

Interactive comment on “Combustion efficiency and emission factors for US wildfires” by S. P. Urbanski

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Response to Referee #2

We would like to thank all three referees for their constructive comments and suggestions. The referees' comments and suggestions have greatly improved our manuscript. We truly appreciate the effort that all three referees invested in reviewing our manuscript. The original comments of referee #N are labeled RN.X and our response is labeled AN.X. We have proposed significant revisions to some sections of the manuscript. These significant revisions may respond to the comments of multiple referees and are provided as a supplement. The proposed significant revisions are labeled SR.Y and are provided in order according to page and line number of the original manuscript. When one of the referee's comments has been addressed with significant

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revisions to the manuscript the relevant SR.Y are referenced.

Referee #2

The manuscript presents an analysis from three wildfires and one prescribed fire in mixed conifer forest fuels in the northern Rocky Mountains, US. Measurements of CO₂, CO, CH₄, and H₂O were made using CRDS techniques from an airborne platform. This proved to be a unique opportunity to measure biomass burning (BB) emissions from temperate wildfires, as the majority of BB studies in temperate fuels have been made on prescribed fires, which the author suggests may not be representative of emissions from wildfires. Emission factors for CO, CO₂, and CH₄ are presented along with 14 additional species that were extrapolated using EF-MCE linear relationships from the literature. EFs from this work are compared with EFs from previous field studies of temperate forest fires and from 18 prescribed fires from the literature. A relationship between fuel composition and the modified combustion efficiency (MCE) is observed from a noted decrease in average MCE with the increase in ratio of heavy fuel consumption to total fuel consumption. This work suggests that EF variability may be strongly influenced by fuel composition. Considering that the majority of fuel consumption by wildfire occurs in the western US and these fuels often have significant accumulation of heavy fuels, low MCE fires (high CWD) and their emissions may best represent many “typical” temperate wildfire emissions. If representative of temperate wildfires, measured and estimated emission factors from this work suggest a large underestimation of wildfire emissions as reported from published BB reviews and inventories that are based on higher MCE fires. This effort is an important step towards improving the accuracy emission inventories and towards maintaining compliance with National Ambient Air Quality Standards. As the author stresses, it is critical that BB inventories and models are “supplied” with the most accurate emission factors possible. This proves difficult because wildfires are difficult to study and wildfire fuel types in the US are numerous and diverse. Devising techniques that acknowledge this and work with these limitations is imperative towards improving the accuracy of current models.

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I recommend publication of this manuscript after the author addresses some minor concerns outlined below.

General Comments: R2.1. The author discusses the shortage of temperate wildfire measurements which are needed for wildfire emissions inventories. A valid point was made in General Comment #4 by Referee 1 and I emphasize it here: Could other literature fires be considered as part of the database on wild forest fires? For example, boreal fires are mostly wildfires and the MCE of boreal forest fires (Akagi et al., studies with an airborne data only, found in supplementary tables) often reflect greater amounts of smoldering combustion compared to temperate prescribed fires. Additionally, in the review of Akagi et al. (2011), their supplementary information provides a breakdown of temperate forest emission factors by fire type and/or fuel type (e.g. temperate wild fire, temperate prescribed fire, understory fuels, organic soils, debris, etc.). While it is true that temperate wildfire measurements are scarce, there exist several options to best estimate temperate wildfire EF. I would be curious to see a comparison in Table 3 including A11 EF averages of ONLY temperate wildfires, or A11 EF airborne averages from boreal fires, with the hope that the ratios of “this study/A11” will be closer to 1.

A2.1. We have revised our approach for estimating EF for species not measured in our study. Please see our response to Referee #1 comment R1.7. We have updated Table 3 to include A11 boreal data. We have not included the A11 TF wildfire data only – these include EF from pine-oak forest fires at tropical latitudes (Mexico).

R2.2. Estimating the contribution of emissions from wildfires is difficult in multiple levels, as the author explores. One difficulty not mentioned in this paper is the trouble in distinguishing a wildfire from a prescribed fire (fire type). For those people who use wildfire emissions inventories, is there a standard method they use to further classify a temperate fire as wild or prescribed? I mention this because this work suggests that temperate BB EF may be the largest source of error in the management of regional air quality. Temperate EF reflecting both prescribed and wild fires may be most appropriate if our current methods of identifying fire type cannot distinguish between the

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two.

A2.2. We have better defined the focus of our paper to “wildfire season fires in mixed conifer forests of the Rocky Mountains”. We argue that the low MCE of the fires measured in our study were driven largely by: 1) the fuel present - ample loadings of CWD and 2) conditions which facilitate the combustion of CWD and duff – primarily low fuel moistures. Thus the low MCE is not strictly a wildfire characteristic. The North Fork Fire measured in our study was a prescribed that met these criteria. Also, the Shaver fire sampled by Burling et al. (2011) falls in this category as well as the burning of dried logging slash studied by Hobbs et al. (1996). The temperate forest (TF) EF in the literature are based largely on prescribed fires that do not meet these criteria – they burned under conditions of high CWD moisture and high duff moisture, which inhibited consumption of these fuel components, and/or they burned in forests with minimal loads of CWD and duff. Because much of the prescribed burning in the western US is conducted outside the wildfire season, these literature TF EF are probably appropriate for most prescribed fires, but we believe not so for many wildfires (or the minority of prescribed fires that occur during the wildfire season). In our revised paper we state our measurements are applicable to fires that occur during the wildfire season in mixed conifer forests of the Rocky Mountains and likely areas with similar forest types (Lodgepole Pine, Douglas-fir, Engelmann Spruce / Subalpine fir) throughout the western US. We define and describe the western US wildfire season (June – September (June - October in California)), and note that most prescribed burning in the west takes place outside the WF season. This information may be used as guidance for applying our results in emission modeling. Regarding the uncertainties involved in estimating BB emissions, we have added a paragraph describing the many uncertainties. We note that fuel loading is considered the greatest uncertainty for estimating emissions from wildland fires in temperate and boreal forests. Significant manuscript revisions addressing this comment are: SR.2, SR.10

R2.3. A concern I have is that the primary conclusion from the paper, that consumption

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of heavy fuels favors lower MCE, was not quantitatively supported from data in this work. While the data presented by the author is of great interest to BB and air quality/monitoring communities, the conclusions drawn here are quite broad and need to be limited to what was actually established. Since fuel consumption was not measured for the 4 fires in this work, conclusions should emphasize what was found from these 4 fires, rather than what was found in the 18 fires measured in the literature.

A2.3. We revised the paper to de-emphasize our conclusion that the consumption of heavy fuels favors lower MCE. Our revision emphasizes that our hypothesis regarding the MCE – heavy fuel link is highly speculative. There was some disagreement between the 3 referees regarding the value of this analysis and the role it should play in the paper. Our revisions strike a balance among the referee's varied comments on this matter. Significant manuscript revisions addressing this comment are: SR.11, SR.12

R2.4. My last concern is the representativeness of the small sample size of three wildfires that all occurred in the same state, and in similar fuels. This point was mentioned in General Comment #1 by Referee 1, and I highlight it again here. Temperate wildfires can burn all types of vegetation, from grasses to chaparral shrubs to hardwood forest. While figure 4 shows pretty clear trends between MCEs of SE, SW, NW, and WF, it seems these trends also emphasize the inherent variability of prescribed fires from region to region (SE, SW, NW). We need to consider that the classic "temperate wildfire" MCE may also show similar variability across regions and fuel types, and at this point, it is questionable if there is enough data to say that the three wildfires measured in this work are representative of all US wildfires. While it is true that most wildfires in this nation occur in the west, I would argue that more than three fires are needed given the large fire-to-fire variability.

A2.4. We have refined the scope of our paper to wildfire season fires in mixed conifer forests of the northern Rocky Mountains. In our revised paper we have made it clear that our measurements and analysis pertain to specific forest types. As we argue in A2.2, we believe the low MCE measured for the 4 fires in this study were driven in large

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part by the presence of ample CWD and conditions that promoted the consumption of these fuels (primarily low fuel moisture). In our revised paper we state our measurements are applicable to fires that occur during the wildfire season in mixed conifer forests of the Rocky Mountains and likely similar forest types throughout the western US. In response to referee #1 & referee #3 we have added additional information regarding the vegetation types involved. The four fires sampled in our study burned forest dominated by Lodgepole Pine, Douglas-fir, Engelmann Spruce / Subalpine Fir. These forest types accounted for about 19% of total area burned and about 43% of forest area burned by wildfires in the western US from 2001-2010 (see response to comment 3 of Referee #1(R1.3/A1.3)). We believe the 4 fires sampled in this study are far more diverse than the referee acknowledges. While the fires occurred in only two states (North Fork Fire in Idaho, Hammer Creek & Big Salmon Lake in Montana, and Saddle Complex in both Montana & Idaho) they occurred in two different ecological provinces: the Northern Rocky Mountains (North Fork Fire, Hammer Creek & Big Salmon Lake) and the Middle Rocky Mountains (Saddle Complex) (USDA, 2007; Bailey 1995). Further, the ecosystems while sharing dominant forest types were distinct from one another. The Hammer Creek and Big Salmon Lake fires occurred in the Bob Marshall Wilderness of northwestern Montana. The fire regime of the Bob Marshall Wilderness has regions of mixed severity fire regime - high to low frequency of return (25 to 150 year fire rotation) and stand replacing fire regime – low frequency of return and regions (120 to 350 year fire rotation) (Teske et al., 2012). The Hammer Creek Fire, which burned into previous burns (burns less than a decade old), was located in the South Fork Flathead drainage an area where Douglas-fir/Lodgepole Pine forests are maintained by mixed-severity fire regime (Arno et al., 2000). While dominant species in the Hammer Creek and Big Salmon Lake burn areas were Lodgepole Pine, Douglas-fir, and Engelmann spruce/ subalpine fir, Larch was an important species in both areas (Arno et al., 2000; Keane, 2013). The Hammer Creek fire included areas where Ponderosa Pine was an important species (Arno et al., 2000; Larson, A, 2013). Both the Hammer Creek and Big Salmon Lake fires burned into previous burns (fires in 2003). The Saddle Complex

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was adjacent to Frank-Church River of No Return Wilderness and partially burned into the wilderness area. The Frank-Church River of No Return Wilderness has areas of low severity (high frequency, 4 to 84 year fire rotation), mixed severity (low to high frequency, 35 to 105 year rotation) and stand replacing (low-frequency, 40 to 200 year rotation) (Arno, 1980; Teske et al., 2012). Overall the fire regime of the Frank-Church River of No Return Wilderness is classified as mixed severity regime (Teske et al., 2012). The Saddle Complex did not burn into the area of a recent burn (no burn since 1984), but the area burned was heavily impacted by mortality due to insects (as described in Sect. 2.1. of the paper). The North Fork Prescribed Fire burned in an area that had not been impacted by significant fire since 1910. The North Fork Prescribed Fire burned in the North Fork Clearwater River basin of the Clearwater National Forest, an area maintained by high severity fire regime (in-frequent, i.e. long return interval, stand replacing fires) (Smith and Fischer, 1997). The burn area had been impacted by tree mortality due to insect activity as noted in Sect 2.1 of the paper.

Further, as we argue in the paper, the two fires that were sampled on multiple days exhibited different fire behavior (original Table 1, revised Table S1) and burned in different areas and should be considered as different fires. The fire activity we observed and that reported by the incident management teams varied from day to day (revised Table S1). The canopy fire activity we observed was patchy and intermittent, observations corroborated by the fire management team reports (revised Table S1) and the USFS Rapid Assessment of Vegetation Condition after Wildfire (RAVG) analysis of these wildfires (<http://www.fs.fed.us/postfirevegcondition/index.shtml>). The mix of vegetation involved, terrain, and fuels likely varied among days. Even in forests with the same dominant forest type, surface fuel loadings are extremely variable spatially (see e.g. Keane et al., 2012). The Big Salmon Lake (BSL) Fire grew by over 4 km² between Aug 17 and Aug 28. When sampled on August 28 the BSL fire the east and west fire fronts were separated by 7km. The area burned by the BSL fire had a spatially and temporally variable disturbance history of previous burns and mortality due to insects. The Saddle Complex was sampled on 4 consecutive days and the burned area grew between 2

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and 8 km² on these days. When sampled the Saddle Complex had active fire fronts separated by ~12 km east – west and ~10 km north – south. The Saddle Complex burned in an area that was ~25 to 45% impacted by insect induced tree mortality. Considering: 1) the variability in daily observed fire behavior, 2) the spatial variability in fire severity indicated by the post-fire fire severity assessment, 3) the difference in the daily spatial extent of the fire, 4) the spatial variability of disturbance history, 5) the complex terrain, and 6) the natural spatial variability of wildland surface fuels (e.g. Keane et al., 2012) we believe our treatment of each day as a separate fire (i.e. fire-day), is very reasonable. We consider our fire sample size 9 not 4.

References

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Teske, C. C., Seielstad, C. A., and Queen, L. P.: Characterizing fire-on-fire interactions in three large wilderness areas, *Fire Ecology*, 8, 82-89, doi:10.4996/fireecology.0802082, 2012.

USDA Forest Service ECOMAP Team, 2007 (http://fsgeodata.fs.fed.us/other_resources/ecos)

R2.5. P36, L21: I am not familiar with the Regional Haze Rule, nor Regional Haze Regulations (P36, L14). It may be worthwhile to add a sentence or two detailing what these regulations monitor and/or aim to maintain (e.g. is it PM2.5, BC, NOx, SO2, etc.?) and if the monitored species were measured in this campaign.

A2.5. We have added the following sentences at P36 L14: "The purpose of the Regional Haze Rule is to reduce pollution which causes visibility impairment in national parks and wilderness areas. Particulate matter is the primary contributor to visibility impairment in areas covered by the Regional Haze Rule." We have also changed "Regional Haze Regulations" to "Regional Haze Rule" for consistency. The later term is the official designation although "Regional Haze Regulations" and simply "Regional Haze" are often used.

R2.6. P37, L25-28: It would be appropriate to mention the Akagi et al. (2013) study in this discussion. A central question in their work involved the differences in PF emissions resulting from burn history, along with additional factors such as time of year, fuel moisture, fuel composition, and atmospheric conditions, and how these may influence BB emissions. A brief discussion of key findings would strengthen and complete this section on previous work.

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A2.6. We believe the referee meant P47 not P37. We have included the study of Akagi et al. (2013) in our revised Sect. 3.2 Comparison with other studies. This section has been significantly revised please see: SR9. Also, the data from Akagi et al. (2013) have been added to Fig. 4 and have also been used in our revised EF- MCE analysis (at the suggestion of referee #1). Please see responses to referee #1 (R1.7/A1.7) for details.

R2.7. P69, Table 1: Consider adding a column for "Fuels" or "Vegetation Type" to give the reader an idea of what CWD (if any) may have been present. Since no measurements of fuel consumption were made and the influence of fuels on MCE is an important conclusion of this paper, any fuels data would be very beneficial here.

A2.7. We have revised Table 1 to include "Vegetation Involved as Percent of Burned Area".

R2.8. P44, L13: Following the format of the proceeding sentences, maybe provide 1 or 2 examples of NMOC that have been linked with both flaming and smoldering combustion (ex. C2H2)

A2.8. The text at P44. L5 – P44 14 has been merged into the Introduction at P37, L16 – 22. This section (P36, L23 – P38, L21) has been significantly revised. Specific revisions to the manuscript are: SR.2

R2.9. P45, L13-P46, L2: It seems this information may be more appropriate if moved to Sect. 2.1.3 or 2.3.

A2.9. Text from P45, L13 -21 has been moved into Sect. 2.1. Text from P45L21 – P46L2 has been moved to Sect. 2.3. Please see specific revisions: SR.4 and SR.5

R.2.10. P47, L16: I would like to see the South Carolina airborne prescribed fire data from Akagi et al. (2013) here. Fuels burned in their study were similar to those from B11.

A2.10. We have included the study of Akagi et al. (2013) in our revised Sect. 3.2

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Comparison with other studies. This section has been significantly revised please see: SR9. Also, the data from Akagi et al. (2013) have been added to Fig. 4 and have also been used in our revised EF- MCE analysis (at the suggestion of referee #1). Please see responses to referee #1 (R1.7/A1.7) for details.

R2.11. P48, L8: There are many other factors in addition to time of year that affect fire behavior, as the author mentions. Is there any data on the burn history of the North Fork Prescribed Fire plot? As noted in Akagi et al. 2013, the burn history (and frequency of burning) may also affect fire behavior. I question whether we can call this prescribed fire a wildfire just because it burned during wildfire season, since this paper aims to distinguish the differences in emissions between the two.

A2.11. The area of the North Fork burn had not been impacted by significant fire since the historic fires of 1910. We have added fire history of the North Fork Fire to Sect. 2.1.4. As discussed in our response A2.2, we have better defined the focus of our paper to “wildfire season fires in mixed conifer forests of the Rocky Mountains”. We argue that the low MCE of the fires measured in our study were driven largely by: 1) the fuel present - ample loadings of CWD and 2) conditions which facilitate the combustion of CWD and duff – primarily low fuel moistures. Thus the low MCE is not strictly a wildfire only characteristic. The North Fork Fire measured in our study was a prescribed that met the criteria above.

R2.12. P49, L20: What is the effect of elevation on fires and fire emissions? Is it just used to explain the different types of vegetation?

A2.12. The elevation has been included to explain the different vegetation types and provide some feel for the diversity of the terrain.

R2.13. P51, L11: I would like to see more discussion of the data presented in Table 3. It was mentioned earlier in the paper that EF are inflated by 5% from using the CMB method how do the EF from this study compare to A11 or NEI, given this adjustment? Maybe consider adding a column of “adjusted” EF?

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A2.13. Table 3 includes uncertainties for our measured and estimated species. The 5% inflation of EF is small compared to these uncertainties. Any attempt to adjust for this 5% inflation would be overwhelmed by the stated uncertainties included in Table 3. Section 3.3 (including Table 3) has been significantly revised based on suggestion/comments of referee #1. The discussion of our measurements has been expanded (e.g. it now includes boreal forest EF from Akagi et al. 2011). For details please see specific revisions: SR.8, SR.9, SR.10

R2.14 P53, L19-20: I am confused about the linkage between fuel moisture and MCE. This work seems to support that low MCE was the result of available CWD, made available by low fuel moisture. This sentence, on the other hand, seems to suggest that MCE tends to increase with decreasing fuel moisture for a constant fuel type and fuel mass (as found by recent laboratory studies). Please clarify this in the text.

A2.14. We do not believe the lab results are necessarily inconsistent with our analysis or the findings of others (e.g. Akagi et al., 2011). The lab studies we cite observed an MCE – fuel moisture relationship for homogeneous fuel beds, which we didn’t clarify. The studies also focused on fine fuels. Our analysis (and that of Akagi et al., 2011) pertains to the heterogeneous fuels found in the natural environment. Even if the MCE of fine fuels is higher during the wildfire season compared to a spring/fall prescribed burn, the amount of fine fuel consumed will be similar. Increased emissions from the increased consumption of CWD & duff, which burns with a lower MCE than fine fuels, could easily offset the MCE gain due to drier fine fuels. This is the scenario we layout P53, L25 – P 54, L17. To clarify that the lab studies which reported MCE – fuel moisture link focused on homogenous fine fuels we have revised the text at P53, L18-21 as follows: “In addition to fuel geometry and arrangement, recent laboratory studies suggest a linkage between fuel moisture and MCE, with MCE tending to increase with decreasing fuel moisture for a homogeneous fine fuels constant fuel type and fuel mass (Chen et al., 2010b; McMeeking et al., 2009).”

R2.15. P54, L10-14: This was observed in NC and SC in B11 and Akagi et al. (2013),

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respectively. B11's fires took place in the early spring, and they saw generally higher MCEs for conifer prescribed fires than Akagi et al., who burned under dry conditions during wildfire season and saw relatively lower MCEs than B11. I would add a sentence or two on this to support this speculation.

A2.15. The following text has been added at P54, L17: "The prescribed fires studies of B11 and A13 showed evidence of such an effect. The B11 North Carolina prescribed fires burned in the spring under conditions of high fuel moisture and MCE were high, averaging 0.948. While occurring in nominally similar forests, the prescribed fires studied in A13 were burned during the fall prescribed fire season before the region had fully recovered from a prolonged drought. The average MCE of the A13 fires was 0.931."

R2.16. P34, L9: Add comma after "decade"?

A2.16. Comma added

R2.17. P34, L9 and P37, L10: Change "has been realized" to "has been made"?

A2.17. Changed to: "has been made"

R2.18. P36, L 15-16: Consider changing "quantifying the contribution of wildfires to O3 related air quality degradation is difficult" to "quantifying the contribution of wildfires to O3 formation is difficult"

A2.18. This sentence has been changed to: "Because O3 is a secondary pollutant resulting from complex chemistry, quantifying the individual contribution of wildfires or prescribed fires to O3 formation is difficult".

R2.19. P37, L4: Consider "day-time scale" instead of "day time scale"

A2.19. We have decided to leave the text as originally written.

R2.20. P39, L22: Consider rephrasing "The Big Salmon Lake fire started, cause undetermined, 16 August 2011". This seems a little choppy.

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A2.20. This sentence has been changed to: "The Big Salmon Lake Fire started from an unknown cause on August 16, 2011 in the Bob Marshall Wilderness in northwestern Montana, about 10 km northwest of the Hammer Creek Fire."

R2.21. P40, L10: Change "The Stud Fire which, was also caused by lightning,..." to "The Stud Fire was also caused by lightning and..."

A2.21. The sentence has been changed to: "The Stud Fire was also caused by lightning and began on August 14 in the Salmon-Challis National Forest in Idaho."

R2.22. P41, L21: Delete comma after "in-flight"

A2.22. The comma has been deleted.

R2.23. P42, L24: Delete "a" before "several km"

A2.23. This section has been significantly revised. Please see specific revision: SR.5

R2.24. P43, L7: Change "of compound X, X, was" to "of compound X (X) was"

A2.24. We have changed the text as suggested.

R2.25. P43, L15: Change "CH4 to CO2, CH4/CO2, was" to "CH4 to CO2 (CH4/CO2) was"

A2.25. We have changed the text as suggested.

R2.26. P43, L17: Change "12 the molar mass" to "12 is the molar mass"

A2.26. We have changed the text as suggested.

R2.27. P43, L12-14: Please add a sentence or two on how the two listed Approaches compare in terms of EF (e.g. variability within X%). You may want to move P46, L9-10 here.

A2.27. We have added the following: "The average EF calculated using the two methods agreed within 10%." Please see response to referee #3 (R3.15/A.15) for related

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changes.

R2.28. P44, L24: Add “Akagi et al., 2013” after “Burling et al., 2011”

A2.28. “Akagi et al., 2013” has been added after “Burling et al., 2011”

R2.29. P45, L13: Change to “Fire perimeters, areas of active burning, and regions of smoke. . .”

R2.30. P45, L13-15: Awkward wording, consider removing “the Saddle Complex on 24 August”?

A2.29/A2.30. This sentence had been modified as suggested. Also, this material has been moved to Sect. 2.1 following the suggestion of referee #3. Please see specific revisions SR3 and SR5. Please see response to referee #3 (R3.15/A.15) for related changes.

R2.31. P45, L18: Do we know if the “pockets of burning” were mostly flaming or smoldering combustion?

A2.31. No. We cannot definitively state if the pockets of burning were mostly flaming or smoldering.

R2.32. P46, L20: Add comma after “previously”

A2.32. We have added comma after “previously”. This section has been significantly revised. Please see details please see specific revision: SR.6

R2.33. P47, L3: What is meant by “muted”?

A2.33. We have changed “muted” to “limited”. This section has been significantly revised. Please see details please see specific revision: SR.6

R2.34. P49, L23: Change “involved” to “burned”?

A2.34. We have changed “involved” to “burned”. This section has been significantly revised. Please see details please see specific revision: SR.7

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R2.35. P52, L27-28: Possibly delete this sentence, as this was clearly conveyed earlier in the paragraph

A2.35. This section has been significantly revised in response to comments from referee #1 and referee #3. Please see details please see specific revision: SR.10

R2.36. P53, L8: What is meant by “soundness”? I'd clarify this or suggest a different word.

A2.36. The has been changed to: “the state of decay of dead wood”

R2.37. P54, L4: Consider changing “fuel particles” to “fuels”.

A2.37. We have left this text unchanged. The term fuel particle(s) is has specific meaning in fire science and is commonly used.

R2.38. P54, L27: Add comma after “Turtle burn”

A2.38. Comma added after “Turtle burn”

R2.39. P57, L12: Delete “and EFCO2”, as it is implied from “lower MCE”?

A2.39. Yes, it is implied. However, we prefer to specifically state higher EFCO here in the Conclusion as well as the Abstract.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/13/C953/2013/acpd-13-C953-2013-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 33, 2013.