Reviewer #1 (Bob Yokelson) Comments and Responses

General Comments:

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- 5 This manuscript reports much needed, very important, high-quality boreal forest fire smoke measurements with impressive modeling support and the work should very much be published. Unfortunately, there seems to be an error in the calculation of emission factors (EFs) explained in detail below. If so, that will require revisions to reported values, re-interpretation of the implications, and re-review. As explained below, the data may in fact support earlier EFs rather than suggest they should be higher. I am submitting a quick, rough
- 10 review so the authors can correct this if needed or validate their calculation if appropriate. I'm happy to communicate directly with the authors about the calculations and to review the paper in more detail after the calculations can be verified to be correct, and, if needed, the analysis and conclusions are appropriately modified.
- 15 A second, relatively minor, general comment is that there is some missing context that could be added to the intro or discussion that could help motivate why the authors data is so valuable and perhaps inform the interpretation. I'll summarize that next.
- Bertschi et al., (2003) showed that adjusting EFs for rarely sampled residual smoldering combustion (RSC)
 led to important adjustments in the EFs for all fire types and especially for fires burning heavy or duff fuels. Christian et al., (2007), Burling et al., (2012), Akagi et al., (2013; 2014) and others all supplemented airborne measurements with ground-based measurements on the same fire to explore this, but the relative importance of weakly lofted smoldering and flaming emissions could only be crudely estimated from size-/type-resolved fuel consumption measurements, which are challenging and rare. Yates et al., (2015) showed that even
- 25 airborne measurements can imply a much larger smoldering/flaming ratio late in long-lasting fires. Saide et al., (2015, and references therein) showed that rarely sampled nighttime combustion is both important and underestimated in some cases using commonly assumed diurnal cycles. So there is precedent and ample support in the literature for factoring in smoldering and nighttime combustion, but little data to judge the potential differences in emissions or the relative production. For this reason, Selimovic et al., (2019a, b)
- 30 deployed ground-based smoke monitoring downwind of hundreds of fires burning at all stages for two fire seasons. A priori, one might suspect that ground-based sampling could be biased towards smoldering and airborne sampling to flaming, but these authors found that conserved tracers sensitive to flaming (BC) and smoldering (CO) had a similar ratio from both air and ground. This implies both platforms are relevant and maybe even in sufficient agreement for some purposes. Other findings from this work are
- 35 relevant/comparable to the authors work as well. Even earlier, the widely used Akagi et al., (2011) recommendations for boreal forest fire EFs had been based on averaging ground and airborne measurements together as a "best guess" at overall EFs. Finally, It's very likely that ground-based downwind measurements are best for validating AQ models, but it may be that satellite or aircraft vertical profiles will be needed to best probe overall emissions. Climate assessments may be more interested in smoke in higher layers, which 40 may be missed by towers? However, this work is an extensive and welcome addition to the information
- 40 may be missed by towers? However, this work is an extensive and welcome addition to the information available.

Next some details on why it is unclear if the authors got "much higher EFCO" and whether their work actually implies more smoldering than previously assumed since MCEs are directly measured and similar to some 45 widely used previous work.

To start, I compare the authors EFs at face value to those from some widely-used recommendations: namely Andreae and Merlet 2001, now updated (Andreae, 2019) and Akagi et al., (2011). Akagi et al recommended a 50/50 average of the ground-based and airborne EFs in their boreal recommendations. For boreal the 50/50 ground/air led to EFCO of 127(45) g/kg compared to the authors 145(46) g/kg in their Table 1. So if there

EF is correct it is 14% higher. They are closer to the A11 ground-based average of 157. Andreae (2019) recommend the straight average of 20+ studies, which is 121.4(46.6). Putting the EFs and MCEs together in a table reveals some things.

		EFCO	MCE	EFCH4	n
	A11	127(45)	.881	5.96(3.14)	7 studies multiple fires per study
	A19	121.4(46.6)	.89	5.5(2.5)	20+ studies
5	Wiggins	145(46)	0.879(0.068)	6.05 <mark>(</mark> 2.09)	35 fires

While the authors current calculated EFCO is about 15% larger, the directly-measured MCEs are very close so maybe this new data does not imply more smoldering? Also this works MCE of 0.879(0.068) is not far from Selimovic et al (2019) estimated MCE (based on BC/CO) for similar long-lasting fires in heavy fuels
of 0.87(0.02).

The similarity in MCE along with non-standard notation in eqn 2 and a lack of definition for the authors "S" scalar inspired me to calculate EF directly from their emission ratios (ERs). Using the authors quoted assumption of 45% C for the fuel; I get different EF values than them: EFCO2 1437, EFCO 126.2, EFCH4

15 5.23. These EF values are the same or lower. If my calculation is right, then this new work supports the previous work rather than suggesting the values in use should be increased. It's still good data even if it agrees with previous work. Also %C > 45% is possible for boreal fires. 50% C is often assumed though one study (Santin et al., 2015) did measure fuel C close to 45% for a fire in boreal forest. But using the authors average ERs I have to assume 519 gC/kg to get close to their EF for CH4 and CO; and ~52%C seems to high.

20 I'd be happy to share my calculation (Yokelson et al., 1999) and re-review a revised paper if necessary.

Another possible reason for an ER-EF mismatch is using different averaging schemes for these two quantities? Ideally the averaging scheme should be the same for both quantities. If possible, it might be good to weight for how much smoke was produced at the fire, received at the tower, duration of events, or etc. Exploring how the average depends on the scheme employed is always useful and could be reported along

with a clear explanation of how the averaging was done for the reported values.

A few other things I noticed in order of Page, Line. This is a one-skim set of potentially useful comments. A more careful review could be done after ensuring the calculations are accurate.

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We appreciate Dr. Yokelson's positive comments that our manuscript provides needed and important CO and CH4 emission factor measurements from boreal forest fires.

We agree with his assessment that there was an important omission in our emission factor calculation. 55 Finally, Dr. Yokelson provides a valuable perspective (and references) on past work that has compared ground-based and aircraft-based estimates of emission factors.

We propose the following major revisions to our paper to address these issues.

40 First, our emission factor calculations will be corrected. This is easy and relatively straightforward to implement. More importantly, we will reinterpret our results and their implications in our revised manuscript, taking into account the revised emission factor information. This will require revisions to the title and abstract as well as main text.

Second, we will change equation 2 in our manuscript to standard notation as published in previous studies (Yokelson et al., 1999) and offer more clarity on the definition of the variables.

Third, will modify the introduction and discussion to include more context to motivate the importance 5 of this study and inform the interpretation of our results. We plan to include the studies Dr. Yokelson highlighted in our revised manuscript. Specifically, we plan to integrate work by Bertschi et al. (2003), Christian et al. (2007), Burling et al. (2011), Akagi et al (2014), Santin et al. (2015), Yates et al. (2016), Andreae (2019), and Selimovic et al. (2019a,b), and Yokelson et al. (1999).

- 10 Fourth, as describe above, we have carefully evaluated emission ratio and emission factor observations from past measurements of boreal forest fires in North America and Siberia, taking into account studies reported by Andreae (2019) and also reports provided by Yokelson in his review. A compilation of these studies will now be provided in our revised Table 1. In this context, we note that in comparison to North American boreal forest fires sampled by aircraft, our CO emission ratios are still considerably
- 15 higher, implying our observations do provide evidence for stronger role of smoldering combustion. Fires in boreal Siberia tend to have even higher CO emission ratios than North American fires, which is consistent with well known differences in fire behavior between the continents (Rogers et al., 2015). We look forward to the reviewer's perspective on the new analysis in this revised Table. A draft of the table is included here.

Study	CO Emission	MCE	# Fires	
-	Ratio			
Airborne W	/ildfires North A	merica		
Cofer et al., 1989	0.069 ± 0.004	0.935 ± 0.004	1	
Cofer et al., 1998	0.140 ± 0.012	0.878 ± 0.009	1	
Friedli et al., 2003	0.100 ± 0.020	0.909 ± 0.017	1	
Goode et al., 2000	0.085 ± 0.008	0.922 ± 0.007	4	
Laursen et al., 1992	0.050 ± 0.007	0.953 ± 0.006	1	
Nance et al., 1993	0.078 ± 0.012	0.928 ± 0.011	1	
O'Shea et al., 2013	0.150 ± 0.024	0.871 ± 0.012	4	
Radke et al., 1991	0.116 ± 0.087	0.896 ± 0.075	1	
Simpson et al., 2011	0.110 ± 0.070	0.901 ± 0.061	5	
Fire Weighted Mean	$\textbf{0.102} \pm \textbf{0.033}$	$\textbf{0.908} \pm \textbf{0.027}$	19	
Airborne Manag	gement Fires Nor	rth America		
Cofer et al., 1990	0.086 ± 0.008	0.921 ± 0.007	2	
Cofer et al., 1998	0.095 ± 0.016	0.913 ± 0.013	7	
Radke et al., 1991	0.047 ± 0.032	0.956 ± 0.030	4	
Susott et al., 1991	0.060 ± 0.061	0.943 ± 0.058	1	
Fire Weighted Mean	$\textbf{0.077} \pm \textbf{0.022}$	0.929 ± 0.020	14	
Labora	tory North Amer	rica		
Bertschi et al., 2003	0.151 ± 0.040	0.870 ± 0.030	_	
Burling et al., 2010	0.209	0.827	-	
Mcmeeking et al., 2009	0.091 ± 0.038	0.917 ± 0.068	-	
Mean	0.150 ± 0.039	0.871 ± 0.049		
Siberia – Surface and Airborne				
Cofer et al., 1998 (A)	0.224 ± 0.036	0.817 ± 0.025	1	
McRay et al., 2006 (A & S)	0.249 ± 0.064	0.800 ± 0.043	6	
Vasileva et al., 2017 (S)	0.126 ± 0.007	0.888 ± 0.005	2	
Fire Weighted Mean	$\textbf{0.219} \pm \textbf{0.048}$	0.822 ± 0.033	9	
Ground W	ildfires North Aı	merica		
Wiggins et al., 2016	0.128 ± 0.023	0.887 ± 0.018	3	
This study	0.142 ± 0.051	0.878 ± 0.039	35	
Fire Weighted Mean	$\textbf{0.141} \pm \textbf{0.049}$	$\boldsymbol{0.879 \pm 0.027}$	38	

Fifth, we will add text to the methods to describe how we average the different individual fire events together to come up with a season-wide mean. We will explore the sensitivity of this to the averaging 5 techniques, also reporting at CO₂ anomaly-weighted mean.

We respectfully ask the editors to allow us to update our calculations and revise the corresponding manuscript introduction and discussion prior to the next iteration of reviews. Below we address specific comments but note some of the responses will depend on our corrected results.

Specific Comments:

Comment 1:

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P1, L14: define CRV

Response: In our revised paper, we plan to change the sentence to read: "Here we quantified emission factors for CO and CH4 from a massive regional fire complex in interior Alaska during the summer of 2015 using continuous high-resolution trace gas observations from the Carbon in Arctic Reservoirs Vulnerability Experiment (CRV) tower in Fox, Alaska."

20 Comment 2:

P1, L33 - P2, L2: will these aggressively lofted emissions impact tower? Run some forward/back trajectories? Vertical mixing?

Response: To address this comment, in the methods section of the revised manuscript, we will add the following sentences: "Here we emitted fire emissions into the surface influenced volume of PWRF-

25 STILT, which extends from the surface to the top of the planetary boundary layer, with the assumption that fire emissions were equally distributed within the planetary boundary layer [*Turquety et al.*, 2007; Kahn et al., 2008]. In a previous study using the same tower, a sensitivity study revealed that plume injection height contributed only minimally to variability in simulated fire-emitted CO with PWRF-STILT [Wiggins et al., 2016]."

Comment 3:

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P2, L6: "deadly" AQ is over-simplified Response: We will change "deadly" to "unhealthy."

35 Comment 4:

2, 13: Andreae and Merlet was updated in 2019 Response: We will update all of the appropriate references to Andreae and Merlet (2001) to Andreae (2019).

40 Comment 5:

2, 18: The updated Andreae paper lists more than 20 studies, so there may be more worth including in Table 1.

Response: As described above, we will update Table 1 and corresponding text to include the missing studies of field measurements of boreal forest fire emissions from Andreae (2019), along with other studies from laboratory measurements, studies that measured emissions from land management fires, 45

and studies from Eurasian boreal forest fires that exist in the literature.

Comment 6.

2, 28: Ground-based data downwind of fires has also been collected in Selimovic et al (2019a, b) and e.g. in the Colorado front range (Gilman, Benedict) and MBO (Collier and references there-in). The Wiggins 2016 data doesn't appear in this paper anywhere that I saw.

Response: We will update this sentence to now read: "This approach has been used to estimate CO emission ratios during a moderate fire season in Alaska [*Wiggins et al.*, 2016] and for fires in other ecosystem types [*Gilman et al.*, 2015; *Collier et al.*, 2016; *Benedict et al.*, 2017; *Selimovic et al.*, 2019a,b]." We also added the fires sampled in Wiggins et al. (2016) to Table 1, using the same approach as described by our revised equation 2 and 3 to calculate emission factors.

10 **Comment 7:**

2, 33-34: Akagi et al., 2011 explain how MCE can be used to estimate an arbitrary mix of smoldering and flaming over a continuous range.

Response: We will add the following sentence on Page 2 Line 35 to include an explanation of how MCE and be used to estimate contributions from smoldering and flaming combustion: "The relative

- 15 amounts smoldering and flaming combustion are difficult to measure, but can be estimated using the modified combustion efficiency (MCE) defined as $\Delta CO_2/(\Delta CO_2 + \Delta CO)$. Fire emissions dominated by flaming combustion have an MCE up to 0.99 while emissions dominated by smoldering combustion have an MCE often between 0.65 and 0.85 (Akagi et al., 2011). MCE can be used to understand the relative contributions from both flaming and smoldering fire processes." We also changed our criteria
- 20 for separating the different combustion phases to align with previous studies.

Comment 8:

2, 37: Real fires often don't have phases - rather a dynamic mix of processes. Change "phase" to "process" throughout?

25 Response: This is a good point and we agree with the reviewer. We will change "phase" to "process" where appropriate throughout the manuscript.

Comment 9:

3,22: The tower results, even if lowered are higher than "some" airborne studies. E.g. Cofer 98 is the same. 30 If the EFs stay the same they are "a bit higher" than "some" previous estimates or recommendations.

- Response: We will revise the text to reflect our modified perspective after recomputing the emission factors. The text in section 4.2 will change to "Our emission factors for CO and CH₄ were in agreement with the mean of previous estimates for boreal fires derived from a compilation of all past studies. However, if studies that are not representative of North American boreal wildfires are excluded, including measurements from prescribed fires, laboratory studies, and studies of fires in the Eurasian
- 35 including measurements from prescribed fires, laboratory studies, and studies of fires in the Eurasian boreal forest, our emission factors are 39% higher than average emission factors derived primarily from aircraft studies of wildfires in the North American boreal forest."

Comment 10:

40 4, 6: Confusing, is it just 50 minute samples with ten minutes downtime per hour?

Response: Yes, the tower collects continuous measurements for 50 minutes out of the hour. We will clarify this point by changing the text on Page 4 line 5 to "...to separate the dataset into a set of continuous 50-minute intervals of trace gas observations..."

45 **Comment 11:**

4, 30: EFs are usually given for one species so the meaning of the ratio in the subscript here and in eqn 2 is not apparent. Suggest adopting standard notation?

Response: We will remove the ratio in the subscript of our emission factors to align with standard notion.

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Comment 12:

4, 32: Do these references unambiguously support 45% C? Revisit, consider reference above, and explain in detail in revised text.

Response: We now provide a reference to Santin et al. (2015) and add text to explain the variability can range from 45 - 50%.

Eqn (1) Sum or slope or simple subtraction?

Response: To clarify we will add the following text to Page 4 Line 30: "Excess mole fractions denoted with a Δ symbol refer to observations of trace gas mole fractions during intervals when fire had a dominant influence on tower trace gas variability with background values subtracted."

Eqn (2) Again, notation unusual, something common should work, or explain?
Response: We will change the notation to: EF_X = F_C * (1000g/kg) * MM_X/12.01 * ER_X/C_T
Where Fc refers to the carbon content of the fuel (45), MM_{CO} is the molecular mass of CO, ER_{CO} is the
emission ratio of CO relative to CO₂ and C_T = ΣN_i * ΔC_i/ΔCO₂.

Comment 13:

5, 1-5: I get computing MCE for each sample, but what's the point of the categories that don't seem to be used?

20 Response: We use the categories to separate our emission factor calculations and aid in the interpretation of our results as shown in Table 1, in Figure 4, and as discussed in section 3.1. Specifically, we use these categories to allow the reader to visually identify whether there is a trend toward one or another emissions type throughout the fire season. As shown in Figure 4, intervals with smoldering, mixed, and flaming emissions types were interspersed throughout the fire season.

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Comment 14:

Sec 2.3: There is not much detail on how AKFED is driven. One thing that stands out though is that the day/night split for fuel consumption is likely not right for 64 N! See Vermote et al, (2009); MODIS FRP can be higher at "night" than during the "day" in high latitude summer. This is relevant later.

- 30 Response: We will add the following text to Section 2.3 to better explain how AKFED is created: "AKFED burned area is mapped using perimeters from the Alaska Large Fire Database combined with imagery from the Moderate Resolution Imaging Spectroradiometer (MODIS). Both above and belowground carbon consumption are modeled based on elevation, day of burning, pre-fire tree cover, and the difference normalized burn ratio (dNBR) [Veraverbeke et al. 2015]. AKFED predicts carbon
- 35 emissions from fires with a temporal resolution of 1 day and a spatial resolution of 450 m."

We created the diurnal cycle of emissions specifically for the analysis here. We conducted additional analysis of the active fires and fire radiative power (FRP) from the MODIS fire detection products measured during the 2015 fire season in Alaska to assess our approah. The satellite data analysis

- 40 reveals that the product of total number of active fires and FRP during the daytime Terra and Aqua overpasses accounts for 83% of total fire activity (the sum of fire activity from both daytime and nighttime overpasses). This is in line with our 90% day /10% night emissions split prescribed in the model. In this context, its important to note that if there was an afternoon satellite overpass 3 hour after Aqua (at 4:30pm), it would like be higher than the 10:30am Terra overpass, because relative
- 45 humidity is lower and temperatures are considerably higher in mid-afternoon as measured from our earlier eddy covariance observation [Liu et al., 2005]. So the 83% estimate from MODIS is likely an underestimate of daytime fire activity. Vermote et al. (2009) concluded MODIS FRP can be higher at "night" than during the "day" in the boreal forest during summer, where "night" FRP is defined as the sum of FRP from both Terra overpasses and "day" is the sum of FRP from both Aqua overpasses.

However, in this analysis for the summer of 2015, we found that the sum of FRP from both Aqua overpasses was higher than the sum from both Terra overpasses.

We changed the text to provide more justification for the 90/10 emission split we used:"Analysis of the
product of fire radiative power and fire detections from the MODIS MCD14ML C6 product showed that 83% of fire activity occurred during daytime overpasses (10:30am and 1:30pm) relative to the sum across both daytime and nighttime overpasses during the 2015 Alaskan wildfire season (data not shown). The satellite observations provide broad support for the diurnal cycle we prescribed for emissions in the model."

Here is a figure showing the FRP, sum of active fires, and product of the two that illustrates how FRP and total number of active fires was considerably elevated during daytime overpasses during the 2015 fire season in Alaska. Aqua



Figure 1. Panel A shows the mean FRP (MW) normalized by area (km²) for all fires that occurred during the 2015 fire season in Alaska organized by the time and satellite of detection. B shows the total number of active fires, and C shows the product of A and B or the product of the area normalized mean FRP and the number of fires.

Comment 15:

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6, 1-9: It's likely that some weaker smoke peaks are more distorted by background variability and increase the range of values, but there is not necessarily bias.

10 Response: We agree with the reviewer and believe our methodology is strict but unbiased.

Comment 16:

6, 9: Many references support high correlation of EFCH4 with MCE.

Response: In addition to the references already listed in section 4.3 we will add the following

- 15 references to strengthen this point. The text will change to "A strong linear relationship existed between the CH₄ emission factor and MCE across the different sampling intervals (Figure 5). Linear relationships between CH₄ emission factors and MCE have also been observed in previous studies [*Yokelson et al.*, 2007; *Burling et al.*, 2011; *Van Leeuwen and van der Werf*, 2011; *Yokelson et al.*, 2013; Urbanski, 2014; *Smith et al.*, 2014; *Strand et al.*, 2016, *Guerette et al.*, 2018]. The relationship shown in
- 20 Figure 5 implies MCE can be used as a metric for CH₄ emission factors from North American boreal forest wildfires when measurements of CH₄ are not available."

Comment 17:

6, 38-39: This needs to be thought through a bit. Does the high impact of night smoke at the tower comparedto the assumed low fraction of smoke produced at night mean day smoke was under-sampled? Or does this imply AKFED underestimates night smoke?

Response: We will modify the analysis to span the same time intervals of the diurnal cycle that was applied to AKFED (0600 to 1800 for day and 1800 to 0600 for night). The text will read: "Overall, 73% of the fire emissions that impacted the tower occurred during the day (0600 to 1800 local time) and

30 27% occurred at night (1800 – 0600 local time)." AKFED has a daily resolution, but we accounted for diurnal variability in emissions by applying a diurnal cycle as explained in section 2.3. Our imposed diurnal cycle could be underestimating night smoke, or we could be measuring a slightly greater proportion of night smoke at the tower.

35 Comment 18:

7, 3: "emissions" to "consumption"

Response: We will change "emissions" to "consumption." The sentence will read: "The relative contributions of consumption from flaming and smoldering fires are uncertain for boreal forest fires...."

40 Comment 19:

7, 8: 15 total previous fires sampled may be too low if you check updated compilations. Response: We will update the total number of previous fires sampled using studies included in updated compilations.

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Comment 20:

7, 9: Convection entrains some smoldering.

Response: We changed the text in section 4.1 to the following: "... airborne sampling techniques struggle to measure emissions from less energetic smoldering combustion that emits smoke lower in 50 the atmosphere [Selimovic et al., 2019a,b]. Emissions from smoldering boreal forest fires can

sometimes be entrained in the convective columns of certain flaming fires and can be sampled by aircraft, but nighttime emissions or residual smoldering emissions from fires that have weak convective columns usually cannot [Ward and Radke, 1993; Bertschi et al., 2003; Burling et al., 2010]."

Comment 21: 5

7, 10-17: True, but a tower could potentially undersample flaming. Flaming is associated with rapid fuel consumption so not a negligible concern. Try forward trajectories from high injection altitudes to see if they impact tower or compare to column data?

- Response: We believe the tower is at an optimal location and height to sample integrated emissions from both flaming and smoldering fires. The tower is on average 295 km away from the fires we 10 sampled and located on a ridge that is over 600m above sea level. The long distance the emissions have to travel in order to reach the tower allows for mixing throughout the planetary boundary layer. Most of the fire emissions from boreal forest fires in Alaska remain in the PBL as shown by a MISR plume height analysis in Wiggins et al. (2016). To clarify, we will include the following text in section 3.2:
- "CRV tower is sufficiently downwind to integrate both flaming and smoldering processes from fires across interior Alaska."

Comment 22:

7, 19: Night may have been oversampled? But maybe not if there really is more emissions at night than was 20 assumed in AKFED? As noted above, there is sometimes more MODIS FRP at night than day in boreal regions. Also, as above, towers may not be sensitive to the entire range of injection altitudes? Explore? Response: We will add the following text to explain why the tower is not sensitive to injection altitude: "In a previous study using the same tower, a sensitivity analysis that included modifying the vertical resolution of the surface influenced volume of PWRF-STILT revealed that plume injection

25 height contributed only minimally to variability in simulated fire-emitted CO with PWRF-STILT [Wiggins et al., 2016]."

Comment 23:

7, 20-21: Quote these values from 2016 paper in Table 1?

30 Table 1 header or caption: It's enormous! Move part elsewhere. The text mentions CH4 data which I did not see in table.

Response: We will add values from Wiggins et al. (2016) and other recent studies and updates as requested by the reviewer. We will also remove the reference to CH₄ data. We will move some of the caption to table footnotes.

35 Comment 24:

7, 27: Cofer 98 agrees with this study's current values and the real average may be in middle of all this data somewhere.

Response: This section of the discussion will change to reflect a new interpretation of our corrected 40 emission factor data.

Comment 25:

7, 28-31: This data should certainly be used, but rarely does new data replace old data completely. More often new data contributes to an evolving literature average - sometimes with weighting by n factor. 45 Response: We agree with the reviewer and offer a weighted average to use instead.

Comment 26:

7, 36: You can often see smoke by satellite even when you can't detect FRP.

Response: Although this is true, FRP is often used to estimate emissions and missing detections correspond to missing emissions. We will change the original sentence "This residual smoldering 50

combustion could substantially contribute to trace gas emissions, but is difficult to detect and quantify using remote sensing because of low radiative power associated with this phase of combustion" to the following for clarity: "This residual smoldering combustion could substantially contribute to trace gas emissions but is usually excluded from FRP based fire emissions inventories because of the difficulty in detecting low FRP associated with this process of combustion."

Comment 27:

8, 3-4: To claim a difference with the studies above you would have to know proportion of above-/belowground fuel consumption that goes with those studies.

10 Response: We will revisit this discussion section following our corrected results. We cannot directly compare with the overall magnitude of emissions, but we can compare with the emission ratios measured in previous studies.

Comment 28:

- 15 8, 8: This is a common error to assume that increased EFs will lead to increased, modeled health impacts. Models use EF*biomass burned to get a-priori emissions. Then the modeled impacts are compared to downwind monitors and the a-priori emissions are adjusted to best match reality. A higher EF may change the details of the tweaking procedure, but not change the downwind PM. What would change the latter is discovering a problem with the PM monitors.
- 20 Response: We appreciate the reviewer's comment, but respectfully disagree. There has been a long standing low discrepancy between fire emissions and observed PM_{2.5} [Huang et al., 2013; Redding et al., 2016; Christopher et al., 2019; Liu et al., 2020]. Higher emission factors will require much less tweaking to the a-priori emissions by increasing the accuracy of the magnitude of the emissions.
- 25 Comment 29: 8, 10: "lead to" Response: We will change "lead" to "lead to."

Comment 30:

30 8, 17: Towers are not completely new. There was a long history of sampling prescribed fires from towers carried out by the Fire Lab.

Response: We agree with the reviewer and added the following text to clarify our approach refers to towers in the boreal forest: "Our tower-based approach to calculate emission factors has been used in other ecosystems, and is a technique that significantly improves our understanding of trace gas emissions specifically from boreal forest fires."

Reviewer #2 Comments and Responses

General comments:

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The discussion paper presents important research into the characteristics of wildfire emissions using established techniques, but novel analysis. Teasing apart the contributions of various fire events and the combustion stage (Flaming vs. smoldering) is a new and valuable way to understand nuances of boreal fire relevant to many needs, such as human health, carbon cycling, and smoke planning. However, the paper falls

45 short in many ways, and will need some extensive modification to reach its potential. I strongly suggest a refocus on a more relevant outcome from the work (rather than the fact that previous work was not catching smoldering as well as they could), a fully revised Discussion (some ideas below), and some attention to references (see notes below). This work is very important, and when presented well will make a great contribution to the literature on this subject.

Response: We appreciate the reviewers comment that this paper offers important insight into the characteristics of boreal forest fire emissions. We will systematically revise the discussion in response the the reviewer's comments and those from the other reviewers.

5 Specific comments:

Comment 1:

The title will need modification. It is unclear what "larger" refers to – larger than what? Than previous studies (yes, but I know that only when I get to the end of theAbstract). It could be larger than flaming
 combustion. The point is that having an unreferenced comparative adjective can be troublesome, especially in a title where you want to be clear. The title could be the same, but with the first four words dropped: "Contribution of . . .". Also, it is my opinion that, while this may show larger contribution than previous studies, this work has a lot of other implications and contribution that could be highlighted in the title. In some ways the community would not be too surprised to learn that the smoldering fire signal has not been
 captured in previous studies, so

highlighting this part of it is not needed to make this an impactful paper/study. Response: Our title will change to align with our updated calculations and their implications. Our new title is "Boreal forest fire CO and CH₄ emission factors from tower observations in Alaska during the extreme fire season of 2015." We chose this title to highlight that our emission factors were measured

20 during an extreme fire year using an approach that integrates emissions from fires over longer time scales than traditional aircraft based studies. We appreciate the suggestion to broaden the implications of our study and avoid highlighting undersampled smoldering fire emissions. It is likely our new emission ratios are higher than reports in previous aircraft studies, and we discuss this in the main text.

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Comment 2:

2. The comparison to previous studies would more naturally go into the discussion, rather than the introduction/background. I suggest revising to put Table 1 into the discussion where you can make the case more directly, rather than introducing the previouswork without yet seeing your results.

30 Response: We appreciate the reviewer's suggestion, and will bring the reader's attention to Table 1 in the introduction. We will add new discussion and analysis of the implications from an updated version of Table 1.

Comment 3:

35 3. There is a blatant and concerning misuse of terminology on Page 2, line 34: The sentence "Smoldering combustion can be defined as combustion with a degree of combustion completeness, or modified combustion efficiency, less than 0.9 [Urbanski2014]." First, MCE and combustion completeness (CC) are very different things. CC is the proportion of fuels consumed/combusted, while MCE is defined as the proportion of a gas to CO2. Second, the Urbanski paper puts MCE of 0.65 to 0.85 as "smoldering", and references Akagi et al. 2011 so I don't know where the 0.9 figure

comes from. The choice of the thresholds stated on page 5 lines 1-4 need to be better justified. Response: We agree with the reviewer that combustion completeness needs to be removed from the sentence. We have updated our criteria for separating the combustion processes to align with previous studies. The revised text now reads "The relative amounts smoldering and flaming combustion are

45 difficult to measure, but can be estimated using the modified combustion efficiency (MCE) defined as $\Delta CO_2/(\Delta CO_2 + \Delta CO)$. Fire emissions dominated by flaming combustion have an MCE while emissions dominated by smoldering combustion have an MCE often between 0.65 and 0.85 [Akagi et al., 2011; Urbanski et al., 2014]. MCE can be used to understand the relative contributions from both flaming and smoldering fire processes."

Comment 4:

4. I found a couple of instances where the citations used are inappropriate. While I mention only 2 here, I would suspect others, so the citations need to be fully vetted for appropriateness. First: "Rogers et al. 2015" in Page 1 line 33 is not a review of borealfire regime. It may mention this, it is not what that study provides

- 5 to the literature. Second: "Bertschi et al. 2003" in Page 7 line 34 is of laboratory experiments and work in savannah ecosystems, not boreal forest fires. In both of these cases, it could be argued that no reference is needed. If you do include a reference, it needs to be a paper or resource where the statement made is shown or studied, not where it was stated. I suggest the co-authors assist with improving the citations. Response: We thank the reviewer for pointing out our errors in citations. We will thoroughly revisit
- 10 the citations throughout the manuscript and make adjustments where necessary. We will edit the reference in Page 1 line 33 to include Johnstone et al. (2011), but prefer to keep the reference to Rogers et al. (2015) because although the primary goal of this paper was to highlight differences between the boreal fire regime in North America and Eurasia, it highlights the high energy crown fires that occur in the North American boreal forest. Rogers et al. (2015) is also used as a reference in section 4.1 of the
- 15 discussion. In the introduction we will will make the following citation changes: change McGuire et al. (2010) to Kasischke (2000), remove a reference to French et al. (2004), replace Turquety et al. (2004) with Harden et al. (2000), and add Fromm et al. (2000).

Comment 5:

20 5. The discussion would benefit from more regarding the implications of the results. What is the data showing us that is relevant? Some possible ideas to highlight/discuss

(these need to be discussed with co-authors, so are only representative):

Response: We plan to completely revise the main text of the discussion. The discussion will have a detailed discussion of the implications of our findings relative to past work summarized in Table 1.

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a. Figure 5 (Page 6 line 9) shows a linear relationship between CH4 and MCE. Provide a short discussion of this in the discussion – what does this mean for using the data? **Response:** We added the following text to the discussion: "We found a strong linear relationship between CH₄ emission factors and MCE that has also been observed in previous studies [*Yokelson et*]

- al., 2007; Burling et al., 2011; Van Leeuwen and Van Der Werf, 2011; Yokelson et al., 2013; Akagi et al., 2014; Smith et al., 2014; Urbanski et al., 2014; Strand et al., 2016, Guerette et al., 2018]. There is a wide range of slopes between CH₄ and MCE that have been found in prior studies and could be dependent on fuel type and burning conditions [Smith et al., 2014]. This implies MCE could be used as a metric for CH₄ emissions when measurements of CH₄ are not available, but care should be taken to ensure
- 35 the MCE and CH₄ relationship used is for the correct ecosystem."

b. Page 6 line 22 – ". . . attributed to boreal fire emissions." – As opposed to what?? Or why? A bit of discussion on what other factors contribute to the signal, and why there are some difference in the model will help non-atmospheric modelers better understand why these results are so powerful

Response: We changed the text to read: "The forward model simulations combining AKFED fire emissions with PWRF-STILT confirmed that the elevated CO signals at the CRV tower can be attributed primarily to boreal forest fire emissions (Figure 7), as opposed to fossil fuel or other CO emissions sources. The AKFED model had a Pearson's correlation coefficient of 0.61 with observed

- 45 daily mean CO and had a low bias of approximately 7%. Differences between the model simulations and observations were likely caused by errors in the magnitude and timing of fire emissions within AKFED as well as the limited spatial resolution and incomplete representation of atmospheric transport within PWRF-STILT."
- 50 c. The temporal distribution data (Fig 10) is very interesting and could be helpful for

exposure assessment for health studies. (although PM, rather than CO would be of interest).

Response: We added the following sentence: "The timing of emissions is important for quantifying the impact on human health, and enhanced nighttime emissions (Figure 10) when the boundary layer is much lower could increase surface concentrations and exacerbate negative health effects."

d. Page 7, line 24: I am not sure I see a temporal trend in the old data, and I am not sure why this would be something to note. This statement is best dropped. Table 1 presents past results that are collected in a variety of settings, so (in my assessment)

10 represents some data on the range of variability, not a record of change over time. I hope these comments inspire the authors to revise the manuscript for a more useful. Response: We appreciate the insight offered by the reviewer to improve our discussion. We agree the implications need to be revisited and the discussion will be significantly revised based on our corrected emission factor calculations and themes suggested by the reviewer.

Reviewer #3 Comments and Responses

General comments:

- 20 The authors present an impressive set of CO, CH4 and CO2 measurements in boreal forest fire smoke to calculate emission factors for CO and CH4. Such accurate data are neccesarry to soundly test models used to quantify the impact of big fires on the air quality and climate. Therefore, the paper is highly suitable to be published in this journal. Yet, due to error in the emission factors calculation I suggest a revision of this manuscript. Re-interpretation of results should be done by the authors before resubmission. Firstly, Eq 2 on Page 4 should be revised, using e.g. Eq. 1 and 2 in
- 25 Yokelson et al. I'm looking forward to review the revised manuscript in detail. I add here only a potential useful comment. Are there any other measurements of specific tracers to be used to quantify the smoldering/flaming contributions? Flaming is likely under-represented in the used sampling height. Response: We appreciate the reviewer's comment that our study provides accurate CO and CH4 emission factor data needed to quantify the impact of boreal fires on air quality and climate. We agree
- 20 that the manuscript will need revision and reinterpretation of results to reflect the corrected emission factor calculations. We have added text in section 2.1 and 3.2 to explain why the tower is an ideal location to measure emissions from both smoldering and flaming fires. The text in section 2.1 reads "In a previous study using the same tower, the authors conducted a sensitivity study on the CRV tower and found little influence of plume injection height on CRV tower trace gas observations [Wiggins et]
- 35 al., 2016]." The text in section 3.2 reads "CRV tower is sufficiently downwind to integrate both flaming and smoldering processes from fires across interior Alaska."

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Boreal forest fire <u>CO and CH₄ emission factors derived</u> from tower observations in Alaska <u>during the extreme fire season of 2015</u>

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- 10 Abstract. With recent increases in burned area within boreal forests that have been linked to climate warming, there is a need to better understand the composition of emissions and their impact on atmospheric composition. Most previous studies have estimated boreal fire emission factors from daytime samples collected by aircraft near fire plumes or at the surface near actively burning fires. Here we quantified emission factors for CO and CH₄ from a massive regional fire complex in interior Alaska during the summer of 2015 using continuous high-resolution trace gas observations from the Carbon in Arctic Reservoirs Vulnerability
- 15 Experiment (CRV) tower in Fox, Alaska, Averaged over the 2015 fire season, the CO/CO₂ emission ratio was 0.142,± 0.051, and the CO emission factor was 127 ± 40 g CO per kg of dry biomass consumed. The CO/CO₂ emission ratio was about 39% higher than the mean of previous aircraft estimates for fresh emissions from boreal North America wildfires. The mean CH₄/CO₂ emission ratio was 0.010 ± 0.004 and the CH₄ emission factor was 5.3 ± 1.8 g CH₄ per kg of dry biomass consumed, with a mean similar to previous reports. CO and CH₄ emission factors varied in synchrony, with higher CH₄ emission factors observed during periods
- 20 with lower modified combustion efficiency (MCE). By coupling a fire emissions inventory with an atmospheric model, we identified that at least 35 individual fires contributed to trace gas variations measured at the CRV tower, representing a <u>sample size</u> that is nearly the same as the total number of boreal fires measured in all previous <u>field campaigns</u>. The model also indicated that typical mean transit times between trace gas emission and tower measurement were 1-3 days, indicating that the time series sampled combustion across day and night burning phases. The high and variable CO emission factor estimates reported here provide
- 25 evidence for a more prominent role of smoldering combustion, highlighting the importance of continuously sampling of fires across time-varying environmental conditions that are representative of typical burning conditions.

1 Introduction

Boreal forest fires influence the global carbon cycle and climate system through a variety of pathways. Boreal forest fires initiate succession, influence landscape patterns of carbon accumulation, and directly release carbon dioxide and other trace gases into the atmosphere [*Johnson*, 1996]. One of the largest reservoirs of global terrestrial carbon resides in organic soils underlying boreal forests [*Apps et al.*, 1993; *Kasischke*, 2010], and fires in the boreal forest can consume significant amounts of aboveground and belowground biomass [*Harden et al.*, 2000; *French et al.*, 2004; *Boby et al.*, 2010; *Walker et al.*, 2018]. Many boreal forest fires are stand replacing and high energy [*Johnstone et al.*, 2011; *Rogers et al.*, 2015], with enough convective power to inject smoke into the upper troposphere and lower stratosphere where it can be transported across the Northern Hemisphere [*Fromm et al.*, 2000, Fromm et al., 2010].

35 <u>al., 2000;</u> Forster et al., 2001; Turquety et al., 2007; Peterson et al., 2018].

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Emissions from boreal fires can significantly influence atmospheric composition throughout the Northern Hemisphere. Fire plumes from regional fire complexes in Alaska and western Canada, for example, have been shown to influence air quality over Nova Scotia [*Duck et al.*, 2007] and across the south-central US [*Wotawa et al.*, 2001; *Kasischke et al.*, 2005] and Europe [*Forster et al.*, 2001]. Similarly, emissions, from boreal forest fires in Russia, have caused unhealthy, air quality in Moscow

- 5 [Konovalov et al., 2011] and have affected ozone and other trace gases concentrations across the western US [Jaffe et al., 2004]. Over the past few decades, annual burned area in several regions in boreal North America has increased [Gillett et al., 2004; Kasischke and Turetsky, 2006; Veraverbeke et al., 2017], and future projections suggest further increases may occur in response to changes in fire weather and a lengthening of the fire season [Flannigan et al., 2001; de Groot et al., 2013; Young et al., 2017]. As a consequence, fires are likely to play an increasingly important role in regulating air quality and climate feedbacks during the remainder of the 21st century.
 - Emission factors provide a straightforward way to convert fire consumption of dry biomass into specific trace gas species, such as CO, CH₄, and CO₂. This technique is commonly used to model emissions of select species and or to compare model results with in-situ or remotely sensed observations. The most frequently used boreal forest fire emission factors are derived from metaanalyses that average together information from individual field campaigns [Andreae and Merlet, 2001; Akagi et al., 2011;
- 15 <u>Andreae</u>, 2019]. These syntheses often include in-situ airborne and ground based measurements along with laboratory measurements of combusted fuels. There is no consensus on how to combine information from different studies, and in past work individual studies have been given equal weight when estimating biome-level means, even when the number of fires and duration of sampling has varied considerably from one field campaign to another. A summary of previous studies that measured CO emission ratios for poreal forest fires is shown in Table 1.
- 20 In past work, the most common approach for measuring emission factors from boreal fires is to fly aircraft near or withinplumes, measuring trace gases using infrared gas analyzers mounted in the aircraft or by collecting flasks of air that are measured later in the laboratory. Over a period of more than 25 years, a total of <u>39</u> boreal fires have been sampled by aircraft, including <u>19</u> wildfires and <u>14</u> prescribed <u>land management</u> fires from boreal North America and 6 prescribed fires in Siberia (Table 1). Aircraft sampling is a highly effective approach for sampling large and remote wildfires, especially for characterizing reactive trace gas
- emissions that have lifetimes of hours to days. It also important to recognize potential limits associated with sampling fires in this way. Aircraft observations are mostly confined to periods with good visibility, often sampling well-developed fire plumes during mid-day and during periods with relatively low cloud cover. These conditions represent a subset of the environmental variability that a large wildland fire may experience in boreal forest ecosystems as it burns over a period of weeks to months. An alternative approach for measuring in-situ emission factors involves using a surface tower that continuously samples trace gas concentrations.
- [Wiggins et al., 2016] and for fires in other ecosystem types [Collier et al., 2016; Benedict et al., 2017; Selimovic et al., 2019a,b].

_Environmental conditions, including weather, vegetation, and edaphic conditions are known to influence the composition of emissions, in part by regulating the importance of flaming and smoldering combustion processes, [Ward and Radke, 1993;
 35 Yokelson et al., 1997; Akagi et al., 2011; Urbanski, 2014]. The relative amounts smoldering and flaming combustion are difficult to measure, but can be estimated using the modified combustion efficiency (MCE) defined as ΔCO₂/(ΔCO₂ + ΔCO), where the Δ notation denotes the fire-associated dry air mole fraction of a sample gas after background levels have been removed. Fire emissions dominated by flaming combustion have an MCE fromfrom 0.92 – 1.0, while emissions dominated by smoldering combustion have an MCE flaming et al., 2011; Urbanski, 2014]. MCE can be used to understand the
 40 relative contribution of flaming and smoldering combustion processes to the composition of trace gases and aerosols in air

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measured downwind of a fire. Flaming combustion is more efficient at oxidizing organic matter directly to CO₂ gas than smoldering combustion, and as a consequence, smoldering combustion produces more CO, CH₄, and organic carbon aerosol [*Ward and Radke*, 1993; *Urbanski et al.*, 2008]. Smoldering combustion can be defined as combustion with a modified combustion efficiency, less than 0.85, [*Urbanski*, 2014]. Flaming combustion requires the presence of organic material that burns efficiently in a high oxygen

- 5 environment [*Ryan et al.*, 2002], and often occurs in boreal forests when fires consume dry aboveground fuels, including vegetation components with low moisture content, litter, and fine woody debris [*French et al.*, 2004]. Smoldering, in contrast, is a dominant combustion process, for burning of belowground biomass and larger coarse woody debris. Residual smoldering combustion in boreal forests can continue to occur for weeks after a flaming fire front has passed through, especially in peatland areas with carbon rich organic soils [*Harden et al.*, 2000; *Bertschi et al.*, 2003], Over the lifetime of a large fire, smoldering combustion is more likely to occur during periods with lower temperatures and higher atmospheric humidity that increases the moisture content of fine
- fuels [*Stocks et al.*, 2001; *Ryan*, 2002].

Here we used trace gas observations of CO, CH₄, and CO₂ from the CRV tower to estimate emission factors from boreal forest fires that burned during the near-record high Alaska fire season of 2015. The summer of 2015 was the second largest fire season in terms of burned area since records began in 1940 with about 2.1 million hectares burned [*Hayasaka et al.*, 2016; *Partain*

- 15 et al., 2016]. An unseasonably warm spring and early snowmelt allowed fuels to dry early in the season [Partain et al., 2016]. In mid-June, thunderstorms caused an unprecedented number of lightning strikes (over 65,000) that ignited over 270 individual fires on anomalously dry fuel beds over the course of a week [Hayasaka et al., 2016; Veraverbeke et al., 2017]. Fires expanded rapidly during several hot and dry periods through mid-July, and then slowed down as multiple precipitation events and cool, damp weather minimized fire growth for the rest of the summer fire season.
- 20 The CRV tower captured an integrated signal of trace gas emissions from <u>multiple</u> fires across interior Alaska during the 2015 fire season [*Karion et al.*, 2016]. The data stream was comprised of continuous sampling from June 15 August 15 with more than 5858,000, samples, each with a 30 s duration. The CRV tower experienced enhanced and highly correlated CO, CH₄, and CO₂ trace gas signals from fires for about 7% of the duration of the fire season. We identified events when fire emissions had a dominant influence on trace gas variability at CRV tower and used these events to derive emission factors. <u>Analysis of these data</u>
- 25 indicate that smoldering processes may have a higher contribution to total wildfire emissions, from North American boreal forests than previous estimates derived from aircraft measurements. To quantify the spatial and temporal variability of individual fires and their influence on CO, CH4, and CO2 at the CRV tower, we coupled a fire emissions inventory, the Alaska Fire Emissions Database (AKFED) [Veraverbeke et al., 2015] with an atmospheric transport model, the Polar Weather Research and Forecasting Stochastic Time Integrated Lagrangian Transport (PWRF-STILT) model [Henderson et al., 2015]. This modeling analysis further revealed
- 30 that the number of 2015 wildfires sampled with our approach is comparable to the total number of North American boreal forest, fires sampled in past work.

2 Methods

2.1 CARVE (CRV) Tower Observations

Atmospheric CO, CH₄, and CO₂ mole fractions were measured using a cavity ring-down spectrometer (CRDS, Picarro models 2401 and 2401m) [*Karion et al.*, 2016] at the CRV tower in Fox, Alaska (64.986°N, 147.598°W, ground elevation 611m above sea level). The tower is located about 20 km northeast of Fairbanks Alaska <u>on top of a hill in hilly terrain</u> (Figure 1), and within the interior forested ecoregion in interior Alaska [*Cooper et al.*, 2006]. There are three separate inlets on CRV tower at different heights above ground level from which the spectrometer draws sample air. The spectrometer samples air from the highest

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level for 50 minutes out of every hour, and then draws air from the other levels for 5 minutes at each level [Karion et al., 2016]. The data stream from this spectrometer has gaps every 50 minutes as the spectrometer cycles to the lower inlets. We used observations from air drawn from the top intake height at a height of 32 m above ground level because this level had the highest measurement density and the smallest sensitivity to local ecosystem CO2 fluxes near the tower [Karion et al., 2016]. All raw 30 s average measurements were calibrated according to Karion et al. [2016]. Each 30 s average measurement served as an individual

point in our calculation of emission factors described below.

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2.2 Emission Factors and Modified Combustion Efficiency

We isolated intervals when fire had a dominant influence on trace gas variability observed at CRV tower to calculate emission factors. An interval with dominant fire influence was defined as a continuous period that had: 1) a minimum of at least thirty trace gas measurements (with each measurement representing a mean over 30 seconds), 2) a mean CO over the entire interval exceeding 0.5 ppm, and 3) significant correlations between CO and CO2, and CH4 and CO2, with r² values for both relationships exceeding 0.80.

We used the gaps in the data stream when the spectrometer sampled air from the lower levels to separate the dataset into a set of continuous 50-minute intervals of trace gas observations with less than 15 s between each new 30 s averaged measurement 15 and by applying a minimum sampling size, criterion of at least 30 measurements. We calculated the mean CO mole fraction for each interval and removed all intervals with a mean CO less than 0.5 ppm. For each interval with high levels of CO, we then extracted CO, CH₄, and CO₂ mole fractions and calculated correlation coefficients between all three gases. Only periods with high and significant correlations between CO:CO₂ and CH₄:CO₂ ($r^2 > 0.80$; p < 0.01, n > 30) were retained, because covariance among these co-emitted species is a typical signature of fire emissions [Urbanski, 2014].

- 20 We calculated background mole fractions of CO and CH4 by taking an average of observations prior to any major fire activity in interior Alaska during DOY 170-172.5. This yielded a CO background of 0.110 ppm and a CH₄ background of 1.90 ppm. We modeled hourly CO2 background concentrations to account for the influence of net ecosystem exchange (NEE) using a Bayesian approach multi-variable linear regression model trained on CRV tower observations during 2012, a year with little to no fire influence on trace gas variability. We assumed negligible influence from fossil fuel combustion on background mole fraction
- 25 variability, The hourly CO_2 model was linearly interpolated to have the same temporal resolution as the CRV tower data. The variables used in the CO₂ model include hourly observations of temperature, vapor pressure deficit, precipitation, day of year, latent heat flux, and hourly CO2 observations from Barrow, AK (Figure 2). Meteorological variables were acquired from the National Climatic Data Center Automated Weather Observing System for Fairbanks International Airport (http://www7.ncdc. noaa.gov/CDO/cdopoemain.cmd). This location was chosen due to its proximity to the CRV tower. We obtained 3-hourly latent
- heat flux from the NOAH2.7.1 GLDAS/NOAH experiment 001 for version 2 of the Global Land Data Assimilation System 30 (GLDAS-2) [Rodell et al., 2015]. Hourly in-situ CO2 observations from a clean air site at Barrow, AK were attained from the Earth System Research Laboratory Global Monitoring Division [Thoning et al., 2007]. In a sensitivity analysis we found that the removal of the background had only a small effect, because the background did not change appreciably during the duration of each 50minute time interval used to compute an emission factor.
- 35 We estimated emission ratios (*ER*) by calculating the slope from a type II linear regression of CO and CH₄ excess mole fractions (ΔX) relative to CO₂ (Δ CO₂) (Equation 1). Excess mole fractions denoted with a Δ symbol refer to observations of trace gas mole fractions during intervals when fire had a dominant influence on tower trace gas variability with background values subtracted. Emission factors (EF_{λ}) were calculated using equation 2, where F_{C} is the mass fraction of carbon in dry biomass, MM_{x} is the molar mass of CO or CH₄, 12.01 is the molar mass of carbon, ER_x is the emission ratio, and C_T is given by equation 3, where

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n is the number of carbon containing species measured, N_i is the number of carbon atoms in species *i*, and ΔC_i is the excess mole fraction of species *i* [*Yokelson et al.*, 1999; *Akagi et al.*, 2011] (Equation 2). Here we computed C_T by allowing *i* in equation 3 to cycle over CO₂, CO, and CH₄ (n = 3). We assumed the fraction of carbon in combusted fuels, F_C , was 0.45 [*Santin et al.*, 2015], but note that F_C can range from 0.45 – 0.55 [*Akagi et al.*, 2011].

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$$ER_X = \frac{\Delta X}{\Delta CO_2} = \frac{X_{Fire} - X_{Background}}{CO_2 Fire - CO_2 Background}$$

$$EF_{X} = \frac{MM_{X}}{12.01} * F_{C} * \frac{1000g}{kg} * \frac{ER_{X}}{C_{T}}$$

$$C_{T} = \sum_{i=1}^{n} N_{i} * \frac{\Delta C_{i}}{\Delta CO_{2}}$$
(3)

We calculated the modified combustion efficiency (MCE) for each emission factor interval. Modified combustion efficiency is defined as the excess mole fraction of CO₂ divided by the sum of the excess mole fractions of CO and CO₂ [*Ward and Radke*, 1993]. MCE was used to separate events into three categories: smoldering, mixed, or flaming. These categories reflect the dominant combustion process contributing to trace gas anomalies at the CRV tower during the summer of 2015. Periods with an MCE less than 0.85 were considered to consist of mostly smoldering combustion, periods with a MCE of greater than or equal to 0.85 and less than 0.92 were classified as consisting of a mixture of smoldering and flaming combustion, and period with an MCE greater than 0.92 were classified as flaming [*Urbanski*, 2014]. We performed this classification to allow for a visualization of how combustion processes varied from interval to interval (and day to day) during the 2015 fire season.

2.3 Transport Modeling

We coupled a fire emission model, the Alaskan Fire Emissions Database (AKFED) [Veraverbeke et al., 2015] with an atmospheric transport model, the Polar Weather Research and Forecasting Stochastic Time Integrated Lagrangian Transport model (PWRF-STILT) [Henderson et al., 2015] to estimate fire contribution to trace gas variability from CRV tower observations

- 20 following Wiggins et al. [2016]. Here we emitted fire emissions into the surface influenced volume of PWRF-STILT, which extends from the surface to the top of the planetary boundary layer, with the assumption that fire emissions were equally distributed within the planetary boundary layer [*Turquety et al.*, 2007; *Kahn et al.*, 2008]. In a previous study using the same tower, a sensitivity study revealed that plume injection height contributed only minimally to variability in simulated fire-emitted CO with PWRF-STILT [*Wiggins et al.*, 2016]. Daily burned area from AKFED is mapped using thermal imagery from the Moderate Resolution
- 25 Imaging Spectroradiometer (MODIS) within fire perimeters from the Alaska Large Fire Database. Both above and belowground carbon consumption is modeled based on elevation, day of burning, pre-fire tree cover, and difference normalized burn ratio (dNBR) derived from 500m MODIS surface reflectance bands [Veraverbeke et al., 2015]. AKFED predicts carbon emissions from fires with a temporal resolution of 1 day and a spatial resolution of 450 m. We regridded AKFED to the same spatial resolution as the atmospheric transport model (0.5°) for the model coupling. To account for diurnal variability in emissions, here we imposed a
- 30 diurnal cycle on daily emissions following *Kaiser et al.* [2009], where the diurnal cycle is the sum of a constant and a Gaussian function that peaks in early afternoon with 90% of emissions occurring during the day (hours 0600 to 1800) and 10% at night (hours 1800 to 0600). <u>Analysis of the product of fire radiative power and the number of fire detections from the MODIS MCD14ML C6 product showed that 83% of fire activity occurred during the daytime overpasses (10:30am and 1:30pm) relative to the sum across both daytime and nighttime overpasses during the 2015 Alaskan wildfire season (data not shown). The satellite 35 observations, although temporally sparse (with only 4 over passes per day), are consistent with the diurnal cycle we prescribed for</u>

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fire emissions in the model. We convolved AKFED with the PWRF-STILT footprints to determine individual fire contributions to CO anomalies at CRV tower. This was achieved by calculating the total CO contribution from each individual 0.5° grid cell from the AKFED × PWRF-STILT combined model and utilizing the fire perimeters from the Alaska Large Fire Database (data provided by Bureau of Land Management (BLM) Alaska Fire Service, on behalf of the Alaska Wildland Fire Coordinating Group (AWFCG)

- 5 and Alaska Interagency Coordination Center (AICC)) to identify the location of individual fires. AKFED uses the same fire perimeter database for burned area and carbon emissions estimates [Veraverbeke et al., 2015]. We determined an individual fire's contribution to CO at the CRV tower by setting all emissions in AKFED for a particular grid cell to zero and rerunning the model coupling with PWRF-STILT. The difference between the original model and the updated coupling is equal to an individual fire's contribution to CO at the CRV tower. Due to the 0.5° grid cell size used for model coupling, more than one fire perimeter existed
- 10 in some individual grid cells. In these cases, the contribution for each fire was determined by weighting the total signal contribution by fire size.

We also used the influence functions or "footprints" (ppm per μ mol/m²/s) from the atmospheric model to quantify the contribution of day and night emissions and mean transport times between the point of emission and measurement at the CRV tower. The footprints are on a 0.5° latitude-longitude grid with a temporal resolution of 1 h during hours 0600 to 1800 (day) local

- 15 time and 3 h during hours 1800 to 0600 (night) and provide an estimate of the impact of upwind surface fluxes on CRV tower trace gas measurements at a given time. We analyzed the footprints for each time period associated with an emission factor period to confirm CRV tower observations represented an integration of emissions from multiple fires and captured variability in emissions across the diurnal fire cycle. <u>The footprints associated with each emission factor event also were used to determine how much of</u> the signal was coming from burning on previous days and the fraction of emissions emitted during day and night periods. We
- 20 found that 99% of the fire emissions that influenced CRV tower trace gas concentrations occurred within 3 days of the sampling interval used to derive the emission factor for an individual event at the CRV tower, with 76% occurring within the first 24 hours, 21% during the next 24 hours, and 2% occurring three days prior to the event (Figure 10). Overall, 73% of the fire emissions that impacted the tower occurred during the day (0600 to 1800 local time) and 27% occurred at night (1800 0600 local time).

3 Results

25 3.1 Emission Factors and Modified Combustion Efficiency

During the 2015 Alaska fire season, we observed synchronized enhancements of CO, CH₄, and CO₂ well above background concentrations in CRV tower observations from DOY 173 – 196 (Figure 3). We identified <u>55</u>, individual events that each span about 50 minutes each to calculate emission factors from the elevated trace gas observations (Figure 4; Table 2). CO/CO₂ emission ratios ranged from 0.025 to 0.272 and CH₄/CO₂ emission ratios ranged from 0.002 to 0.020. MCE ranged from 0.786 to

- 30 0.975 (Table 2). CO emission factors ranged from 25 to 223 g CO per kg biomass combusted, and CH4 emission factors ranged from 1.18 to 10.7 g CH4 per kg biomass combusted. The mean CO/CO2 emission ratio was 0.141 ± 0.051 the mean CO emission factor was 127 ± 40 g CO per kg biomass combusted, and the mean MCE was 0.878 ± 0.039. Concurrently, the mean CH4/CO2 emission ratio was 0.010 ± 0.004 and the mean CH4 emission factor was 5.32 ± 1.82 g CH4 per kg biomass combusted.
- _____A strong linear relationship existed between the CH4 emission factor and MCE across the different sampling intervals
 (Figure 5). Linear relationships between CH4 emission factors and MCE have also been observed in previous studies [*Yokelson et al.*, 2007; *Burling et al.*, 2011; *Van Leeuwen and van der Werf*, 2011; *Yokelson et al.*, 2013; *Urbanski*, 2014; *Smith et al.*, 2014;

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Strand et al., 2016, Guerette et al., 2018]. The relationship shown in Figure 5 implies MCE can be used as a metric for CH_4 emission factors from North American boreal forest wildfires when measurements of CH_4 are not available.

We classified each fire-affected sampling interval as being dominated by smoldering, mixed, or flaming combustion processes using thresholds on MCE. This analysis revealed that intervals with different combustion phases were interspersed

- 5 throughout the fire season, with no clear progression over time, or clustering of flaming or smoldering processes during periods with high or low levels of burning. We identified 12 smoldering events, 37 mixed events, and 6 flaming events throughout the fire season (Figure 4, within examples shown in Figure 6 and summarized Table 3). Smoldering events had a mean CO/CO₂ ratio of 0.214±0.030, a mean CO emission factor of 183±21 g CO per kg biomass combusted, a mean CH₄/CO₂ ratio of 0.014±0.003, a mean CH₄ emission factor of 6.89 ± 1.18 g CH₄ per kg biomass combusted, and a mean MCE of 0.824±0.020. Mixed events
- 10 consisting of both smoldering and flaming combustion had a mean CO/CO₂ emission ratio of 0.13 L ± 0.024, a mean CO emission factor of 120 ± 20 g CO per kg biomass combusted, a mean CH₄/CO₂ emission ratio of 0.010 ± 0.003, a mean CH₄ emission factor of 5.28 ± 1.5 Lg CH₄ per kg biomass combusted, and a mean MCE of 0.884 ± 0.019. Flaming events had a mean CO/CO₂ emission ratio of 0.060 ± 0.020, a mean CO emission factor of 59 ± 19 g CO per kg biomass combusted, a mean CH₄/CO₂ emission ratio of 0.060 ± 0.020, a mean CO emission factor of 2.49 ± 0.78 g CH₄ per kg biomass combusted, and a mean MCE of 0.944 ± 0.018
 15 (Table 3).

In our primary analysis described above, each individual fire event was weighted equally in computing a season-wide mean. As a sensitivity analysis, we computed the mean emission ratios weighting each event according to its mean ΔCO mole fraction, and, alternately, according to its mean ΔCO_2 mole fraction. Weighting by CO caused the CO emission ratio to increase from 0.141 to 0.146 but the CH4 emission ratio did not change. Weighting by CO₂ caused the emission ratios to slightly increase

20 yielding a CO emission ratio of 0.144 and the same CH₄ emission ratio of 0.010. The variation revealed by this analysis highlights the challenge of combining information from different individual fires, and the importance of moving toward flux-weighted estimates in future work.

3.2 The Influence of Individual Fires on Trace Gas Variability at the CRV Tower

The forward model simulations combining AKFED fire emissions with PWRF-STILT confirmed that the elevated CO
 signals at the CRV tower can be attributed primarily to boreal forest fire emissions (Figure 7) and not to fossil fuels or other CO sources, The AKFED model had a Pearson's correlation coefficient of 0.61 with observed daily mean CO and had a low bias of approximately 7%. Differences between the model simulations and observations were likelycaused by errors in the magnitude and timing of fire emissions within AKFED as well as the limited spatial resolution and incomplete representation of atmospheric transport within PWRF-STILT. Nevertheless, the broad agreement between the model and the observations, including the timing of the large burning event between DOY 173 and 179, provides some confidence that our model can be used to explore the influence

and contribution of individual fires.

We identified 35 individual fires that contributed to at least 1% of the CO mole fraction time series at CRV tower (Figure 8; Figure 9; Table 3). On average, these fires were 295 ± 131 km away from CRV tower, located mostly to the west of Fairbanks, in the direction of the prevailing summer surface winds. This analysis revealed that the CRV tower was sufficiently downwind to

35 integrate emissions from multiple fires through the full planetary boundary layer and across several day-night cycles. The total CO emitted from these fires accounted for 75% of the excess CO mole fraction signal during DOY 160 – 200. The remaining CO signal originated from many smaller fires that were widely distributed across interior Alaska. The Tozitna fire was responsible for the greatest percentage of the total CO anomaly integrated over the 2015 fire season at the CRV tower (accounting for 8% of the integrated, CO anomaly, at CRV), The fires that significantly contributed the most to the CO anomaly at CRV tower were not account of the season at the CRV tower were not account of the season at the CRV tower were not account of the season at the CRV tower were not account of the season at the CRV tower were not account of the season at the CRV tower were not account of the count of the season at the CRV tower were not account of the count of the count of the count of the count of the season at the CRV tower were not account of the count of the c

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necessarily the closest fires to the tower or the largest fires of the 2015 fire season in terms of burned area. Combined, however, this set of 35 fires accounted for 0.97 Mha, or approximately $\frac{46\%}{100}$ of the total burned area reported during the 2015 fire season [Veraverbeke et al., 2017].

4 Discussion

5 4.1 <u>Measurement technique</u>, and <u>ecosystem type as drivers of variability in boreal forest fire, emission ratios</u>,

The most widely used emission factors for boreal forest fires are derived from syntheses that average together data from individual field campaigns [*Andreae and Merlet*, 2001; *Akagi et al.*, 2011; *Andreae*, 2019]. In order to investigate the possible influence of sampling strategy employed by previous studies, and variations caused by ecosystem type, we compiled available studies that report CO emission ratios for boreal forest fires and organized the studies into several categories with common

- 10 characteristics, including aircraft sampling of North American boreal forest wildfires, aircraft sampling of North American boreal forest management or prescribed fires, combustion of North American boreal forest fuels measured in the laboratory, and sampling of Siberian boreal fires from both aircraft and surface platforms (Table 1). All previous studies combined have sampled a total of 39 individual boreal forest fires for CO emission ratios, and additional measurements have been made by combusting fuels in a laboratory setting. We found several important differences in emission ratios that may be linked with the measurement technique
- 15 and ecosystem type. First, solely considering emission ratio measurements from boreal North America, our surface tower measurements of about 35 fires, along with earlier tower measurements from Wiggins et al. [2016] have a considerably higher mean (0.141) than the mean of aircraft measurements sampling wildfires (0.102) or management and prescribed fires (0.077). We believe these differences are linked, in part, with sampling strategy. Aircraft-based studies often sample fires that have a strong contribution
- 20 from flaming combustion, which releases enough energy to generate well-defined plumes at an altitude accessible by the aircraft. This methodology provides an opportunity to comprehensively measure the vertical and horizontal distribution of emissions from an individual fire and their atmospheric evolution in a smoke plume. However, airborne sampling techniques are often limited to daytime periods with good visibility, making it difficult to comprehensively measure emissions over a diurnal cycle or over the full lifetime of a fire which may span several periods with inclement weather. Due to these sampling constraints, aircraft studies
- 25 are less likely to measure emissions from less energetic smoldering combustion, since these emissions are more likely to remain near the surface [*Ward and Radke*, 1993; *Selimovic et al.*, 2019a]. Emissions from smoldering boreal forest fires can sometimes be entrained in the convective columns of certain flaming fires and can be sampled by aircraft, but nighttime emissions or residual smoldering emissions from fires that have weak convective columns usually cannot be measured in this way [*Bertschi et al.*, 2003; *Burling et al.*, 2012]. Near the end of the lifetime of a long-lived fire, aircraft measurements have sometimes observed a larger
- 30 smoldering to flaming ratio [Yates et al., 2016]. A few previous studies have investigated the differences in emissions measurements from ground and aircraft sampling of the same fire, reporting significant differences between the relative abundance of the emissions observed depending on the sampling method [Christian et al., 2007; Burling et al., 2011; Akagi et al., 2014]. Emission ratios derived from aircraft measurements are more likely to sample fires during times when flaming combustion processes are dominant [Babbitt et al., 1996, Akagi et al., 2014], yet rarely sample residual smoldering combustion that can

35 substantially contribute to emissions over the full lifetime of an individual fire [Bertschi et al., 2003].
Second, we also separated aircraft-based studies that measured emissions from wildfires from those that measured emissions from prescribed slash and land management fires, where trees are bulldozed, dried and intentionally arranged to promote

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Deleted: → The footprints associated with each emission factor event also were used to determine how much of the signal was coming from burning on previous days and the fraction of emissions emitted during day and night periods. We found that 99% of the fire emissions that influenced CRV tower trace gas concentrations occurred within 3 days of the sampling interval used to derive the emission factor for an individual event at the CRV tower, with 76% occurring within the first 24 hours, 21% during the next 24 hours, and 2% occurring three days prior to the event (Figure 10). Overall, 64% of the fire emissions that impacted the tower occurred during the day (0900 to 1800 local time) and 36% occurred at night (1900 − 0600 local time). ¶

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Deleted: → Emission factors provide a straightforward way to convert fire emissions of dry biomass into specific trace gas species, such as CO, CH₄, and CO₂. This technique is commonly used to model emissions of select species and or to compare model results with in-situ or remotely sensed observations. There are limited previous studies on boreal forest fire emission factors, and almost all derived emission factors were determined from aircraft sampling (Table 1) [Cofer et al., 1990; Radke et al., 1991; Nance et al., 1993; Cofer et al., 1998; Goode et al., 2000; Simpson et al., 2011]. In total, all previous aircraft-based studies combined sampled 15 individual prescribed and/or wildfires and derived emission factors that were likely most representative of flaming fires that occurred in the afternoon and were strong enough to generate well-defined plumes. Our emission factors for CO were 35% higher than previous estimates for wildfires derived from the aircraft measurements. We believe a primary contributor to this difference is sampling methodology. Our CRV tower-based sampling was able to integrate over day-night burning cycles, flaming combustion at active fire fronts as well as residual smoldering combustion in soils that persists for days behind the fire line, and emissions associated with a wide range of environmental conditions that occurred during 2015 fire season. This integration was possible because the tower was located several hundred kilometers downwind of the core fire complex located western Alaska.

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Deleted: (Deleted:) maximum fuel consumption [*Cofer et al.*, 1990; *Cofer et al.*, 1998]. Land management fires consume dried aboveground fuels with a different fuel structure and moisture content than fuels consumed in a wildfire, where combustion from soil organic material layers is a dominant component of bulk emissions [Boby et al., 2010; *Dieleman et al.*, 2020], Although the number of land management fires is relatively small, the mean from these studies suggest flaming processes are a more important contributor to

- 5 this fire type than for wildfires, and some consideration of this difference should be factored into regional and global syntheses. Third, three studies report emission ratios from laboratory combustion of fuels collected from North American boreal forests including biomass samples from black spruce, white spruce, and jack pine, as well as moss and surface organic material (duff). The laboratory studies have considerable variability that can be attributed to the type of fuel combusted and fuel moisture content. This work indicates duff consumption yields much higher emission ratios for CO and CH4 than combustion of black spruce
- or jack pine needles and other fine fuels [*Bertschi et al.*, 2003; *Mcmeeking et al.*, 2009]. The fuels used in laboratory studies are usually dried and burned individually, although some studies have attempted to mimic natural fires by placing dried fine fuels on top of damp fuels that undergo residual smoldering combustion [*Bertschi et al.*, 2003]. The structure, composition, and moisture content of fuels are well known as key drivers of the composition and magnitude of emissions. Although these laboratory studies provide valuable information on emissions from individual fuel components, they are not able to capture the full complexity of a wildfire.

Fourth, emission factors from the Siberian boreal forest are often grouped together with emission factors from North American boreal forest in biome-level syntheses [*Andreae*, 2019]. Yet, Table 1 shows emission ratios from wildfires in boreal Siberia tend to be higher than emission ratios from North American wildfires. Although more measurements are needed, higher CO emission ratios for Siberian fires appears consistent with past work showing that boreal fire behavior is fundamentally different

- 20 between North American and Siberian continents as a consequence of differences in tree species and their impacts on fire dynamics. Notably, as consequence of the presence of black spruce in many boreal forests of North America, fires tend to burn hotter and faster, traveling through the crowns of trees and inducing higher levels of tree mortality [Rogers et al., 2015]. This occurs because black spruce is a well-known fire embracer, retaining dead branches that serve as ladder fuels– carrying fire into the overstory where seeds in serotinous cones are activated by fire. Black spruce trees are absent from Siberia, where many pine and larch tree
- 25 species lack ladder fuels and are known as fire resistors. In Siberian ecosystems ground fires are more common [Rogers et al., 2015], a finding that appears consistent with the higher CO emission ratios (and stronger contribution of smoldering combustion) shown in Table 1.

4.2 Evidence for a stronger role of smoldering combustion in emissions from North American boreal wildfires,

Our mean emission factor for CO (127 ± 59 g CO per kg of dry biomass consumed) is similar to the mean reported in past
 30 syntheses for boreal forests, including estimates by *Andreae* [2019] (121 ± 47 g CO per kg of dry biomass consumed) and *Akagi et al.* [2011] (127 ± 45 g CO per kg of dry biomass consumed). However, if studies that are not representative of North American boreal forest wildfires are excluded (including measurements from prescribed fires, laboratory studies, and studies of fires from the Siberian boreal forests) and we focus on emission ratios, to avoid uncertainties introduced by the limited number of measurements that report the carbon content of combusted fuels, our estimate is 39% higher (and significantly different at a p <

35 0.01 level using a Student's t test) than the mean derived from aircraft studies of North American boreal wildfires (Table 1). Considering the higher emission ratio of our measurements, we believe the CRV observations we analyzed here provide evidence that boreal forest fires in North America have a stronger contribution from smoldering combustion than what has been estimated in previous reports. Our CRV tower-based sampling was able to integrate over day-night burning cycles, flaming combustion at active fire fronts as well as residual smoldering combustion in soils that persists for days after the fire front moves

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studies and time intervals-

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5 <u>extinction.</u> Collectively, these fires appeared to experience time-varying environmental conditions that were less ideal for flaming combustion than the fire plumes sampled in past work by aircraft.

Following ignition, <u>North American</u> boreal forest fires generally <u>expand with flaming combustion in the crown.</u> <u>Smoldering combustion in organic soil layers and coarse woody debris behind the fire front that can continue for weeks after ignition [*Bertschi et al.*, 2003]. This residual smoldering combustion could substantially contribute to trace gas emissions but is</u>

- 10 usually excluded from FRP based fire emissions inventories because of the difficulty in detecting low FRP associated with this process of combustion. The relative contributions of consumption, from flaming and smoldering are uncertain for boreal forest fires, although several previous studies have assumed 80% of aboveground carbon is consumed in flaming combustion, 20% is consumed in smoldering combustion, and vice versa for belowground carbon [*French et al.*, 2002; *Kasischke and Bruhwiler*, 2002]. Our results suggest that the smoldering process of combustion contributes to more to CQ emissions that previously thought.
- 15 In the context of interpreting the CRV measurements, it's important to note that MISR satellite observations from Alaskan wildfires indicate most fire plumes reside within the planetary boundary layer, which is typically between 1 and 3 km during midday in summer [val Martin et al. 2010; Wiggins et al., 2016]. Combining this length scale with the mean distance of the 35 fires that most influenced CO at CRV (295 km), we obtain a factor of about 100 for a back-of-the-envelope ratio of horizontal to vertical mixing processes. This implies that mesoscale atmospheric circulation played an important role in delivering fire-emitted
- 20 trace gases to CRV. This ratio is considerably larger than what would be inferred from the location and sampling strategy of several past studies that have used surface towers to sample fires near or within fire perimeters [Collier et al., 2016; Benedict et al., 2017; Selimovic et al., 2019a,b], band highlights the unique role that a remote tower can have in providing an integrated assessment of a large regional fire complex.

Finally, we note that during the latter half of June and early July of 2015, weather in Alaska was very hot and dry, allowing
 a record number of fires to rapidly expand in size, and yielding the second highest level of annual burned area in the observed record. The extreme fire weather conditions would be expected to reduce fuel moisture content, thus promoting flaming combustion processes. This raises the question of whether longer term monitoring of many normal and low fire years (which tend to co-occur in cooler and wetter conditions) would provide evidence for an even larger role of smoldering combustion for wildfire emissions from these ecosystems. Another related question is whether even within a fire season, do day-to-day or week-to-week variations

30 in fire weather influence variability in emission ratios. We explored this latter question with the datasets described here but were unable to uncover structural relationships between daily meteorological variables such as vapor pressure deficit and CO emission ratios. Together, these questions represent important directions for future research and emphasize the critical need of sustained long-term support for trace gas monitoring networks and field campaigns.

35 <u>4.3 Synthesis of emission factor observations across studies</u>

With many new field campaigns measuring fire emissions, there is a need to revisit how information from different studies is combined to generate the most reliable set of emission factors for regional and global atmospheric models. Several ideas for an improved synthesis have emerged from our study.

First, it may make sense to separately report emission factors for Siberian and North American boreal forest fires, given 40 what we know about differences in species composition, fire dynamics, and measurements of emission factors between the two

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→ Our modeling study confirms that the entire day/night fire cycle was captured by anomalous trace gas observations at CRV tower that was used to calculate emission factors. Wiggins et al. [2016] used a similar tower-based approach to estimate boreal forest emission factors during a moderate fire year, and they found CO and CH4 emission factors that were higher than the compiled mean from previous studies. We found a strong linear relationship between CH4 emission factors and MCE that has also been observed in previous studies [Van Leeuwen and Van Der Werf, 2011; Yokelson et al., 2013; Urbanski, 2014].

Although Table 1 appears to suggest CO emission factors from boreal forest fires are increasing over time, it is more likely that studies using the tower approach are better suited to sample a more thorough representation of all the phases of combustion that can occur in boreal forest fires. The tower approach is not limited by the time or scale of sampling, unlike aircraft measurement techniques. Aircraft based emission factors are often biased towards flaming fires. because most measurements are acquired during the afternoon when active fire plumes are visible. The emission factors derived from this study provide a more robust estimate of the mean, and indicate that the smoldering phase and nighttime emissions of boreal fires have likely been underestimated in previous studies. The improved emission factors from this study can be used in future modeling efforts to convert carbon emissions to CO and CH4 trace gas emissions from horeal forest fires more accurately 4.3 Relative Contributions of Smoldering and Flaming Combustion

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Second, it's important to further explore ways to weight the information content from different studies, considering the number of fires sampled, the duration and intensity of sampling, the representativeness of the sampling approach, and the

- 5 representativeness of the fire complexes that were sampled relative to the typical pattern of burning within a biome. Here using a remote surface tower, we were able to get an integrated estimate of CO and CH4 emission ratios from about 35 wildfires from an ecologically significant regional fire complex. While these observations represent a step change in CO and CH4 data availability for North American boreal forest fires, more work is needed to find a way to systematically combine this information with other observations generated using different sampling techniques.
- 10 Third, even for an individual fire, steps toward flux-weighting different emission factors would be an important path toward reducing uncertainties, yet this goal remains technically challenging given existing measurement techniques. Our sensitivity analysis, in which we computed a weighted-mean CO emission ratio using CO or CO₂ concentrations during each fire-affected sampling interval, provided an indication of the robustness of our mean estimates to weighting scheme. This approach falls short, however, of providing a flux-weighted estimate given atmospheric processes that may decouple concentration from flux, including,
- 15 for example, variations in windspeed, diurnal variations in planetary boundary layer height, and the distance between the emissions source and measurement point. To make progress on this issue, a closer integration is needed in future field campaigns between instantaneous measurements of fire behavior (temperature, fire radiative power, and spread rate), measurements of emissions composition, and post-fire sampling of fuel structure and consumption during times when fire dynamics were fundamentally different. This coordination across disciplines in both study design, data analysis, and modeling is rare and may provide a path
- 20 <u>toward creating the observations needed to dynamically model the temporal evolution of the chemical composition of wildland</u> <u>fire emissions over the lifetime of an individual fire and during different phases of a fire season</u>

4.4 Implications of a larger contribution of smoldering combustion

that often leads to climate cooling [Tosca et al., 2010; Jayarathne et al., 2018].

Smoldering combustion produces significantly more CO and PM2.5 than flaming combustion [*Bertschi et al.*, 2003; *Chen et al.*, 2007; *Stockwell et al.*, 2016], and <u>our work suggests North American</u> boreal forest fire emissions of these species are likely
higher than previous thought, This conclusion implies changes to the overall impact of boreal forest fires on human health, atmospheric composition, and climate. Emissions from boreal forest fires have the potential to be transported long distances across the Northern Hemisphere [*Forster et al.*, 2001], implying large-scale impacts. CO can lead to enhanced tropospheric ozone production downwind of a fire [*Lapina et al.*, 2006], and higher concentrations of CO from fires may indirectly contribute to radiative forcing by consuming hydroxyl radicals and extending the lifetime of CH4 [*Levine and Cofer*, 2000]. PM2.5 emissions, in
contrast, can significantly degrade regional air quality, endanger cardiovascular and respiratory health, and influence the radiative balance of the planet [*Reid et al.*, 2016]. The timing of emissions is important for quantifying the impact on human health, and enhanced nighttime emissions (Figure 10) when the boundary layer is much lower could increase surface concentrations and exacerbate negative health effects. Much of the PM2.5 emitted by smoldering fires is composed of organic carbonaceous aerosol

35 5 Conclusions

Using a remote tower downwind of a large regional fire complex in interior Alaska, we measured CO and CH4 emission factors from about 35 individual fires during the summer of 2015. This is more than the number of individual wildfires that have

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been sampled in North America, in all previous studies combined. Our results suggest smoldering combustion processes in North American boreal forest fires contribute more trace gas emissions than previously thought, and as a consequence, total CO emissions may have been underestimated in model simulations of boreal forest fire impacts on atmospheric composition. Long-term monitoring from remote towers may provide a means to quantitatively sample fire complexes in other biomes, integrating across day-night variations in fire behavior, periods with different environmental conditions, and across multiple fires in different stages

of growth and extinction,

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Figure 2. A) Observations of CO₂ mole fraction from CRV tower in 2012 (black) along with model estimates of the CO₂ background (green) at CRV using the approach described in the main text. Very few fires occurred during 2012, and as a consequence most of the CO₂ variability in the observations and in the model is associated with terrestrial net ecosystem exchange. B) In 2015 wildfires in interior Alaska contributed significantly to CO₂ variability at the CRV tower, causing positive anomalies in the observations shown in black, particularly between days 170 and 190. The modeled background for 2015 is shown in red.

The CO2 mole fraction observations and model estimates have a 1 hour temporal resolution

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Figure 3. <u>Trace</u> gas observations at the CRV tower during the summer of 2015 for A) CO, B) CH₄, and C) CO₂. The trace gas observations are plotted at a 30 s temporal resolution. Daily active fire detections derived the MODIS instrument on Terra and Aqua satellites (MCD14ML C6) are shown in panel D₄.

-(Deleted: Panels A-C: trace
-(Deleted: (black) from
-(Deleted: . Panel D: number of daily
)(Deleted: fires from
1	Deleted: (red).





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Figure 4. CRV tower observations of A) $CO_{,,,,} = CO_{,,,} = CO_{,,,,} = CO_{,,,,,} = CO_{,,,,,,} = CO_{,,,,,,} = CO_{,,,,,,} = CO_{,,,,,,} = CO_{,,,,,,,} = CO_{,,,,,,,} = CO_{,,,,,,} = CO_{,,,,,,,} = CO_{,,,,,,,} = CO_{,,,,,,,} = CO_{,,,,,,,,$



Figure 5. Relationship between CH4 emission factor, and modified combustion efficiency (MCE). The strong linear relationship indicates that periods with more smoldering combustion (with a smaller MCE) produce significantly higher levels of CH₄ emissions. The relationship was defined by a slope of -46.37 \pm 4.13 g CH₄ per kg dry biomass per MCE, an X intercept of -0.47 \pm

5 0.05 g CH₄ per kg dry biomass an R² of 0.54, and a significance value of p <0.01.

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5 <u>Table 2.</u>





Figure 7. CRV observations of CO (black) compared with the modeled CO anomaly from fires (red) derived from PWRF-STILT 5 driven by AKFED fire emissions. The trace gas observations and model predictions are shown at a 1 hr temporal resolution.







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Figure 9. A) Top, 5 individual fire contributions to the CO anomaly simulated at CRV tower. Black shows original PWRF-STILT
 <u>AKFED</u> model, red depicts contributions from the Tozitna fire, green from Kobe fire, blue from Blair fire, gold from Aggie Creek fire, and purple from Spicer Creek fire. B) The total CO anomaly from the 35 fires that contributed to at least 1% of the
 modeled CO anomaly at CRV tower (red) compared to the original model (black).



Tables

Study	CO Emission	MCE	Number of				
	Ratio		fires sampled				
North American wildfires sampled by aircraft							
Cofer et al., 1989	0.069 ± 0.004	$\underline{0.935 \pm 0.004}$	1				
Cofer et al., 1998	0.140 ± 0.012	$\underline{0.878 \pm 0.009}$	1				
Friedli et al., 2003	$\underline{0.100\pm0.020}$	$\underline{0.909 \pm 0.017}$	<u>1</u> 5				
Goode et al., 2000	$\underline{0.085 \pm 0.008}$	$\underline{0.922 \pm 0.007}$	<u>4</u>				
Laursen et al., 1992	$\underline{0.050\pm0.007}$	$\underline{0.953 \pm 0.006}$	<u>1</u>				
Nance et al., 1993	$\underline{0.078 \pm 0.012}$	$\underline{0.928 \pm 0.011}$	<u>1</u>				
O'Shea et al., 2013	0.150 ± 0.024	0.871 ± 0.012	<u>4</u>				
Radke et al., 1991	0.116 ± 0.087	0.896 ± 0.075	<u>1</u>				
Simpson et al., 2011	0.110 ± 0.070	0.901 ± 0.061	<u>5</u>				
Fire-weighted mean	$\underline{0.102 \pm 0.033}$	$\underline{0.908 \pm 0.027}$	<u>19</u>				
North American	management fire	s sampled by airc	raft				
Cofer et al., 1990	$\underline{0.086 \pm 0.008}$	$\underline{0.921 \pm 0.007}$	<u>2</u>				
Cofer et al., 1998	0.095 ± 0.016	$\underline{0.913 \pm 0.013}$	<u>7</u>				
Radke et al., 1991	0.047 ± 0.032	$\underline{0.956 \pm 0.030}$	<u>4</u>				
Susott et al., 1991	0.060 ± 0.061	0.943 ± 0.058	10				
Fire-weighted mean	0.077 ± 0.022	0.929 ± 0.020	<u>14</u>				
North American fuels sampled in the laboratory							
Bertschi et al., 2003	0.151 ± 0.040	0.870 ± 0.030	-				
Burling et al., 2010	0.209	0.827					
Mcmeeking et al., 2009	0.091 ± 0.038	0.917 ± 0.068					
Mean	0.150 ± 0.039	0.871 ± 0.049					
Siberian wildfires	- sampled by air	craft or surface to	ower				
Cofer et al., 1998 (A)	0.224 ± 0.036	0.817 ± 0.025	1				
McRay et al., 2006 (A & S)	0.249 ± 0.064	0.800 ± 0.043	6				
Vasileva et al., 2017 (S)	0.126 ± 0.007	0.888 ± 0.005	2				
Fire-weighted mean	0.219 ± 0.048	0.822 ± 0.033	<u>1</u> 5				
North American	wildfires sample	ed by surface tow	/er				
Wiggins et al. 2016	0.128 ± 0.023	0.887 ± 0.018	3				
This study	0.142 ± 0.051	$\frac{0.878 \pm 0.039}{0.878 \pm 0.039}$	35				
Fire-weighted mean	0.141 ± 0.049	0.879 ± 0.027	38				
a ne mergareet mettin	0.1	0.017 - 0.027	00				

Table 1. Comparison of CO emission ratio and modified combustion efficiency (MCE) from previous studies that sampled emissions from boreal forest fires. The studies are organized according to wildfire domain (North America or Siberia), management 20 practice (wildfire or management, fire), and sampling approach (aircraft, laboratory, or surface tower). Siberian studies are indicated as aircraft studies (A), surface based studies (S), or a combination of the two (A & S), The CO emission ratio column has units of ppmv ppmv⁻¹ and uses CO₂ as the reference gas. MCE was calculated as 1/(1+CO emission ratio) when not directly reported in the study. The weighted mean of emission ratios and MCE for all previous studies is shown in the row labeled fire-weighted mean, with each study weighted according to the number of fires sampled.

Deleted: and CH4 Deleted: ratios, emission factors, Deleted: measured in-situ Deleted: fires. Number of fires sampled per study is given in parenthesis following the location. Aircraft based studies are denoted with an (A) following the sampling strategy and ground based studies are denoted with a (G) following the sampling strategy. Deleted: based on the type of Deleted: they measured, prescribed fires or wildfires. We did not include studies that measured slash and tramp prescribed fires or laboratory fires. Deleted: use Deleted: This column is separated into the mean per study and by fire combustion phase including flaming, mixed, and smoldering when available. Emission factors were Deleted: from emission ratios assuming the carbon content of combusted fuel was 45% when not provided by the study. MCE was calculated Deleted: given Deleted: , emission factors, Deleted: aircraft based Deleted: by Formatted: Font: 10 pt Deleted: 1 Deleted: ¶

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$ \begin{array}{c c} (pro)^{-1} & Falto & Falto & Falto & Falto & Falto & Falto & Phase \\ (prov / prov)^{-1} & (gcV1/kg & gcV1/kg & gcV1/kg & grV homas) \\ \hline \begin{tabular}{lllllllllllllllllllllllllllllllllll$	Event	Ν	Time of Event	CO Emission	CO Emission	CH ₄ Emission	CH ₄ Emission	MCE	Combustio
$ \begin{array}{c} (p) (n) - p(n) - p(n) - (g \in O \times g) \\ (p) (p) (p) (p) (p) (g \in O \times g) \\ (p) (p) (p) (p) (p) (p) (p) (p) (p) (p)$			(DOY)	Ratio	Factor	Ratio	Factor		Phase
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $				(ppmv / ppmv)	(g CO / kg dry biomass)	(ppmv / ppmv)	(g CH4 / Kg dry biomass		
9 173	1	82	173.27 - 173.30	0.161 ± 0.004	144 ± 4	0.012 ± 0.0003	6.1 ± 0.2	0.861 ± 0.004	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	2	95	173.32 - 173.35	0.151 ± 0.004	136 ± 4	0.011 ± 0.0002	5.8 ± 0.2	0.869 ± 0.004	Mixed
83 173.40 173.43 0.149 ± 0.008 135 ± 8 0.011 ± 0.006 5.5 ± 0.3 0.876 ± 0.008 Mixed 95 173.84 173.87 0.130 ± 0.006 124 ± 8 0.004 ± 0.0007 73 ± 0.5 0.885 ± 0.006 Mixed 95 $175.15 + 175.18$ 0.080 ± 0.007 131 ± 7 0.008 ± 0.004 4.2 ± 0.3 0.375 ± 0.07 Mixed 95 $175.15 + 175.18$ 0.009 ± 0.002 87 ± 1 0.005 ± 0.0001 2.2 ± 0.1 0.916 ± 0.002 Mixed 86 175.22 1143 ± 0.007 131 ± 7 0.008 ± 0.004 4.2 ± 0.3 0.375 ± 0.007 Mixed 87 175.40 1073 ± 0.001 87 ± 1 0.005 ± 0.000 2.5 ± 0.1 0.376 ± 0.001 Mixed 89 $175.40 + 175.44$ 0.187 ± 0.012 164 ± 12 0.013 ± 0.001 8.1 ± 0.004	3	95	173.36 - 173.39	0.141 ± 0.003	128 ± 3	0.010 ± 0.0002	5.5 ± 0.1	0.877 ± 0.003	Mixed
95 173.45 -173.48 0.136 0.008 124 8 0.014 0.0009 7.3 0.385 0.006 Mixed 95 173.45 -173.47 0.136 0.008 152 8 0.004 -0.009 7.3 -0.008 Mixed 3.3 0.22 -0.885 -0.008 Mixed 95 175.15 -175.12 0.041 -0.001 4.23 -0.01 0.926 +<	4	83	173.40 - 173.43	0.149 ± 0.008	135 ± 8	0.011 ± 0.0005	5.5 ± 0.3	0.870 ± 0.008	Mixed
95 173.84 -173.87 0.179 ± 0.008 152 ± 8 0.008 ± 0.0003 4.3 ± 0.2 0.855 ± 0.008 Mixed 95 175.15 -175.18 0.009 ± 0.001 78 ± 0.3 0.004 ± -1c4 2.3 ± 0.1 0.916 ± 0.002 Mixed 88 175.27 -175.30 0.091 ± 0.002 87 ± 1 0.005 ± 0.0001 2.9 ± 0.1 0.917 ± 0.001 Mixed 81 175.27 -175.30 0.091 ± 0.001 87 ± 1 0.005 ± 0.0002 2.4 ± 0.1 0.917 ± 0.001 Mixed 95 175.60 -175.70 0.006 ± 0.003 59 ± 3 0.005 ± 0.001 2.9 ± 0.1 0.943 ± 0.003 Faming 95 175.60 -175.77 0.129 ± 0.001 119 ± 1 0.009 ± 0.001 8.1 ± 0.01 S.5 ± 0.1 0.842 ± 0.012 Smolderi 95 175.80 -175.83 0.147 ± 0.002 133 ± 2 0.011 ± 0.0001 5.4 ± 0.1 0.885 ± 0.003 Mixed 95 175.92 -175.96 0.198 ± 0.001 112 ± 4 0.012 ± 0.0001 5.4 ± 0.1 0.883 ± 0.003	5	95	173.45 - 173.48	0.130 ± 0.006	120 ± 6	0.009 ± 0.0004	5.0 ± 0.3	0.885 ± 0.006	Mixed
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	6	95	173.84 - 173.87	0.136 ± 0.008	124 ± 8	0.014 ± 0.0009	7.3 ± 0.5	0.880 ± 0.008	Mixed
95 175.15 175.18 0.080 ± 0.001 78 ± 0.3 0.004 ± -(1-4) 2.3 ± -0.1 0.972 ± -(1-5) Planning 95 175.12 15.175.25 0.091 ± 0.001 87 ± 1 0.005 ± 0.0001 2.5 ± 0.1 0.875 ± 0.003 Mixed 88 175.27 175.30 0.091 ± 0.001 187 ± 1 0.005 ± 0.0001 2.4 ± 0.101 Mixed 95 175.40 175.44 0.187 ± 0.012 164 ± 12 0.013 ± 0.0002 2.6 ± 0.1 0.984 ± 0.003 Filaming 95 175.56 175.70 0.129 ± 0.001 119 ± 1 0.009 ± 0.001 8.1 ± 0.6 0.892 ± 0.014 Smoleri 95 175.80 1.75.83 0.147 ± 0.002 133 ± 2 0.011 ± 0.001 5.4 ± 0.1 0.886 ± 0.001 Mixed 95 175.88 1.75.9 0.198 ± 0.004 172 ± 4 0.012 ± 0.001 5.4 ± 0.1 0.833 ± 0.004 Smoleri 16 175.98 1.76.96 0.198 ± 0.003 110 ± 0.1001 5.4 ± 0.1 0.838 ± 0.007 Mixed 17.17.17.17.99<	7	85	174.27 - 174.30	0.170 ± 0.008	152 ± 8	0.008 ± 0.0003	4.3 ± 0.2	0.855 ± 0.008	Mixed
95 175.19 175.22 0.014 ± 0.002 87 \pm 2 0.005 ± 0.0002 2.5 \pm 0.1 0.917 ± 0.001 Mixed 88 175.27 175.30 0.013 ± 0.001 87 \pm 1 0.005 ± 0.0002 2.5 \pm 0.1 0.917 ± 0.001 Mixed 95 175.30 0.137 ± 0.012 164 ± 12 0.013 ± 0.0002 4.5 ± 0.1 0.842 ± 0.012 Smolderi 95 175.66 175.70 0.129 ± 0.011 119 ± 1 0.009 ± 0.0002 4.5 ± 0.1 0.884 ± 0.001 Mixed 95 175.80 175.70 0.129 ± 0.001 119 ± 1 0.009 ± 0.0002 4.9 ± 0.2 0.086 ± 0.003 Mixed 95 175.80 175.91 0.153 ± 0.003 169 ± 3 0.011 ± 0.0001 5.4 ± 0.1 0.838 ± 0.003 Mixed 95 175.80 1076.0 0.193 ± 0.003 169 ± 3 0.011 ± 0.0001 5.4 ± 0.1 0.838 ± 0.003 Mixed 95 176.16 0.108 ± 0.001 119 ± 1 0.0108 ± 0.001 5.5 ± 0.1 0.038 ± 0.003	8	95	175.15 - 175.18	$0.080 \pm < 0.001$	78 ± 0.3	$0.004 \pm < 1e4$	$2.3 \pm < 0.1$	$0.926 \pm < 1e3$	Flaming
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	9	95	175.19 - 175.22	0.143 ± 0.007	131 ± 7	0.008 ± 0.0004	4.2 ± 0.3	0.875 ± 0.007	Mixed
$ \begin{array}{c} 88 & 175.27 + 175.30 & 0.091 \pm 0.001 & 87 \pm 1 & 0.005 \pm 0.0001 & 2.9 \pm 0.01 & 0.917 \pm 0.001 & Mixed \\ 89 & 175.40 + 175.44 & 0.187 \pm 0.012 & 164 \pm 12 & 0.013 \pm 0.0008 & 6.4 \pm 0.5 & 0.0842 \pm 0.012 & Smoldering \\ 55 & 175.75 + 175.77 & 0.129 \pm 0.001 & 119 \pm 1 & 0.009 \pm 0.0001 & 4.5 \pm 0.1 & 0.886 \pm 0.001 & Mixed \\ 35 & 175.75 + 175.77 & 0.237 \pm 0.015 & 198 \pm 15 & 0.017 \pm 0.0010 & 8.1 \pm 0.6 & 0.899 \pm 0.002 & Mixed \\ 95 & 175.86 + 175.79 & 0.237 \pm 0.015 & 198 \pm 15 & 0.009 \pm 0.0001 & 4.5 \pm 0.1 & 0.872 \pm 0.002 & Mixed \\ 95 & 175.92 + 175.96 & 0.198 \pm 0.004 & 172 \pm 4 & 0.012 \pm 0.000 & 4.9 \pm 0.2 & 0.866 \pm 0.003 & Mixed \\ 95 & 175.92 + 175.96 & 0.198 \pm 0.004 & 172 \pm 4 & 0.012 \pm 0.0001 & 5.4 \pm 0.1 & 0.838 \pm 0.003 & Smoldering \\ 95 & 175.06 - 176.00 & 0.193 \pm 0.003 & 169 \pm 3 & 0.011 \pm 0.0001 & 5.4 \pm 0.1 & 0.838 \pm 0.003 & Smoldering \\ 95 & 177.06 - 177.09 & 0.108 \pm 0.001 & 102 \pm 1 & 0.0104 \pm 0.0001 & 5.4 \pm 0.1 & 0.892 \pm 0.001 & Mixed \\ 95 & 177.16 + 177.14 & 0.122 \pm 0.001 & 119 \pm 1 & 0.0104 \pm 0.0001 & 5.4 \pm 0.1 & 0.892 \pm 0.001 & Mixed \\ 95 & 177.15 + 177.18 & 0.129 \pm 0.001 & 119 \pm 1 & 0.0114 \pm 0.0001 & 5.5 \pm 0.1 & 0.886 \pm 0.001 & Mixed \\ 95 & 177.19 + 177.22 & 0.102 \pm 0.002 & 96 \pm 2 & 0.008 \pm 0.0002 & 4.4 \pm 0.1 & 0.886 \pm 0.001 & Mixed \\ 95 & 177.19 + 177.23 + 0.002 & 10.49 \pm 2 & 0.008 \pm 0.0001 & 2.3 \pm 0.1 & 0.984 \pm 0.002 & Mixed \\ 95 & 177.88 + 177.91 & 0.129 \pm 0.001 & 119 \pm 1 & 0.0101 & 0.0011 & 2.5 \pm 0.1 & 0.886 \pm 0.001 & Mixed \\ 95 & 177.88 + 177.91 & 0.129 \pm 0.002 & 199 \pm 2 & 0.008 \pm 0.0002 & 4.4 \pm 0.1 & 0.94 \pm 0.002 & Hixed \\ 95 & 177.88 + 177.91 & 0.129 \pm 0.002 & 111 \pm 6 & 0.029 \pm 0.0011 & 2.3 \pm 0.1 & 0.944 \pm 0.002 & Hixed \\ 95 & 177.88 + 177.91 & 0.124 \pm 0.002 & 199 \pm 2 & 0.004 \pm 0.0001 & 2.3 \pm 0.1 & 0.944 \pm 0.002 & Hixed \\ 95 & 188.10 & 188.13 & 0.106 \pm 0.002 & 29 \pm 2 & 0.008 \pm 0.0002 & 4.5 \pm 0.1 & 0.944 \pm 0.002 & Hixed \\ 95 & 188.10 & 1.88 \pm 3.0 & 0.064 \pm 0.002 & 129 \pm 2 & 0.002 \pm 0.0011 & 12 \pm 0.1 & 0.944 \pm 0.002 & Hixed \\ 95 & 188.10 & 1.018 \pm 0.001 & 120 \pm 0.0003 & 4.2 \pm 0.2 & 0.966 \pm 0.004 & Hixed \\ 95 $	10	58	175.23 - 175.25	0.091 ± 0.002	87 ± 2	0.005 ± 0.0002	2.5 ± 0.1	0.916 ± 0.002	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	11	88	175.27 - 175.30	0.091 ± 0.001	87 ± 1	0.005 ± 0.0001	$2.9 \pm < 0.1$	0.917 ± 0.001	Mixed
89 175.40 175.44 10.13 ± 0.0008 6.4 ± 0.5 0.842 ± 0.012 Smolder 95 175.56 175.77 0.129 ± 0.001 119 ± 1 0.009 ± 0.0001 8.1 ± 0.6 0.809 ± 0.014 Smolder 95 175.77 $0.175.79$ 0.237 ± 0.015 198 ± 15 0.017 ± 0.001 8.1 ± 0.6 0.809 ± 0.014 Smolder 95 175.80 $1.175.79$ 0.125 ± 0.003 139 ± 3 0.009 ± 0.0002 4.9 ± 0.2 0.866 ± 0.003 Mixed 95 175.80 0.119 ± 0.003 169 ± 3 0.011 ± 0.0001 5.4 ± 0.1 0.872 ± 0.003 Smolder 95 176.00 0.193 ± 0.003 169 ± 3 0.011 ± 0.0001 5.4 ± 0.1 0.833 ± 0.003 Smolder 95 177.06 0.170 ± 0.002 119 ± 1 0.011 ± 0.0001 5.4 ± 0.1 0.882 ± 0.001 Mixed 95 177.16 $0.171.8$ 0.129 ± 0.002 198 ± 2 0.004 ± 0.0002 44 ± 0.3 0.892 ± 0.002 Mixed 95 177.19 $0.172.0$ 0.129 ± 0.002 99 ± 2 0.004 ± 0.0	12	95	175.32 - 175.35	0.153 ± 0.003	138 ± 4	0.009 ± 0.0002	4.5 ± 0.1	0.867 ± 0.003	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	13	89	175.40 - 175.44	0.187 ± 0.012	164 ± 12	0.013 ± 0.0008	6.4 ± 0.5	0.842 ± 0.012	Smolderin
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	14	95	1/5.66 - 1/5./0	0.060 ± 0.003	59 ± 3	0.005 ± 0.0002	2.6 ± 0.1	0.943 ± 0.003	Flaming
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	10	22	1/5./5 - 1/5.//	0.129 ± 0.001	119 ± 1	0.009 ± 0.0001	4.5 ± 0.1	0.886 ± 0.001	Mixed
$\begin{array}{c} 95 & 17.5.80 - 17.9.10 \\ 95 & 175.80 - 17.9.10 \\ 1.55 \pm 0.001 \\ 1.55 \pm 0.001 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 95 & 175.90 \\ 1.75 \\ 1.77 \\ 1.1 \\ 1.1 \\ 1$	16	35	1/5.// - 1/5./9	0.237 ± 0.015	198 ± 15 122 + 2	0.017 ± 0.0010	8.1 ± 0.6	0.809 ± 0.014	Smolderin
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	10	95	1/5.80 - 1/5.85	0.147 ± 0.002 0.155 ± 0.002	133 ± 2 120 + 2	0.011 ± 0.0001	5.5 ± 0.1	$0.8/2 \pm 0.002$	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	10	95	175.00 - 175.91	0.133 ± 0.003	139 ± 3 172 ± 4	0.009 ± 0.0002	4.9 ± 0.2	0.800 ± 0.003	Smaldanin
$ \begin{array}{c} 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 \\ 10 $	19	95 80	175.92 - 175.96	0.198 ± 0.004 0.103 ± 0.003	$1/2 \pm 4$ 160 ± 3	0.012 ± 0.0001 0.011 ± 0.0001	6.1 ± 0.1 5 4 ± 0.1	0.833 ± 0.004 0.838 ± 0.003	Smolderin
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	20	95	176.06 - 176.00	0.193 ± 0.003 0.119 ± 0.007	109 ± 3 111 ± 7	0.011 ± 0.0001 0.008 ± 0.0004	3.4 ± 0.1 4.4 ± 0.3	0.838 ± 0.003 0.893 ± 0.007	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	21	85	177.06 - 177.09	0.119 ± 0.007 0.108 ± 0.001	102 ± 1	0.000 ± 0.0004 0.010 ± 0.0001	$53 \pm < 0.1$	0.003 ± 0.007 0.002 ± 0.001	Mixed
95177.15 177.18 0.125 ± 0.001 119 ± 1 0.011 ± 0.0001 5.5 ± 0.1 0.886 ± 0.002 Mixed95 $177.19 - 177.22$ 0.102 ± 0.002 96 ± 2 0.008 ± 0.002 4.4 ± 0.1 0.908 ± 0.002 Mixed94 $177.27 - 177.31$ 0.060 ± 0.002 89 ± 2 0.004 ± 0.001 2.3 ± 0.1 0.944 ± 0.002 Hixed95 $177.80 - 177.83$ 0.094 ± 0.002 89 ± 2 0.004 ± 0.001 1.1 ± 0.1 0.914 ± 0.002 Mixed95 $177.80 - 177.83$ 0.094 ± 0.002 89 ± 2 0.008 ± 0.001 1.1 ± 0.1 0.914 ± 0.002 Mixed95 $177.80 - 177.92$ 0.164 ± 0.006 111 ± 6 0.020 ± 0.0012 10.7 ± 0.7 0.893 ± 0.006 Mixed95 $184.23 - 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ 0.025 ± 0.002 2.5 ± 2 0.002 ± 0.001 1.2 ± 0.1 0.976 ± 0.002 Hixed95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0002 4.5 ± 0.1 0.904 ± 0.002 Mixed54 $188.14 - 188.16$ 0.109 ± 0.001 102 ± 1 0.008 ± 0.0001 4.2 ± 0.2 0.906 ± 0.004 Mixed55 $188.40 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 4.2 ± 0.2 0.906 ± 0.004 Mixed55 $188.45 - 188.48$ 0.131 ± 0.004 102 ± 4 0.013 ± 0.0001 6.9 ± 0.3 0.884 ± 0.002 <td< td=""><td>2</td><td>75</td><td>177 11 - 177 14</td><td>0.103 ± 0.001 0.122 ± 0.002</td><td>102 ± 1 113 ± 2</td><td>0.010 ± 0.0001 0.011 ± 0.0001</td><td>5.5 ± 0.1 56 ± 01</td><td>0.902 ± 0.001 0.892 ± 0.002</td><td>Mixed</td></td<>	2	75	177 11 - 177 14	0.103 ± 0.001 0.122 ± 0.002	102 ± 1 113 ± 2	0.010 ± 0.0001 0.011 ± 0.0001	5.5 ± 0.1 56 ± 01	0.902 ± 0.001 0.892 ± 0.002	Mixed
95177.19 177.22 0.102 ± 0.002 96 ± 2 0.008 ± 0.002 4.4 ± 0.1 0.908 ± 0.002 Mixed94 $177.23 - 177.25$ 0.148 ± 0.011 134 ± 12 0.012 ± 0.0001 6.0 ± 0.5 0.871 ± 0.011 Mixed94 $177.27 - 177.31$ 0.060 ± 0.002 59 ± 2 0.004 ± 0.0001 2.3 ± 0.1 0.944 ± 0.002 Flaming95 $177.80 - 177.83$ 0.094 ± 0.002 89 ± 2 0.004 ± 0.0001 1.1 ± 0.1 0.914 ± 0.002 Mixed95 $177.80 - 177.83$ 0.120 ± 0.006 111 ± 6 0.020 ± 0.0012 10.7 ± 0.7 0.893 ± 0.006 Mixed95 $177.82 - 177.91$ 0.124 ± 0.006 1146 ± 7 0.018 ± 0.007 8.9 ± 0.4 0.859 ± 0.006 Mixed95 $182.33 - 184.26$ 0.232 ± 0.012 156 ± 13 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ 0.025 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.1 0.976 ± 0.002 Flaming64 $188.07 - 188.10$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0004 4.5 ± 0.1 0.904 ± 0.002 Mixed54 $188.14 - 188.16$ 0.109 ± 0.001 102 ± 1 0.008 ± 0.0001 4.3 ± 0.1 0.904 ± 0.002 Mixed52 $188.25 - 188.25$ 0.804 ± 0.007 $77 7 7$ 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.45 - 188.48$ 0.13 ± 0.004 120 ± 4 0.013 ± 0.0001 6.9 ± 0.1 0.873 ± 0.0	24	95	177.15 - 177.18	0.122 ± 0.002 0.129 ± 0.001	119 ± 2 119 ± 1	0.010 ± 0.0001	5.5 ± 0.1	0.886 ± 0.001	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	25	95	177 19 - 177 22	0.102 ± 0.002	96 ± 2	0.008 ± 0.0002	44 + 01	0.908 ± 0.002	Mixed
94 $177.27 - 177.31$ 0.060 ± 0.002 59 ± 2 0.004 ± 0.0001 2.3 ± 0.1 0.944 ± 0.002 Flaming95 $177.80 - 177.83$ 0.094 ± 0.002 89 ± 2 0.008 ± 0.0001 4.1 ± 0.1 0.914 ± 0.002 Mixed95 $177.80 - 177.91$ 0.120 ± 0.006 111 ± 6 0.020 ± 0.0012 10.7 ± 0.7 0.893 ± 0.006 Mixed93 $177.92 - 177.96$ 0.164 ± 0.006 114 ± 6 0.020 ± 0.0007 8.9 ± 0.4 0.839 ± 0.006 Mixed95 $184.23 - 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ 0.025 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.11 0.904 ± 0.002 Mixed95 $188.10 - 188.109$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0007 6.5 ± 0.4 0.812 ± 0.012 Smolderi95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0001 $4.3 \pm < 0.1$ 0.904 ± 0.002 Mixed64 $188.20 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 4.2 ± 0.2 0.902 ± 0.001 Mixed52 $188.40 - 188.44$ 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0002 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.55 - 188.64$ 0.164 ± 0.002 132 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0	26	58	177.23 - 177.25	0.148 ± 0.011	134 ± 12	0.012 ± 0.0009	6.0 ± 0.5	0.871 ± 0.011	Mixed
95 $177.80 - 177.83$ 0.094 ± 0.002 89 ± 2 0.008 ± 0.0001 4.1 ± 0.1 0.914 ± 0.002 Mixed95 $177.88 - 177.91$ 0.120 ± 0.006 111 ± 6 0.020 ± 0.0012 10.7 ± 0.7 0.839 ± 0.006 Mixed93 $177.92 - 177.96$ 0.164 ± 0.006 146 ± 7 0.018 ± 0.0007 8.9 ± 0.4 0.859 ± 0.006 Mixed95 $184.23 - 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 8.9 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ 0.022 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.1 0.976 ± 0.002 Flaming64 $188.07 - 188.09$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0008 6.6 ± 0.5 0.842 ± 0.012 Smolderi95 $188.10 - 188.16$ 0.109 ± 0.001 100 ± 2 0.008 ± 0.0001 $4.3 \pm \langle 0.1$ 0.902 ± 0.001 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.43 - 188.44$ 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.1 0.873 ± 0.002 Mixed36 $188.59 - 188.65$ 0.179 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 185 ± 2 0.012 ± 0.0001 6.9 ± 0.1 $0.879 \pm 0.$	27	94	177.27 - 177.31	0.060 ± 0.002	59 ± 2	0.004 ± 0.0001	2.3 ± 0.1	0.944 ± 0.002	Flaming
95 $177.88 - 177.91$ 0.120 ± 0.006 111 ± 6 0.020 ± 0.0012 10.7 ± 0.7 0.893 ± 0.006 Mixed93 $177.92 - 177.96$ 0.164 ± 0.006 146 ± 7 0.018 ± 0.0007 8.9 ± 0.4 0.859 ± 0.06 Mixed95 $184.23 - 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ $0.022 \pm 0.025 \pm 0.002$ 25 ± 2 0.002 ± 0.0001 1.2 ± 0.1 0.976 ± 0.002 Flaming64 $188.07 - 188.09$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0008 6.6 ± 0.5 0.842 ± 0.012 Smolderi95 $188.10 - 188.16$ 0.109 ± 0.001 102 ± 1 0.008 ± 0.0002 4.5 ± 0.1 0.904 ± 0.002 Mixed54 $188.22 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.23 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.40 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0002 6.1 ± 0.1 0.873 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0001 6.9 ± 0.1 0.860 ± 0.002 Mixed95 $188.45 - 188.48$ 0.163 ± 0.002 145 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.61 - 188.69$ 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 $0.$	28	95	177.80 - 177.83	0.094 ± 0.002	89 ± 2	0.008 ± 0.0001	4.1 ± 0.1	0.914 ± 0.002	Mixed
93 $177.92 - 177.96$ 0.164 ± 0.006 146 ± 7 0.018 ± 0.0007 8.9 ± 0.4 0.859 ± 0.006 Mixed95 $184.23 - 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 - 186.52$ 0.022 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.1 0.976 ± 0.002 Smolderi95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0002 4.5 ± 0.1 0.904 ± 0.002 Mixed54 $188.14 - 188.16$ 0.109 ± 0.001 102 ± 1 0.008 ± 0.0001 4.3 ± 0.1 0.902 ± 0.001 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.20 - 188.22$ 0.104 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.40 - 188.44$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0002 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.53 - 188.55$ 0.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.9 ± 0.3 0.884 ± 0.002 Mixed95 $188.46 - 188.66$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.873 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.848 ± 0.002 Mixed95 $188.71 - 188.74$ 0.133 ± 0.005 126 ± 5 0.0104 ± 0.0002 5.7 ± 0.3 0.874 ± 0.003	29	95	177.88 - 177.91	0.120 ± 0.006	111 ± 6	0.020 ± 0.0012	10.7 ± 0.7	0.893 ± 0.006	Mixed
95 $184.23 + 184.26$ 0.232 ± 0.014 196 ± 15 0.013 ± 0.0007 6.5 ± 0.4 0.811 ± 0.014 Smolderi80 $186.49 + 186.52$ 0.025 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.11 0.976 ± 0.002 Flaming95 $188.10 - 188.10$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0008 6.6 ± 0.5 0.842 ± 0.012 Smolderi95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0001 $4.3 \pm < 0.1$ 0.904 ± 0.002 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0001 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.20 - 188.22$ 0.104 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.40 - 188.44$ 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.55 - 188.65$ 0.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.67 - 188.65$ 0.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.67 - 188.65$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.884 ± 0.003 Smolderi95 $188.75 - 188.78$ 0.025 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.94	30	93	177.92 - 177.96	0.164 ± 0.006	146 ± 7	0.018 ± 0.0007	8.9 ± 0.4	0.859 ± 0.006	Mixed
80 $186.49 - 186.52$ 0.022 ± 0.002 25 ± 2 0.002 ± 0.0001 1.2 ± 0.1 0.976 ± 0.002 Flaming64 $188.07 - 188.09$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0008 6.6 ± 0.5 0.842 ± 0.012 Smolderi95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0002 $4.3 \pm < 0.1$ 0.904 ± 0.002 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.23 - 188.25$ 0.804 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.40 - 188.44$ 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.53 - 188.55$ 0.164 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.71 - 188.78$ 0.015 ± 0.003 5.4 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.879 ± 0.005 Mixed95 $188.71 - 188.78$ 0.055 ± 0.003 26 ± 5 0.0104 ± 0.0002 6.9 ± 0.1 0.848 ± 0.003 Smolderi95 $188.71 - 188.78$ 0.055 ± 0.003 24 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.0	31	95	184.23 - 184.26	0.232 ± 0.014	196 ± 15	0.013 ± 0.0007	6.5 ± 0.4	0.811 ± 0.014	Smolderin
64 $188.07 - 188.09$ 0.188 ± 0.012 165 ± 13 0.013 ± 0.0008 6.6 ± 0.5 0.842 ± 0.012 Smolderi95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0002 4.5 ± 0.1 0.904 ± 0.002 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.23 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.53 - 188.55$ 0.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95 $188.66 - 188.65$ 0.179 ± 0.002 158 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed95 $188.77 - 188.78$ 0.055 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming95 $188.77 - 188.83$ 0.272 ± 0.002 112 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.948 ± 0.002 </td <td>32</td> <td>80</td> <td>186.49 - 186.52</td> <td>0.025 ± 0.002</td> <td>25 ± 2</td> <td>0.002 ± 0.0001</td> <td>1.2 ± 0.1</td> <td>0.976 ± 0.002</td> <td>Flaming</td>	32	80	186.49 - 186.52	0.025 ± 0.002	25 ± 2	0.002 ± 0.0001	1.2 ± 0.1	0.976 ± 0.002	Flaming
95 $188.10 - 188.13$ 0.106 ± 0.002 100 ± 2 0.008 ± 0.0002 4.5 ± 0.1 0.904 ± 0.002 Mixed54 $188.14 - 188.16$ 0.109 ± 0.001 102 ± 1 0.008 ± 0.0001 $4.3 \pm <0.1$ 0.902 ± 0.001 Mixed64 $188.20 - 188.22$ 0.104 ± 0.004 90 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.23 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.40 - 188.44$ 0.134 ± 0.004 120 ± 4 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.012 ± 0.0001 6.0 ± 0.1 0.837 ± 0.002 Mixed36 $188.55 - 0.146 \pm 0.002$ 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.864 ± 0.002 Mixed95 $188.62 - 188.69$ 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.0104 ± 0.0002 3.3 ± 0.1 0.948 ± 0.000 Mixed95 $188.72 - 188.83$ 0.122 ± 0.0002 124 ± 2 0.009 ± 0.0001 4.8 ± 0.10 0.939 ± 0.002 Mixed95 $188.75 - 188.78$ 0.022 ± 0.002 124 ± 2 0.009 ± 0.0001 4.8 ± 0.10 0.939 ± 0.002 Mixed	33	64	188.07 - 188.09	0.188 ± 0.012	165 ± 13	0.013 ± 0.0008	6.6 ± 0.5	0.842 ± 0.012	Smolderin
54188.14188.16 0.109 ± 0.001 102 ± 1 0.008 ± 0.0001 $4.3 \pm < 0.1$ 0.902 ± 0.001 Mixed64188.20 188.22 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52188.23 188.25 0.80 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95188.40 188.44 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.002 Smolderi95188.45 188.48 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36188.59 188.65 0.163 ± 0.002 145 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95188.62 188.65 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.848 ± 0.002 Smolderi74188.66 188.69 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi95188.71 188.76 0.055 ± 0.003 54 ± 3 0.066 ± 0.002 3.3 ± 0.1 0.948 ± 0.003 Flaming95188.86 188.86 0.120 ± 0.002 112 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39188.86 188.86 0.120 ± 0.002 112 ± 2 0.007 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39188.86 188.85 $0.002 \pm 0.22 \pm 0.0001$ <t< td=""><td>34</td><td>95</td><td>188.10 - 188.13</td><td>0.106 ± 0.002</td><td>100 ± 2</td><td>0.008 ± 0.0002</td><td>4.5 ± 0.1</td><td>0.904 ± 0.002</td><td>Mixed</td></t<>	34	95	188.10 - 188.13	0.106 ± 0.002	100 ± 2	0.008 ± 0.0002	4.5 ± 0.1	0.904 ± 0.002	Mixed
64 $188.20 - 188.22$ 0.104 ± 0.004 99 ± 4 0.008 ± 0.0003 4.2 ± 0.2 0.906 ± 0.004 Mixed52 $188.23 - 188.25$ 0.080 ± 0.007 77 ± 7 0.006 ± 0.0004 3.2 ± 0.2 0.926 ± 0.007 Flaming95 $188.40 - 188.44$ 0.194 ± 0.003 169 ± 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.53 - 188.55$ 0.164 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.848 ± 0.002 Smolderi95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed95 $188.75 - 188.78$ 0.055 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming95 $188.79 - 188.83$ 0.272 ± 0.009 223 ± 10 0.012 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 <td>35</td> <td>54</td> <td>188.14 - 188.16</td> <td>0.109 ± 0.001</td> <td>102 ± 1</td> <td>0.008 ± 0.0001</td> <td>$4.3 \pm < 0.1$</td> <td>0.902 ± 0.001</td> <td>Mixed</td>	35	54	188.14 - 188.16	0.109 ± 0.001	102 ± 1	0.008 ± 0.0001	$4.3 \pm < 0.1$	0.902 ± 0.001	Mixed
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	36	64	188.20 - 188.22	0.104 ± 0.004	99 ± 4	0.008 ± 0.0003	4.2 ± 0.2	0.906 ± 0.004	Mixed
95188.40 0.194 ± 0.003 169 \pm 3 0.012 ± 0.0002 6.1 ± 0.1 0.837 ± 0.003 Smolderi95188.45 188.48 0.131 ± 0.004 120 \pm 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36188.55 1.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed54188.55 1.146 ± 0.002 145 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed95188.66 $1.866 + 0.179 \pm 0.002$ 158 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Smolderi74188.66 $1.866 + 0.186 + 0.021$ 1.83 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi95188.71 - 188.74 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed95188.75 - 188.78 0.025 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming95188.75 - 188.80 0.122 ± 0.009 2.03 ± 10 0.012 ± 0.0005 5.7 ± 0.3 0.786 ± 0.009 Mixed95188.84 - 188.85 0.120 ± 0.002 87 ± 2 0.007 ± 0.0001 4.8 ± 0.1 0.948 ± 0.002 Mixed95189.33 - 189.05 0.54 ± 0.012 139 ± 13 0.010 ± 0.0005 5.6 ± 0.3 0.871 ± 0.009 Mixed30189.34 - 189.35 0.009 ± 0.009 86 ± 9 0.006 ± 0.0005 3.2 ± 0.3 0.917 ± 0.009 Mixed30189	37	52	188.23 - 188.25	0.080 ± 0.007	77 ± 7	0.006 ± 0.0004	3.2 ± 0.2	0.926 ± 0.007	Flaming
95 $188.45 - 188.48$ 0.131 ± 0.004 120 ± 4 0.013 ± 0.0006 6.9 ± 0.3 0.884 ± 0.004 Mixed36 $188.53 - 188.55$ 0.146 ± 0.002 132 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.873 ± 0.002 Mixed54 $188.59 - 188.61$ 0.163 ± 0.002 145 ± 2 0.012 ± 0.0001 6.0 ± 0.1 0.860 ± 0.002 Mixed95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.848 ± 0.002 Smolderi74 $188.66 - 188.69$ 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed95 $188.79 - 188.78$ 0.055 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming95 $188.79 - 188.78$ 0.055 ± 0.002 122 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 Mixed30 $189.32 - 189.31$ 0.149 ± 0.008 135 ± 9 0.011 ± 0.0005 5.3 ± 0.5 0.867 ± 0.012 Mixed30 $189.49 - 189.35$ 0.099 ± 0.009 $1002 + 112 \pm 0.0007$ 5.1 ± 0.3 0.917 ± 0.009 Mixed </td <td>38</td> <td>95</td> <td>188.40 - 188.44</td> <td>0.194 ± 0.003</td> <td>169 ± 3</td> <td>0.012 ± 0.0002</td> <td>6.1 ± 0.1</td> <td>0.837 ± 0.003</td> <td>Smolderin</td>	38	95	188.40 - 188.44	0.194 ± 0.003	169 ± 3	0.012 ± 0.0002	6.1 ± 0.1	0.837 ± 0.003	Smolderin
36188.550.146 \pm 0.002132 \pm 20.012 \pm 0.00016.0 \pm 0.10.873 \pm 0.002Mixed54188.59 - 188.610.163 \pm 0.002145 \pm 20.012 \pm 0.00016.0 \pm 0.10.860 \pm 0.002Mixed95188.62 - 188.650.179 \pm 0.002158 \pm 20.014 \pm 0.00026.9 \pm 0.10.864 \pm 0.002Smolderi74188.66 - 188.690.214 \pm 0.011183 \pm 120.015 \pm 0.00087.4 \pm 0.50.824 \pm 0.011Smolderi95188.71 - 188.740.138 \pm 0.005126 \pm 50.010 \pm 0.00045.1 \pm 0.20.879 \pm 0.005Mixed95188.75 - 188.780.055 \pm 0.00354 \pm 30.066 \pm 0.0023.3 \pm 0.10.948 \pm 0.003Flaming95188.79 - 188.830.272 \pm 0.009223 \pm 100.012 \pm 0.00055.7 \pm 0.30.786 \pm 0.009Smolderi52188.84 - 188.870.091 \pm 0.002112 \pm 20.009 \pm 0.00014.8 \pm 0.10.893 \pm 0.002Mixed39188.86 - 188.870.091 \pm 0.00287 \pm 20.007 \pm 0.00014.0 \pm 0.10.916 \pm 0.002Mixed30189.32 - 189.050.154 \pm 0.012139 \pm 130.010 \pm 0.00085.3 \pm 0.50.867 \pm 0.012Mixed30189.34 - 189.350.090 \pm 0.00986 \pm 90.006 \pm 0.00053.2 \pm 0.30.917 \pm 0.009Mixed30189.34 - 189.350.090 \pm 0.00986 \pm 90.006 \pm 0.0076.1 \pm 0.40.858 \pm 0.009Mixed30	19	95	188.45 - 188.48	0.131 ± 0.004	120 ± 4	0.013 ± 0.0006	6.9 ± 0.3	0.884 ± 0.004	Mixed
54 $188.59 - 188.61$ 0.163 ± 0.002 145 ± 2 0.012 ± 0.0001 6.3 ± 0.1 0.860 ± 0.002 Mixed 95 $188.62 - 188.65$ 0.179 ± 0.002 158 ± 2 0.014 ± 0.0002 6.9 ± 0.1 0.848 ± 0.002 Smolderi 74 $188.66 - 188.69$ 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi 95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed 95 $188.79 - 188.78$ 0.55 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming 95 $188.79 - 188.83$ 0.272 ± 0.009 223 ± 10 0.012 ± 0.0005 5.7 ± 0.3 0.786 ± 0.002 Mixed 39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.8 ± 0.1 0.933 ± 0.002 Mixed 95 $189.03 - 189.05$ 0.154 ± 0.012 139 ± 13 0.010 ± 0.0005 5.6 ± 0.3 0.871 ± 0.008 Mixed 30 $189.34 - 189.35$ 0.099 ± 0.09 86 ± 9 0.00	10	36	188.53 - 188.55	0.146 ± 0.002	132 ± 2	0.012 ± 0.0001	6.0 ± 0.1	0.873 ± 0.002	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	1	54	188.59 - 188.61	0.163 ± 0.002	145 ± 2	0.012 ± 0.0001	6.3 ± 0.1	0.860 ± 0.002	Mixed
74 $188.66 - 188.69$ 0.214 ± 0.011 183 ± 12 0.015 ± 0.0008 7.4 ± 0.5 0.824 ± 0.011 Smolderi 95 $188.71 - 188.74$ 0.138 ± 0.005 126 ± 5 0.010 ± 0.0004 5.1 ± 0.2 0.879 ± 0.005 Mixed 95 $188.75 - 188.78$ 0.055 ± 0.003 54 ± 3 0.006 ± 0.0002 3.3 ± 0.1 0.948 ± 0.003 Flaming 95 $188.79 - 188.83$ 0.272 ± 0.009 223 ± 10 0.012 ± 0.0005 5.7 ± 0.3 0.786 ± 0.009 Smolderi 52 $188.84 - 188.85$ 0.120 ± 0.002 112 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed 39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 Mixed 95 $189.03 - 189.05$ 0.154 ± 0.012 139 ± 13 0.010 ± 0.0008 5.3 ± 0.5 0.867 ± 0.012 Mixed 30 $189.34 - 189.35$ 0.090 ± 0.099 86 ± 9 0.006 ± 0.0005 3.2 ± 0.3 0.917 ± 0.009 Mixed 30 $189.49 - 189.52$ 0.165 ± 0.009 147 ± 9 <	12	95	188.62 - 188.65	0.179 ± 0.002	158 ± 2	0.014 ± 0.0002	6.9 ± 0.1	0.848 ± 0.002	Smolderin
95 $166, 1+168, 1+60, 1$	13 14	74	188.66 - 188.69	0.214 ± 0.011	183 ± 12 126 + 5	0.015 ± 0.0008	1.4 ± 0.5	0.824 ± 0.011	Smolderin
95 $168.79 - 188.76$ 0.035 ± 0.003 54 ± 3 0.006 ± 0.0002 3.5 ± 0.1 0.948 ± 0.003 $114ming$ 95 $188.79 - 188.83$ 0.272 ± 0.009 223 ± 10 0.012 ± 0.0005 5.7 ± 0.3 0.786 ± 0.009 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 Mixed59 $189.03 - 189.05$ 0.154 ± 0.012 139 ± 13 0.010 ± 0.0008 5.3 ± 0.5 0.867 ± 0.012 Mixed30 $189.34 - 189.35$ 0.090 ± 0.009 86 ± 9 0.006 ± 0.0005 3.2 ± 0.3 0.917 ± 0.009 Mixed30 $189.49 - 189.52$ 0.165 ± 0.009 147 ± 9 0.012 ± 0.0007 6.1 ± 0.44 0.858 ± 0.009 Mixed48 $195.10 - 195.12$ 0.212 ± 0.017 215 ± 28 0.020 ± 0.0020 9.5 ± 1.2 0.792 ± 0.026 Smolderi95 $195.14 - 195.17$ 0.140 ± 0.007 128 ± 8 0.010 ± 0.0026 5.5 ± 0.3 0.877 ± 0.007 Mixed40 0.072 ± 0.027 125 ± 28 0.010 ± 0.0026 5.5 ± 0.3 0.877 ± 0.029 Mixed95 $195.14 - 195.17$ 0.140 ± 0.007 128 ± 8 0.010 ± 0.0026 5.5 ± 0.3 0.877 ± 0.029	14 1 <i>2</i>	93 05	188./1 - 188./4	0.138 ± 0.003	120 ± 3	0.010 ± 0.0004	3.1 ± 0.2	0.879 ± 0.003	Flamin
52 $166, 72 + 168, 65$ 0.212 ± 0.009 22.5 ± 10 0.012 ± 0.0005 5.7 ± 0.5 0.860 ± 0.009 Smolderi 52 $188.84 - 188.85$ 0.120 ± 0.002 112 ± 2 0.009 ± 0.0001 4.8 ± 0.1 0.893 ± 0.002 Mixed 39 $188.86 - 188.87$ 0.091 ± 0.002 87 ± 2 0.007 ± 0.0001 4.0 ± 0.1 0.916 ± 0.002 Mixed 59 $189.03 - 189.05$ 0.154 ± 0.012 139 ± 13 0.010 ± 0.0008 5.3 ± 0.5 0.867 ± 0.012 Mixed 95 $189.27 - 189.31$ 0.149 ± 0.008 135 ± 9 0.011 ± 0.0005 5.6 ± 0.3 0.871 ± 0.009 Mixed 30 $189.34 - 189.35$ 0.090 ± 0.009 86 ± 9 0.006 ± 0.0005 5.2 ± 0.3 0.917 ± 0.009 Mixed 89 $189.49 - 189.52$ 0.165 ± 0.009 147 ± 9 0.112 ± 0.0007 6.1 ± 0.4 0.858 ± 0.009 Mixed 48 $195.10 - 195.12$ 0.212 ± 0.017 215 ± 28 0.020 ± 0.0020 9.5 ± 1.2 0.792 ± 0.026 Smolderi 37 $195.12 - 195.13$ 0.262 ± 0.027 215 ± 28 0.010 ± 0.0006 5.5 ± 0.3 0.877 ± 0.007 Mixed 95 $195.14 - 195.17$ 0.140 ± 0.007 128 ± 8 0.010 ± 0.0020 9.5 ± 1.8 0.877 ± 0.029 95 $195.14 - 195.17$ 0.140 ± 0.071 127 ± 40 0.010 ± 0.0038 5.3 ± 1.8 0.877 ± 0.029	13 16	95	188.75 - 188.78	0.055 ± 0.003	54 ± 5	0.006 ± 0.0002	5.5 ± 0.1	0.948 ± 0.003	Flaming
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	17	93 52	100./9 - 100.83	0.272 ± 0.009 0.120 \pm 0.002	223 ± 10 112 ± 2	0.012 ± 0.0005 0.000 ± 0.0001	3.7 ± 0.3 4.8 ± 0.1	0.780 ± 0.009 0.802 ± 0.002	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	10	20	100.04 - 100.03	0.120 ± 0.002	112 ± 2	0.009 ± 0.0001	-1.0 ± 0.1	0.095 ± 0.002	Mixed
95 169,05	10 10	39 50	100.00 - 100.07	0.091 ± 0.002 0.154 ± 0.012	$0/\pm 2$ 130 + 12	0.007 ± 0.0001 0.010 ± 0.0002	4.0 ± 0.1 5 3 ± 0.5	0.910 ± 0.002 0.867 \pm 0.012	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	50	95	189.05 - 189.05	$0.13 + \pm 0.012$ 0.149 + 0.008	139 ± 13 135 ± 9	0.010 ± 0.0008 0.011 ± 0.0005	5.5 ± 0.5 5.6 ± 0.3	0.807 ± 0.012 0.871 + 0.008	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	51	30	180 34 180 25	0.149 ± 0.008 0.000 ± 0.000	155±9 86±9	0.011 ± 0.0005	3.0 ± 0.3 3.2 ± 0.2	0.071 ± 0.008	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	52	80	107.34 - 107.33	0.090 ± 0.009 0.165 ± 0.009	147 ± 9	0.000 ± 0.0003 0.012 ± 0.0007	5.2 ± 0.5 6.1 ± 0.4	0.917 ± 0.009 0.858 ± 0.009	Mixed
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	3	48	109.49 = 109.32 105.10 = 105.12	0.103 ± 0.009 0.212 ± 0.010	$1 + 7 \pm 7$ 181 + 20	0.012 ± 0.0007 0.016 ± 0.0014	8.1 ± 0.4 8.0 ± 0.9	0.825 ± 0.009	Smolderin
95 $195.14 - 195.17$ 0.140 ± 0.007 128 ± 8 0.010 ± 0.0006 5.5 ± 0.3 0.877 ± 0.020 Mixed an 0.142 ± 0.051 127 ± 40 0.010 ± 0.0006 5.5 ± 0.3 0.877 ± 0.020 Mixed	4	37	195 12 - 195 13	0.262 ± 0.019	215 ± 20	0.010 ± 0.0014 0.020 ± 0.0020	95 ± 12	0.025 ± 0.018 0.792 ± 0.026	Smolderin
an $0.142 + 0.051$ $127 + 40$ $0.010 + 0.0038$ $5.3 + 1.8$ $0.879 + 0.020$	5	95	195.14 - 195.17	0.140 ± 0.007	128 ± 8	0.010 ± 0.00020	5.5 ± 0.3	0.877 ± 0.007	Mixed
	Mean			0.142 ± 0.051	127 + 40	0.010 ± 0.0038	53 + 18	0.878 ± 0.039	

Table 2. Events of elevated trace gas concentrations at the CRV tower due to fire emissions. Columns show the number of 30 s

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measurements used to calculate emission factors for each event (N), the time of the event, emission ratios (ppmv ppmv⁻¹), emission

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factors (g per kg of dry biomass combusted), and modified combustion efficiency (MCE). Dominant combustion process (CP) is

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described as flaming, mixed, or smoldering.

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-	Fire Name	Distance (km)	Contribution (%)	Total Hectares	Fuel Type	Ignition Sou	urce		
1	Tozitna	229	10.74	31652	Black Spruce	Lightning	5		Formatted: Justified, Line spacing: 1.5 lines
2	Kobe	119	7.20	3444	Black Spruce	Lightning			
3	Blair	82	6.31	15217	Black Spruce	Lightning			
4	Aggie Creek	41	5.63	12829	Black Spruce	Lightning			
5	Spicer Creek	195	5.30	39761	Black Spruce	Lightning			
6	Blind River	252	3.87	24608	Black Spruce	Lightning			
7	Holtnakatna	404	3.44	90308	Mixed	Lightning	10		
8	Blazo	514	3.39	49106	Black Spruce	Lightning	10		
9	Big Creek 2	351	3.23	126637	Black Spruce	Lightning			
10	Chitanana River	241	3.12	17483	Black Spruce	Lightning			
11	Sea	309	3.06	172	Black Spruce	Human			
12	Sushgitit Hills	276	2.92	111712	Black Spruce	Lightning			
13	Big Mud River 1	254	2.72	42076	Black Spruce	Lightning			
14	Lost River	347	2.58	21088	Black Spruce	Lightning	15		
15	Munsatli 2	302	2.36	40682	Black Spruce	Lightning			
16	FWA Small Arms	19		740	Black Spruce	Prescribed			
	Complex		2.31						
17	Tobatokh	280	2.24	21868	Black Spruce	Lightning			
18	Trail Creek	363	2.24	11939	Black Spruce	Lightning			
19	Lloyd	201	2.22	26818	Black Spruce	Lightning	20		
20	Isahultila	342	2.17	60445	Black Spruce	Lightning	20		
21	Nulato	499	2.17	449	Black Spruce	Lightning			
22	Three Day	472	2.17	39378	Black Spruce	Lightning			
23	Hay Slough	188	1.90	37007	Black Spruce	Lightning			
24	Rock	316	1.83	3714	Other	Lightning			
25	Sulukna	329	1.77	6760	Black Spruce	Lightning			
26	Titna	273	1.77	12415	Black Spruce	Lightning	25		
27	Quinn Creek	657	1.49	2002	Other	Lightning			
28	Harper Bend	188	1.45	17555	Black Spruce	Lightning			
29	Hard Luck	328	1.43	5230	Black Spruce	Lightning			
30	Fox Creek	369	1.42	2346	Black Spruce	Lightning			
31	Bering Creek	280	1.36	45654	Black Spruce	Lightning			
32	Eden Creek	324	1.16	18614	Black Spruce	Lightning	•		
33	Falco	390	1.10	1817	Mixed	Lightning	30		
34	Jackson	202	1.00	2969	Black Spruce	Lightning			
35	Dulbi River	404	0.95	22057	Black Spruce	Lightning			

Table 3. All fires that contributed to at least 1% of the total CO anomaly observed at CRV tower ordered by largest CO contribution. The distance column represents the distance of the center of the fire perimeter to CRV tower. Contribution is the 35 percent contribution to the total integral of fire CO at CRV for the entire 2015 fire season. Some fires were grouped together if

they were inside the same 0.5° grid cell during model coupling. For those cases, individual fire contribution to the CO anomaly observed at CRV tower was weighted based on fire size.

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