of the table is very long. I think the table would be much clearer if most of the text is moved to a few footnotes.

Reply (14) The text beginning "*The emission factor reported by…*" has been moved to footnotes from the figure caption.

Comment (15) Figures 4a-b are not used in the text. The Authors should remove them or at least cite them in the text, for example with Figure 3 (page 6321 line 14)

Reply (15) The text in the revised manuscript has been edited so that the subfigures are not referred to individually.

Comment (16) Figure 7. "Only the first 60 s following ignition are dominated by smoke from purely flaming combustion, with increasing contribution from the smouldering phase combustion zone towards the end of this time series (as reflected by the decreasing MCE throughout)." This information is already in the text. I would omit it here.

Reply (17) This section of the figure caption has been removed as suggested, in the revised manuscript.

Comment (18) Figure 7. Figure 9. "Notice the higher correlation (R2) between each of the trace gases and carbon monoxide, than between each of the trace gases and carbon dioxide (Fig. 8).". This is already explained in the text. I would omit it here.

Reply (18) This section of the figure caption has been removed as suggested, in the revised manuscript.

Comment (19) Figures 11a, 12 and 13. I think the MCE observations should be presented consistently. Figure 11a presents MCE ranging from 80% to 100%, whereas it ranges from 78% to 100% in Figure 12 and from 84% to 98% in Figure 13.

Reply (19) The scale bar in Figure 11a is incorrect, and has been corrected to range from 78% to 100%, consistent with the scale bar in Figure 12. The range of MCE values in Figure 13 is dependent on having enough data for each vegetation type in each MCE bin for meaningful comparison between different vegetation types. Only one vegetation type (SOW) has enough data to extend this figure lower than the 84%–86% bin (as is evident when you compare Figure 11c with Figure 12), and no single vegetation type has enough data to extend this figure higher than the 98%–100% bin. Therefore, given that this figure is for an intercomparison of emission factors between different vegetation types, the existing MCE range will be kept for clarity.

References

Lobert, J. M. and Warnatz, J.: Emissions from the combustion process in vegetation, In: Fire in the Environment: the Ecological, Atmospheric, and Climatic Importance of Vegetation Fires, edited by: Crutzen, P. J. and Goldammer, J. G., John Wiley, New York, 15–37, 1993.

Paton-Walsh, C., Smith, T. E. L., Young, E. L., Griffith, D. W. T., and Guérette, É.-A.: New emission factors for Australian vegetation fires measured using open-path Fourier transform infrared spectroscopy – Part 1: methods and Australian temperate forest fires, Atmos. Chem. Phys. Discuss., 14, 4327–4381, doi:10.5194/acpd-14-4327-2014, 2014.

Response to Referee #2

We would like to thank anonymous referee #2 for the positive comments and suggestions. Below we address the referee's specific comments:

Comment (1) Section 4.1: Is the regression used to calculate the emission ratios similar to reduced major axis (RMA) regression? Is this another term for the RMA approach?

Reply (1) To the best of our knowledge after reading more about the RMA approach, we can see no differences between this and the regression approach used for this paper and Part 1 (Paton-Walsh et al. 2014). The purpose of using generalised least squares regression is that this approach takes into account measurement errors for both axes variables as seems to be the case for RMA regression.

Comment (2) Figure 8: Please increase the font size throughout.

Reply (2) We will wait until we see the final typeset version (in the ACP format) of Figure 8, before redrawing if necessary.

Comment (3) Figure 11: Please increase the font size on your axes and axes labels.

Reply (3) As for Reply (2), we will wait until we see the ACP-formatted version, before redrawing if necessary.

Comment (4) Figure 12: This is a nice Figure. Please discuss how other trace species behave w/r/t MCE - i.e. do they show a similar pattern as methane with approximately the same pattern of increasing emission ratio as MCE is reduced? I would actually recommend putting similar plots for the other species in the supplemental materials.

Reply (4) There is a large number of data in Figure 12 (thousands of individual measurements from all of the 21 fires). There is such a large number of data for CH_4 because CH_4 mole fractions are retrievable from FTIR spectra even at ambient concentrations. This wealth of data provides us with samples spanning the full range of modified combustion efficiencies (from 78% up to almost 100%). This is also the case for a number of other trace gases whose mole fractions in the smoke from these fires are well above the detectability limits of the spectroscopy (CO, NH_3, CH_3OH, CH_3COOH, CH_2O, C_2H_4). For these gases, which are produced predominantly during smouldering-phase combustion, we see the same relationship as is evident in Figure 12 for CH_4. For gases that are close to the detectability threshold (HCN, HCOOH, C_2H_2, C_2H_6) of the instrumental setup, we do not see a wealth of data, indeed, some of these gases were only detected at a few of the 21 fires (see Table 4). As such the corresponding plots for these gases are data sparse and it is difficult to draw the same conclusions. These plots have been made available as supplemental materials in this revised version.

References

Paton-Walsh, C., Smith, T. E. L., Young, E. L., Griffith, D. W. T., and Guérette, É.-A.: New emission factors for Australian vegetation fires measured using open-path Fourier transform infrared spectroscopy – Part 1: methods and Australian temperate forest fires, Atmos. Chem. Phys. Discuss., 14, 4327–4381, doi:10.5194/acpd-14-4327-2014, 2014.