

Responses to Reviewer #1 Comments

Emission factors from combustion are very important to emission inventory and air quality modeling studies. The authors carried out a series of experiments and reported the gaseous, PM_{2.5} mass and speciated emission factors from peat combustion. Overall, the experimental methods are reasonable and the data are robust. After the following questions have been well addressed, it is suitable for publishing.

- 1. Although the authors simply described the apparatus including the combustion chamber and the instruments used in this work. More details should be given, in particular, the characterization details about the combustion chamber. One of my most important concerns is the wall loss for these gaseous and particle phase pollutants in the chamber. Did you consider the wall loss correction when calculating the emission factors?*

Response (including Parts A and B):

- **Part A:** The Experimental setup section is reorganized to streamline the flow of description with the addition of the following description (Lines 124-128):

A blower supplied filtered air near the bottom of the chamber. The ventilation rate was controlled by the blower and exhaust fan at $\sim 2.65 \text{ m}^3 \text{ min}^{-1}$, resulting in a smoke residence time of ~ 3 min in the chamber assuming a well-stirred flow model.

- **Part B:** The reported EFs in this study represent the upper limit estimates as wall losses were not corrected. The turbulent diffusion and gravitational settling need to be considered while estimating the wall losses. Theoretically, wall losses can occur inside the combustion chamber, in the exhaust stack, sampling lines, and inside the OFR, but it is difficult to quantify given the variabilities in fuel composition and combustion conditions. For this study, estimated wall losses for the most sticky gas (i.e., NH₃) is at most $< 14 \%$, which will be lower for less sticky gases. The particle wall losses depend on particle sizes: $< 5 \%$ for $10 \text{ nm} - 1 \text{ }\mu\text{m}$ and $\sim 10 \%$ from $2.5 \text{ }\mu\text{m}$ particles. Overall, these gaseous and particle wall losses are $< 15 \%$, well within the measurement uncertainties of $\pm 30 \%$ for air quality modeling. A new section (2.5 Estimation of wall losses) is added to address this (Lines 241-264):

Gas and particle wall losses can result in some underestimation of measured EFs, but it is well within the measurement uncertainties of $\pm 15 \%$. Losses can occur inside the combustion chamber, in the exhaust stack, sampling lines, and inside the OFR. Due to the low surface-to-volume ratio of the chamber (2.9 m^{-1}) and short residence time (~ 3 min) in this study, the gas and particle losses are expected to be low in the combustion chamber. Grosjean (1985) estimated an NH₃ loss rate of $4-17 \times 10^{-4} \text{ min}^{-1}$ in a small Teflon chamber (3.9 m^3) with a surface-to-volume ratio of 3.8 m^{-1} , resulting in $< 0.5 \%$ NH₃ wall loss. Even though the NH₃ accommodation coefficient might be higher for aluminum than Teflon (Neuman et al., 1999), the chamber wall loss in this study is expected to be $< 5 \%$ for NH₃. To reduce wall losses of sticky gases, the FTIR sampled exhaust gas from the stack without dilution, as shown in Fig. 2. Approximately 9% NH₃ would encounter the stack wall due to turbulent diffusion (Hinds,

1999). The maximum NH_3 loss in the stack is <9 % and the maximum overall NH_3 loss is <14 %. Losses of less sticky gases would be lower.

The wall loss rates by McMurry and Grosjean (1985) and Wang et al. (2018) indicate <5 % particle number losses for 10 nm–2.5 μm in a similar chamber. Particle losses by turbulent diffusion in the stack are also negligible (<0.5 %). For a 2 m-long horizontal heated sampling line in this study (Fig. 2), particle losses by diffusion and gravitational settling are negligible (<0.1 %) for 10 nm - 1 μm particles and ~6 % for 2.5 μm particles. Earlier measurements showed that the dilution tunnel had ~100% penetration for 0.5-5 μm particles (Wang et al., 2012). Therefore, maximum particle losses in this study are estimated to be <5 % for 10 nm - 1 μm and <10 % for 2.5 μm . Past studies (Bhattarai et al., 2018; Karjalainen et al., 2016; Lambe et al., 2011) showed that particle number losses through the OFR may be ~50 % for 20 nm and <10 % for >100 nm particles, with a negligible effect on mass concentration.

2. *As for C2-C6 VOCs, HCN and NH_3 measurements with FTIR, the IR bands for quantifying each species and the details about the quantification methods should be described in the text and tables. It is better to show the typical IR spectrum for these species in the supplement materials. As for NH_3 , it is a sticky molecule and easily interfered by human activity as pointed out by the authors (line 350). How did you consider these factors on NH_3 measurement? In addition, it is necessary to do uncertainty analysis to all of the measured EFs.*

Response (including Parts A, B, and C):

- **Part A:** The following description about the FTIR was added (Lines 148-153), along with example spectra of six reference gases (i.e., HCN, NH_3 , ethene, 1,3-butadiene, hexane, and benzene) and an Everglades (FL) peat sample spectrum in the new Figure S3:

An exhaust gas sample was drawn into the FTIR and the infrared (IR) absorption spectra in the wave number range of 900 – 4200 cm^{-1} were measured. The instrument software compares the measured absorption spectra with reference gas absorption spectra in the calibration library to identify gas species and calculate concentrations. Examples of reference gas spectra and an Everglades peat sample spectrum are plotted in Fig. S3.

- **Part B:** To reduce NH_3 contamination by background air and human, activities inside the combustion chamber are minimized to only those necessary and the chamber was purged before starting the experiment. During the experiments, the door of the combustion chamber was closed, and intake air passed through a charcoal bed to remove VOCs and a high-efficiency particulate air (HEPA) filter to provide particle-free clean air for dilution. For this study, EF_{NH_3} , from an impregnated filter are used as discussed in Section 3.2.2 (Lines 404-410):

Figure S9a shows some scatter in EF_{NH_3} determined by FTIR and the impregnated filter, especially at high concentrations. The regression slope shows that EF_{NH_3} by the FTIR was ~22 % lower than that of filters with a correlation coefficient of 0.76. Variable baselines in

the FTIR measurements along with some nitrogen content in the diluted air and breath NH_3 (Hibbard and Killard, 2011) in the testing laboratory may have contributed to these variations. The impregnated filter collects all of the NH_3 over the sampling period, including amounts that are below the FTIR detection limits, so it is probably better representing the time-integrated EF_{NH_3} .

- **Part C:** For this study, EF uncertainties are reported as standard deviations of multiple runs for each peat. These uncertainties are shown in Tables 2-4, Supplemental Tables S4-S7, Figures S4, S6, and S12.

3. *Most of these results were shown in tables. It is somewhat difficult to follow. For example, when discussing the influence of aging and the moisture of peat on the EFs, it is more easy to understand their differences if showing in figures. In addition, when comparing the measured EFs with the literature results, it is better to discuss the reasons why you obtained a different value. For example, the measured $\text{EF}(\text{CO}_2)$ from this work is lower than that in literature. Some objective comments should be given in the text.*

Response:

- Tables are useful to examine quantitative differences among peat types and to compare with other studies; however, we agree with the Reviewer that graphic presentations are more intuitive for readers. We have added Figure S4 for EFs of CO , CO_2 , and CH_4 based on Table S4; Figure S6 for EFs of NH_3 , HCN , NO , NO_2 , NO_x , and N_2O based on Table S5; Figure S10 for comparison of fresh vs. aged EFs for $\text{PM}_{2.5}$ mass and carbonaceous compounds based on Table 6; and Fig. S12 for comparison of WSOC and OC.

4. *Even for the same pollutant, EF varied obviously among different samples. Was there a quantitative relationship between the EFs and the element composition in the peat samples or combustion conditions?*

Response:

- Indeed, EFs varied by different test runs and by peat types. The linear relationship between the EFs and full elemental composition are more apparent for the sum of gaseous and particulate carbon, and for EF_{NO} , with inverse association between EF_{CH_4} and fuel oxygen contents. To illustrate the relationship between EFs and peat fuel content, a new Figure S5 (including Figs. S5a–i) is added to discuss the association between EFs and fuel elemental composition. The following discussions were added to the text:

- **Lines 352-358:**

Emission factors depends on both fuel composition and combustion conditions. Figure S5a shows that total measured gas and particle carbon increases with fuel carbon content for the six types of peat. EF_{CO_2} increases with fuel carbon content (Fig. S5b) except for the Putnam (FL) peat, which has the highest fuel carbon (56.6%) but low EF_{CO_2} . It has high EF_{CO} and EF_{TC} (Figs. S5c-d), consistent with its low MCE (0.65 ± 0.04). EF_{CO} and EF_{TC} do not have a clear

trend with fuel carbon content; however, EF_{CH_4} increases with fuel carbon (Fig. S5e) but decreases with fuel oxygen content (Fig. S5f).

- **Lines 373-375:**

Figure S5g shows that EF_{NO} increases with fuel nitrogen content, while EF_{NO_2} is not dependent on fuel nitrogen content (Fig. S5h). Because EF_{NO} is higher than EF_{NO_2} , EF_{NO_x} and EF_{NO_y} also increase with fuel nitrogen content (not shown).

- **Lines 396-398:**

EF_{NH_3} (0.4–8.3 g kg⁻¹) are of the same magnitude as EF_{HCN} (Fig. S6a) and independent of fuel nitrogen content (Fig. S5i) except for the Everglades (FL) peat (9–18 g kg⁻¹) which has the highest fuel nitrogen content.

5. *For OFP experiments, did you consider the OH suppression?*

Response:

- The average CO and NO_x concentrations upstream of the OFR are 6.3 ppm and 34 ppb, respectively, corresponding to external OH reactivity (OHR) of 45 s⁻¹. The OFR used in this study is a commercial product from Aerodyne and its photon flux ratio at 185nm/254 nm has not been fully characterized (<https://sites.google.com/site/pamwiki/hardware/estimation-equations>). Therefore, the OH exposure equations derived based on the "Penn State" lamps (Li et al., 2015; Peng et al., 2015; Peng et al., 2016) are not applicable to this OFR. The OH exposures based on calibration using 500 ppb SO₂ represent an upper limit of the actual OH exposures inside the OFR. The following sentence is added to the text (**Lines 163-165**):

Due to external OH reactivity from CO, NO_x, and other reactants, these OH_{exp} levels represent upper limits of the actual OH exposures inside the OFR (Li et al., 2015; Peng et al., 2015).

6. *In the introduction, the previous relevant researches before this work should be well reviewed.*

Response:

- This paper pioneers the EF comparisons between fresh and aged multipollutant mixtures through laboratory controlled burns with atmospheric aging simulated by an oxidation flow reactor. Relevant past studies for both in-situ field and laboratory peat combustion have been reviewed and cited with a total of 31 references. Where appropriate, the EFs for peat burning are compared throughout the text. Since most peat burning measurements were conducted in Southeast Asia, the following sentence and references are added in the "Introduction" section (**Lines 69-73**):

For Southeast Asia, fire-related regional air pollution and its effects on atmospheric visibility, forest ecosystem, and human health have been addressed in many studies (e.g., Behera et al., 2014;Betha et al., 2013;Bin Abas et al., 2004;Dall'Osto et al., 2014;Engling et al., 2014;Fujii et al., 2017;Heil and Goldammer, 2001;Hu et al., 2019;Kundu et al., 2010;Levine, 1999;Tham et al., 2019).

7. *Table S7 was missed.*

Response:

- Table S7 is now included.

References:

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Table S7. Individual and averaged emission factors for fresh vs. aged PM_{2.5} mass and carbon.

Peat Type	Sample ID	Emission Factors in g/kg																		
		EF _{PM2.5}		EF _{OC}		EF _{EC}		EF _{TC}		EF _{WSOC}		EF _{Levoglucosan}		WSOC/OC		WSOC/PM _{2.5}		OC/PM _{2.5}		
		FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	FRESH ^a	AGED ^b	
Odintsovo, Russia (n=6)	PEAT030	44.937	39.709	27.157	20.511	1.057	0.629	28.214	21.140	17.753	13.680	19.703	12.880	65.37%	66.70%	39.51%	34.45%	60.43%	51.65%	
	PEAT031	42.173	33.042	25.645	16.629	0.160	0.611	25.805	17.240	14.391	10.785	14.330	9.059	56.11%	64.86%	34.12%	32.64%	60.81%	50.33%	
	PEAT032	43.117	46.130	22.248	15.632	1.177	0.521	23.425	16.153	16.075	13.064	12.174	6.922	72.26%	83.58%	37.28%	28.32%	51.60%	33.89%	
	PEAT033	40.491	47.894	26.964	16.901	0.515	1.011	27.479	17.912	15.453	11.415	16.263	7.612	57.31%	67.54%	38.16%	23.83%	66.59%	35.29%	
	PEAT034	50.522	45.404	28.736	20.008	0.822	0.532	29.558	20.540	16.962	12.552	18.328	8.660	59.03%	62.73%	33.57%	27.64%	56.88%	44.07%	
	PEAT035	34.649	31.065	20.019	13.467	0.900	0.823	20.919	14.290	12.320	11.234	13.704	7.432	61.54%	83.42%	35.56%	36.16%	57.78%	43.35%	
Average ± SD		42.65 ± 5.22	40.54 ± 7.15	25.13 ± 3.32	17.19 ± 2.67	0.77 ± 0.38	0.69 ± 0.19	25.90 ± 3.23	17.88 ± 2.61	15.49 ± 1.94	12.12 ± 1.15	15.75 ± 2.88	8.76 ± 2.17	61.94 ± 6.04%	71.47 ± 9.46%	36.37 ± 2.34%	30.51 ± 4.68%	59.01 ± 4.97%	43.10 ± 7.38%	
C.o.V.		12.24%	17.64%	13.22%	15.53%	48.66%	27.92%	12.47%	14.58%	12.55%	9.46%	18.30%	24.77%	9.75%	13.24%	6.44%	15.35%	8.43%	17.12%	
Pskov, Siberia (n=6)	PEAT023	39.719	39.648	27.252	23.769	1.200	1.289	28.452	25.058	8.901	9.395	1.615	1.594	32.66%	39.53%	22.41%	23.70%	68.61%	59.95%	
	PEAT025	32.975	27.337	26.845	16.698	0.844	0.598	27.688	17.296	7.647	7.736	3.041	1.023	28.49%	46.33%	23.19%	28.30%	81.41%	61.08%	
	PEAT026	34.739	21.511	24.636	17.981	1.431	0.824	26.067	18.805	9.003	8.094	2.327	1.048	36.54%	45.02%	25.91%	37.63%	70.92%	83.59%	
	PEAT027	27.468	18.934	21.274	11.310	0.000	0.459	21.274	11.769	9.065	7.385	2.358	0.719	42.61%	65.30%	33.00%	39.00%	77.45%	59.73%	
	PEAT028	42.074	45.032	31.372	22.388	0.069	0.949	31.441	23.337	na ^b	9.580	3.301	1.555	na ^b	42.79%	na ^b	21.27%	74.57%	49.72%	
	PEAT029	26.547	31.524	24.397	16.274	0.620	0.535	25.017	16.809	8.494	9.258	3.120	1.309	34.82%	56.89%	32.00%	29.37%	91.90%	51.62%	
Average ± SD		33.92 ± 6.29	30.66 ± 10.20	25.96 ± 3.40	18.07 ± 4.52	0.69 ± 0.58	0.78 ± 0.31	26.66 ± 3.44	18.85 ± 4.80	8.62 ± 0.59	8.57 ± 0.95	2.63 ± 0.64	1.21 ± 0.34	35.02 ± 5.20%	49.31 ± 9.78%	27.30 ± 4.93%	29.88 ± 7.19%	77.48 ± 8.41%	60.95 ± 12.07%	
C.o.V.		18.54%	33.27%	13.11%	24.99%	84.02%	40.19%	12.91%	25.49%	6.83%	11.06%	24.41%	28.15%	14.85%	19.84%	18.06%	24.06%	10.86%	19.80%	
Northern Alaska, USA (n=5)	PEAT013	17.305	16.320	14.619	13.122	0.543	0.338	15.162	13.460	6.877	5.314	1.790	2.269	47.04%	40.50%	39.74%	32.56%	84.88%	80.41%	
	PEAT014	30.090	27.119	22.257	17.531	0.640	0.884	22.897	18.415	7.294	7.413	4.297	4.195	32.77%	42.29%	24.24%	27.34%	64.64%	61.08%	
	PEAT019	27.733	29.102	20.459	18.942	0.272	0.378	20.731	19.319	6.651	7.321	8.043	6.285	32.51%	38.65%	23.98%	25.16%	73.77%	65.09%	
	PEAT020	30.406	28.527	17.196	15.718	0.932	1.090	18.127	16.808	7.190	8.167	2.726	1.971	41.81%	51.96%	23.65%	28.63%	56.55%	55.10%	
	PEAT022	14.390	23.086	12.270	9.053	0.593	0.063	12.863	9.116	5.686	5.287	1.525	0.652	46.34%	58.40%	39.51%	22.90%	85.27%	39.21%	
	Average ± SD		23.98 ± 7.57	24.83 ± 5.31	17.36 ± 4.09	14.87 ± 3.91	0.60 ± 0.24	0.55 ± 0.42	17.96 ± 4.06	15.42 ± 4.17	6.74 ± 0.64	6.70 ± 1.32	3.68 ± 2.67	3.07 ± 2.20	40.09 ± 7.10%	46.36 ± 8.47%	30.22 ± 8.59%	27.32 ± 3.65%	74.81 ± 11.60%	60.89 ± 15.13%
C.o.V.		31.56%	21.37%	23.58%	26.31%	39.58%	76.78%	22.60%	27.06%	9.52%	19.69%	72.67%	71.44%	17.70%	18.26%	28.41%	13.38%	15.50%	24.85%	
Putnam County Lakebed, Florida (n=4)	PEAT008	57.197	54.119	37.217	39.897	1.314	1.064	38.531	40.961	9.282	10.598	1.807	1.487	24.94%	26.56%	16.23%	19.58%	65.07%	73.72%	
	PEAT009	46.012	42.248	36.055	28.139	2.177	0.923	38.232	29.062	10.505	10.919	1.448	1.185	29.13%	38.80%	22.83%	25.84%	78.36%	66.60%	
	PEAT005	48.798	57.969	34.423	37.209	0.922	0.909	35.345	38.119	8.369	14.017	1.439	0.659	24.31%	37.67%	17.15%	24.18%	70.54%	64.19%	
	PEAT006	60.509	61.350	38.850	43.910	0.898	0.913	39.748	44.823	9.380	13.576	1.991	1.133	24.14%	30.92%	15.50%	22.13%	64.20%	71.57%	
	Average ± SD		53.13 ± 6.84	53.92 ± 8.32	36.64 ± 1.87	37.29 ± 6.69	1.33 ± 0.60	0.95 ± 0.07	37.96 ± 1.86	38.24 ± 6.71	9.38 ± 0.87	12.28 ± 1.77	1.67 ± 0.27	1.12 ± 0.34	25.63 ± 2.36%	33.49 ± 5.78%	17.93 ± 3.34%	22.93 ± 2.70%	69.54 ± 6.51%	69.02 ± 4.39%
	C.o.V.		12.88%	15.44%	5.10%	17.95%	44.99%	7.84%	4.91%	17.54%	9.32%	14.40%	16.38%	30.68%	9.21%	17.27%	18.62%	11.78%	9.37%	6.36%
Everglades National Park, Florida (n=7)	PEAT010	18.275	34.139	17.884	15.235	1.182	0.449	19.066	15.684	7.744	8.662	0.502	0.000	43.30%	56.86%	42.38%	25.37%	97.86%	44.63%	
	PEAT011	28.566	25.526	15.597	16.153	1.260	0.569	16.857	16.722	6.547	6.783	0.452	0.311	41.98%	41.99%	22.92%	26.57%	54.60%	63.28%	
	PEAT012	16.258	15.327	12.117	9.890	0.474	0.376	12.591	10.266	4.528	4.451	0.000	0.000	37.37%	45.01%	27.85%	29.04%	74.53%	64.53%	
	PEAT015	29.133	28.619	25.568	22.561	1.110	0.670	26.678	23.230	9.816	9.891	0.000	0.635	38.39%	43.84%	33.69%	34.56%	87.77%	78.83%	
	PEAT016	21.566	38.113	19.459	20.460	0.696	1.279	20.155	21.739	8.001	9.232	0.480	0.000	41.12%	45.12%	37.10%	24.22%	90.23%	53.68%	
	PEAT017	24.871	48.690	21.387	23.115	0.752	0.615	22.139	23.730	7.153	9.802	0.389	0.154	33.44%	42.41%	28.76%	20.13%	85.99%	47.47%	
PEAT018	26.755	43.879	21.147	23.937	0.000	0.727	21.147	24.664	9.934	11.183	0.772	0.673	46.97%	46.72%	37.13%	25.49%	79.04%	54.55%		
Average ± SD		23.63 ± 5.05	33.47 ± 11.39	19.02 ± 4.36	18.76 ± 5.18	0.78 ± 0.45	0.67 ± 0.30	19.80 ± 4.39	19.43 ± 5.34	7.67 ± 1.88	8.57 ± 2.26	0.37 ± 0.28	0.25 ± 0.30	40.37 ± 4.40%	45.99 ± 5.06%	32.83 ± 6.69%	26.48 ± 4.46%	81.43 ± 14.03%	58.14 ± 11.72%	
C.o.V.		21.38%	34.02%	22.90%	27.61%	57.45%	44.12%	22.19%	27.47%	24.52%	26.39%	75.66%	117.11%	10.90%	11.01%	20.36%	16.85%	17.23%	20.16%	
Borneo, Malaysia (n=4)	PEAT036	19.068	15.123	15.517	13.240	0.166	0.385	15.683	13.624	2.665	3.710	0.479	0.239	17.17%	28.02%	13.97%	24.53%	81.38%	87.55%	
	PEAT038	26.513	23.955	20.370	14.800	0.203	0.441	20.573	15.241	4.050	5.086	0.671	0.713	19.88%	34.37%	15.28%	21.23%	76.83%	61.78%	
	PEAT039	21.895	25.917	17.835	12.836	0.382	0.346	18.217	13.181	4.189	5.794	0.881	0.621	23.49%	45.14%	19.13%	22.36%	81.46%	49.53%	
	PEAT041	23.073	25.276	18.473	16.566	0.352	0.000	18.824	16.566	3.500	5.924	1.090	0.673	18.95%	35.76%	15.17%	23.44%	80.06%	65.54%	
Average ± SD		22.64 ± 3.08	22.57 ± 5.03	18.05 ± 2.00	14.36 ± 1.70	0.28 ± 0.11	0.29 ± 0.20	18.32 ± 2.02	14.65 ± 1.55	3.60 ± 0.69	5.13 ± 1.01	0.78 ± 0.26	0.56 ± 0.22	19.87 ± 2.66%	35.82 ± 7.06%	15.89 ± 2.24%	22.89 ± 1.42%	79.93 ± 2.16%	66.10 ± 15.85%	
C.o.V.		13.62%	22.29%	11.09%	11.82%	38.90%	68.01%	11.05%	10.59%	19.20%	19.78%	33.85%	38.88%	13.38%	19.72%	14.11%	6.19%	2.71%	23.98%	
All 25% Peat Samples: (n=32)																				
Average ± SD		32.74 ± 12.07	34.11 ± 12.45	23.29 ± 6.97	19.50 ± 8.09	0.74 ± 0.49	0.66 ± 0.31	24.03 ± 7.17	20.16 ± 8.27	8.88 ± 3.90	8.98 ± 2.85	4.41 ± 5.86	2.61 ± 3.35	39.09 ± 14.23%	48.61 ± 14.37%	28.09 ± 8.83%	27.11 ± 5.17%	73.78 ± 11.87%	58.63 ± 13.58%	
C.o.V.		36.87%	36.50%	29.92%	41.50%	65.73%	47.50%	29.85%	41.01%	43.84%	31.72%	132.87%	128.31%	36.41%	29.55%	31.43%	19.06%	16.09%	23.16%	
Putnam County Lakebed, Florida (60% moisture content) (n=3)	PEAT042	39.744	37.143	31.436	20.836	1.043	0.434	32.479	21.270	8.024	7.610	1.629	0.823	25.52%	36.52%	20.19%	20.49%	79.10%	56.10%	
	PEAT043	36.704	34.970	29.506	24.842	0.915	1.827	30.421	26.669	8.126	7.878	1.456	0.924	27.54%	31.71%	22.14%	22.53%	80.39%	71.04%	
	PEAT044	31.344	31.960	27.063	20.761	1.028	0.276	28.090	21.038	14.577	9.025	1.038	0.798	53.86%	43.47%	46.51%	28.24%	86.34%	64.96%	
Average ± SD		35.9																		

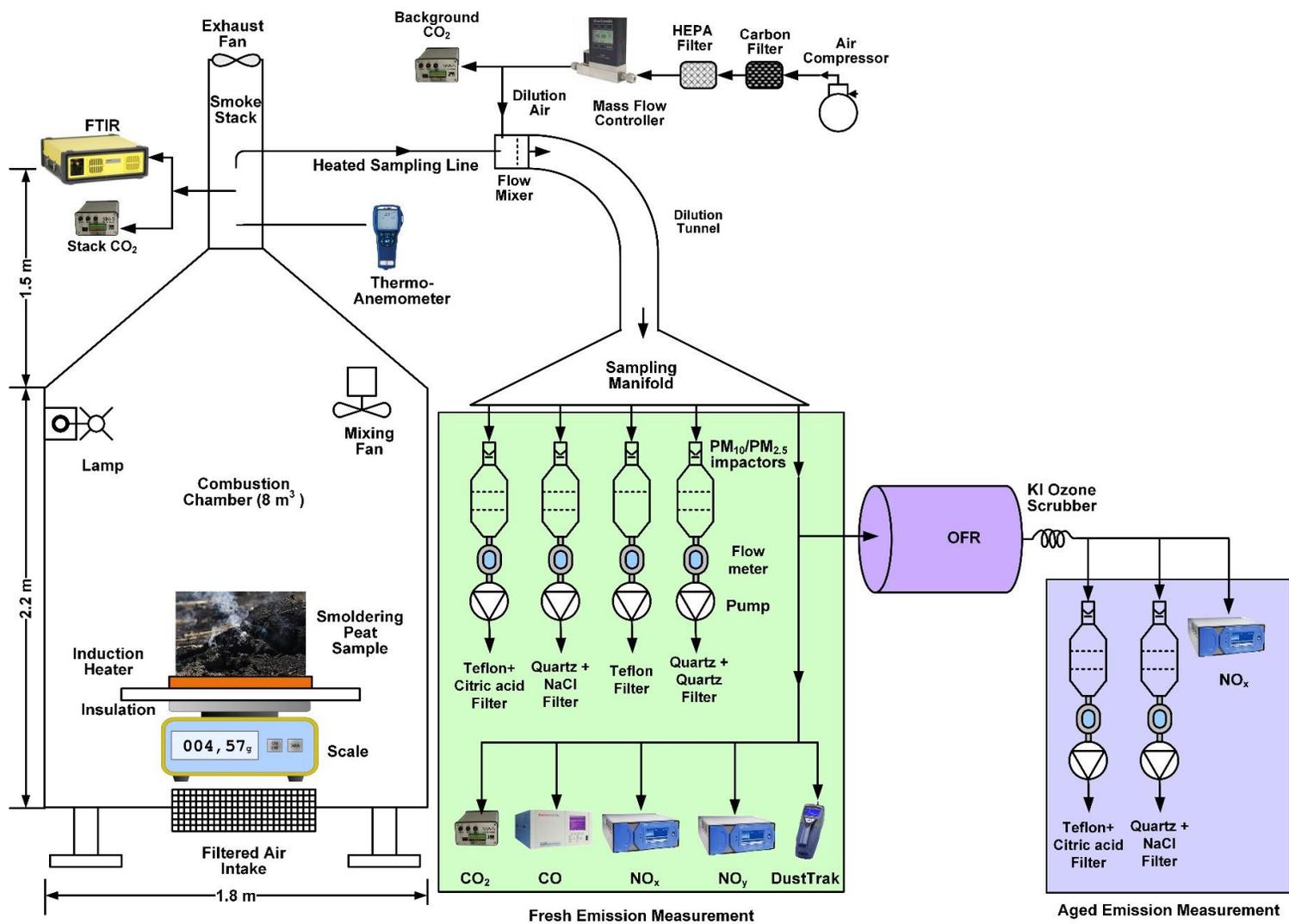


Figure 2. Configuration for peat combustion experimental set up. (FTIR: Fourier Transform Infrared Spectrometer; OFR: oxidation flow reactor; OFR lamps were operated at 2 and 3.5 volts to simulate aging of ~2 and 6.79 days, respectively)

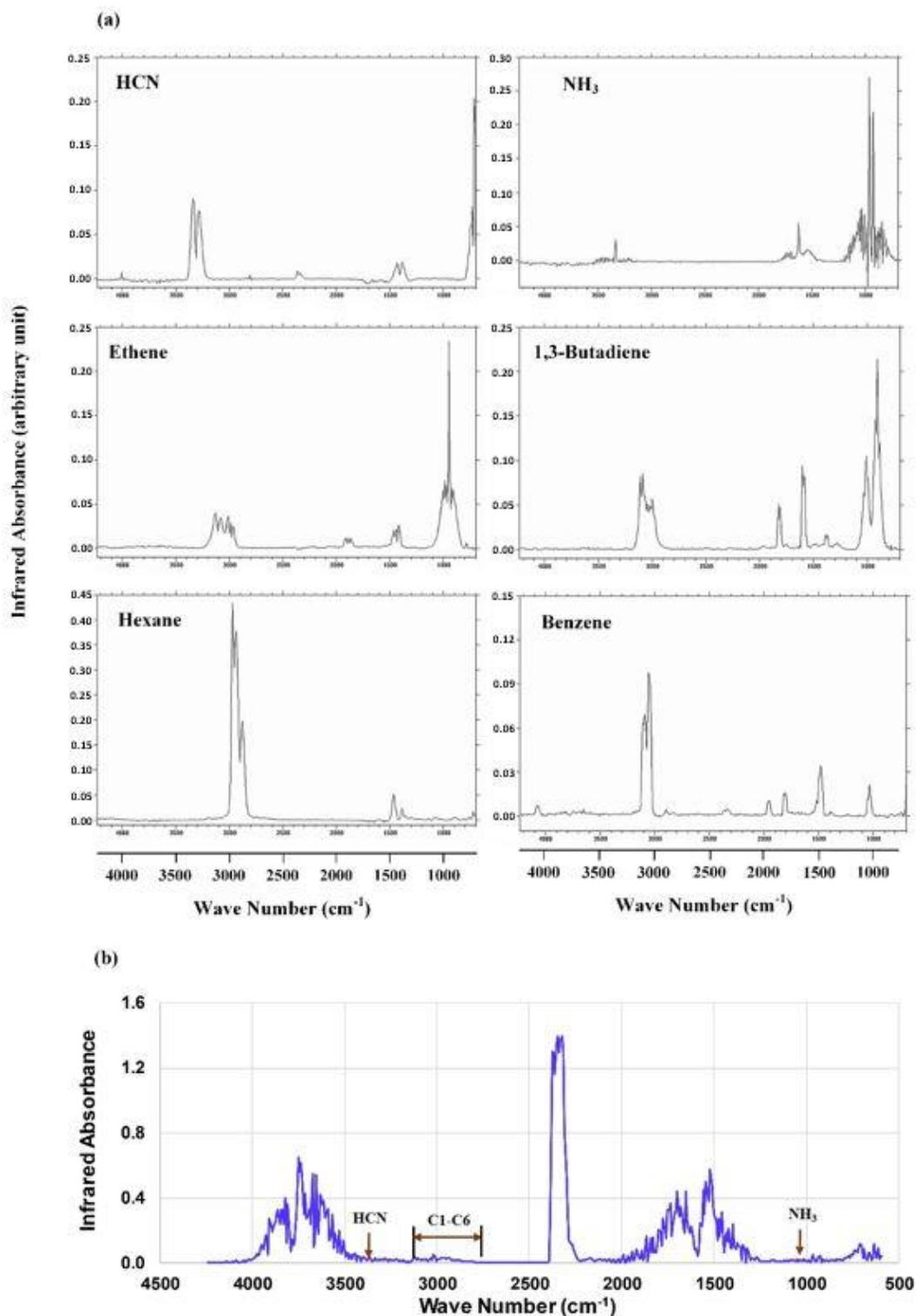


Figure S3. Examples of: a) FTIR spectra from reference gas absorption of HCN, NH₃, Ethene, 1,3-Butadiene, Hexane, and Benzene; and (b) FTIR spectrum of exhaust gas from an Everglades, Florida peat sample. The arrows show the characteristic absorption wavelength ranges of indicated gases.

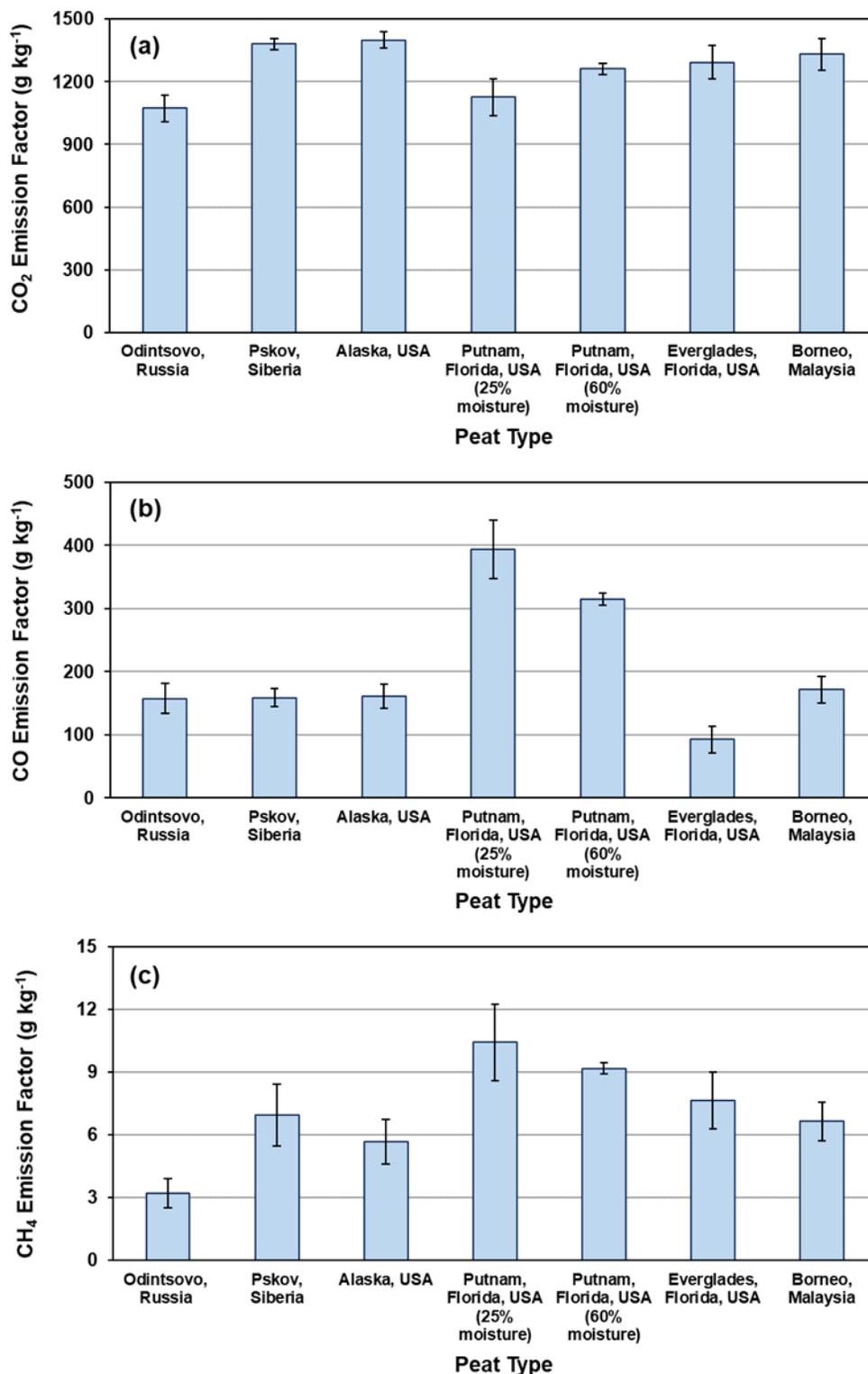


Figure S4. Comparison of emission factors of: (a) CO₂; (b) CO; and (c) CH₄ among different peats. All peats were tested with 25% fuel moisture content except that the Putnam County Lakebed peat was also tested with 60% fuel moisture content. Error bars indicate one standard deviation of the mean emission factor.

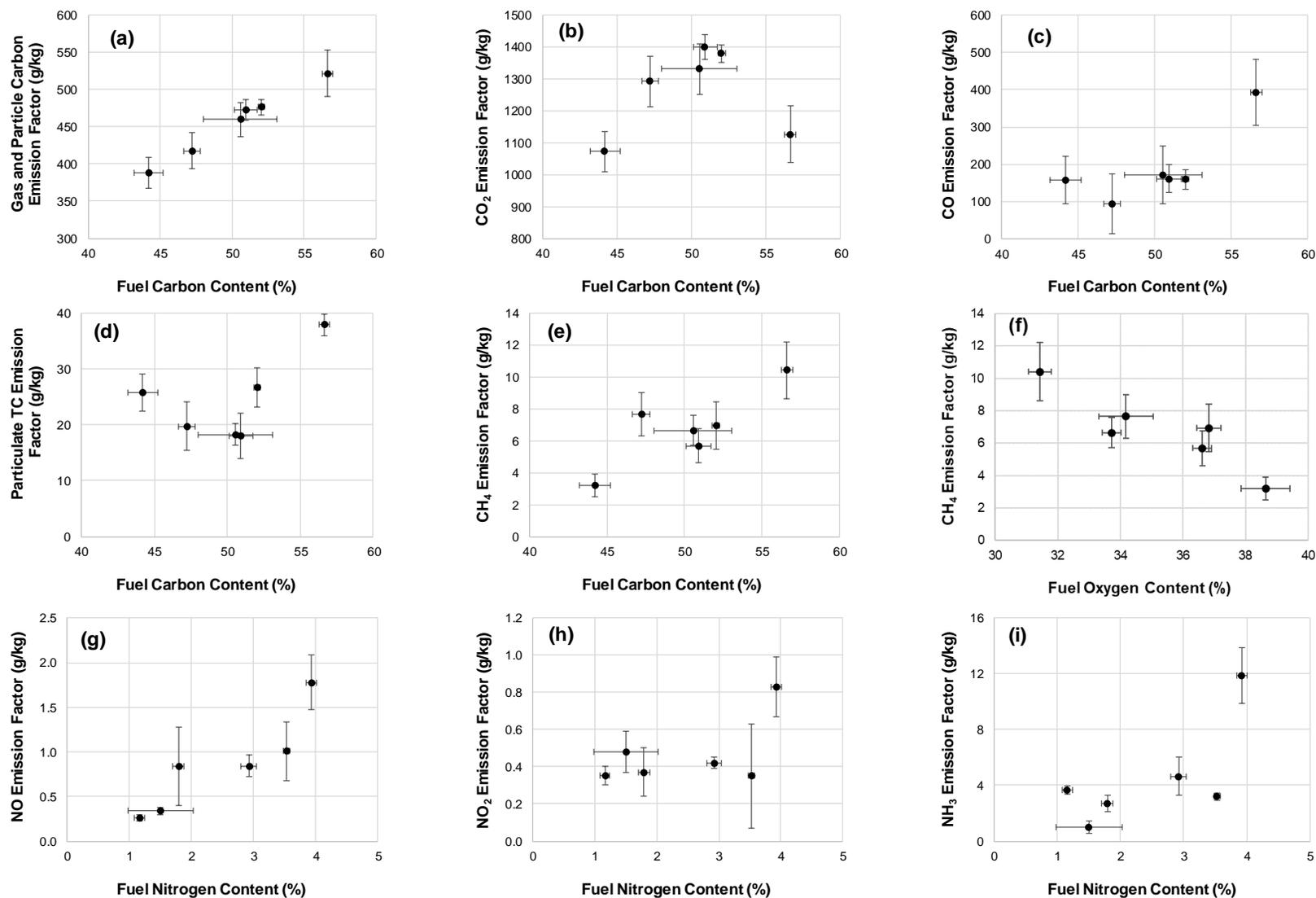


Figure S5. Emission factors of six types of peats for: (a) measured gas and particulate carbon; (b) CO₂; (c) CO; (d) particulate total carbon (OC+EC); (e) CH₄ as a function of fuel carbon content; (f) CH₄ as a function of fuel oxygen content; (g) NO; (h) NO₂; and (i) NH₃ as a function of fuel nitrogen content.

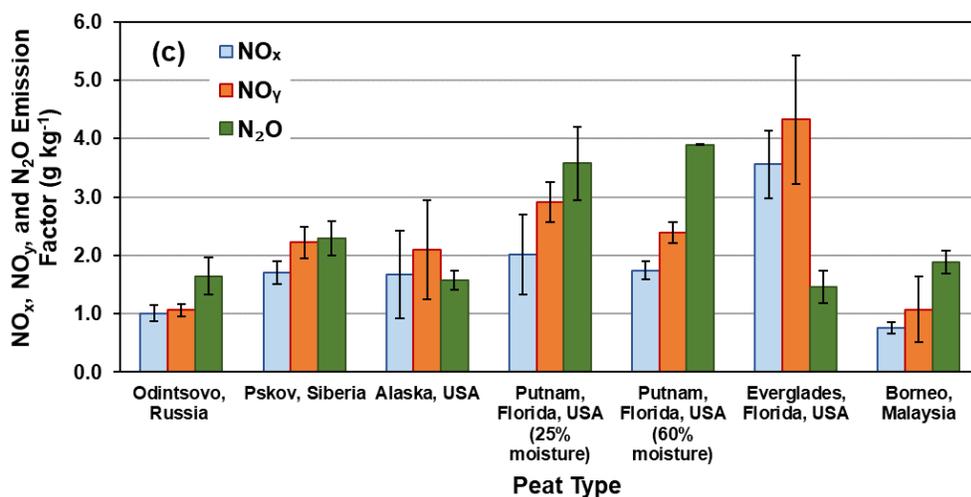
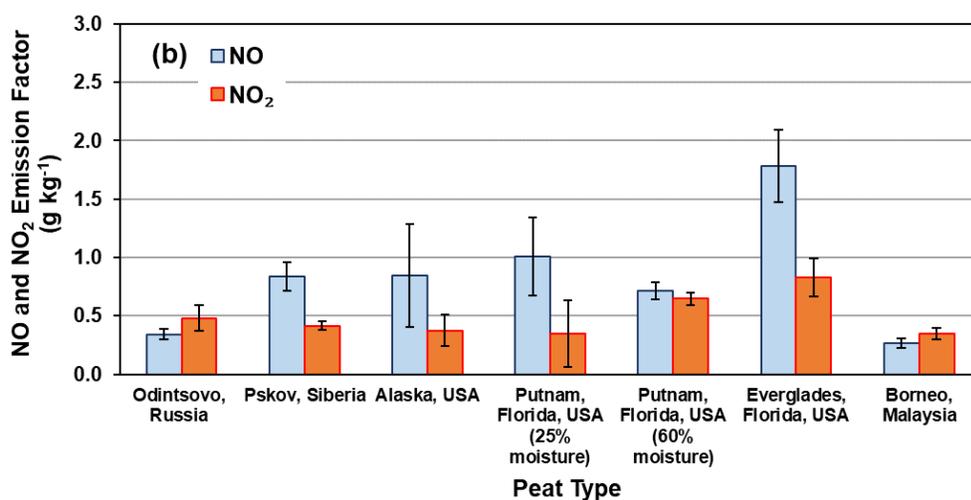
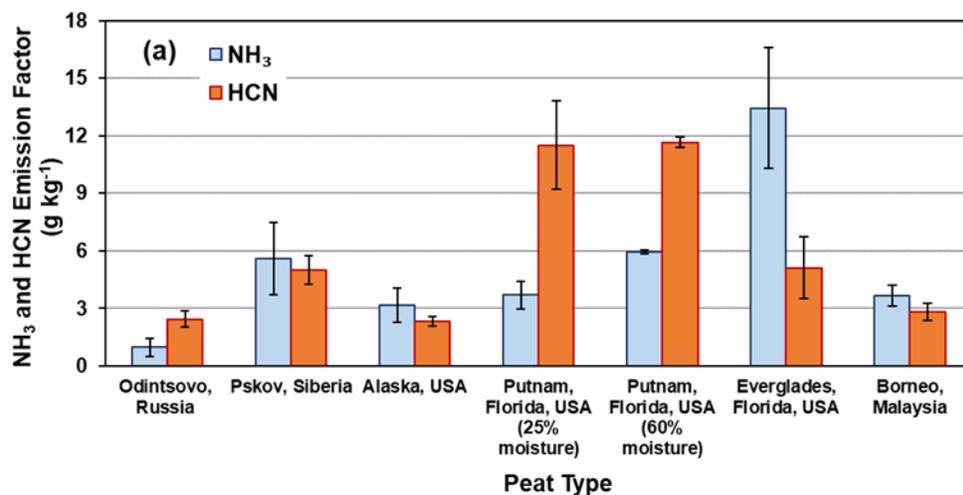


Figure S6. Comparison of fresh emission factors of: (a) NH₃ and HCN; (b) NO and NO₂; and (c) NO_x (as NO₂), NO_y (as NO₂), and N₂O among different peats. All peats were tested with 25% fuel moisture content except that the Putnam County Lakebed peat was also tested with 60% fuel moisture content. Error bars indicate one standard deviation of the mean emission factor.

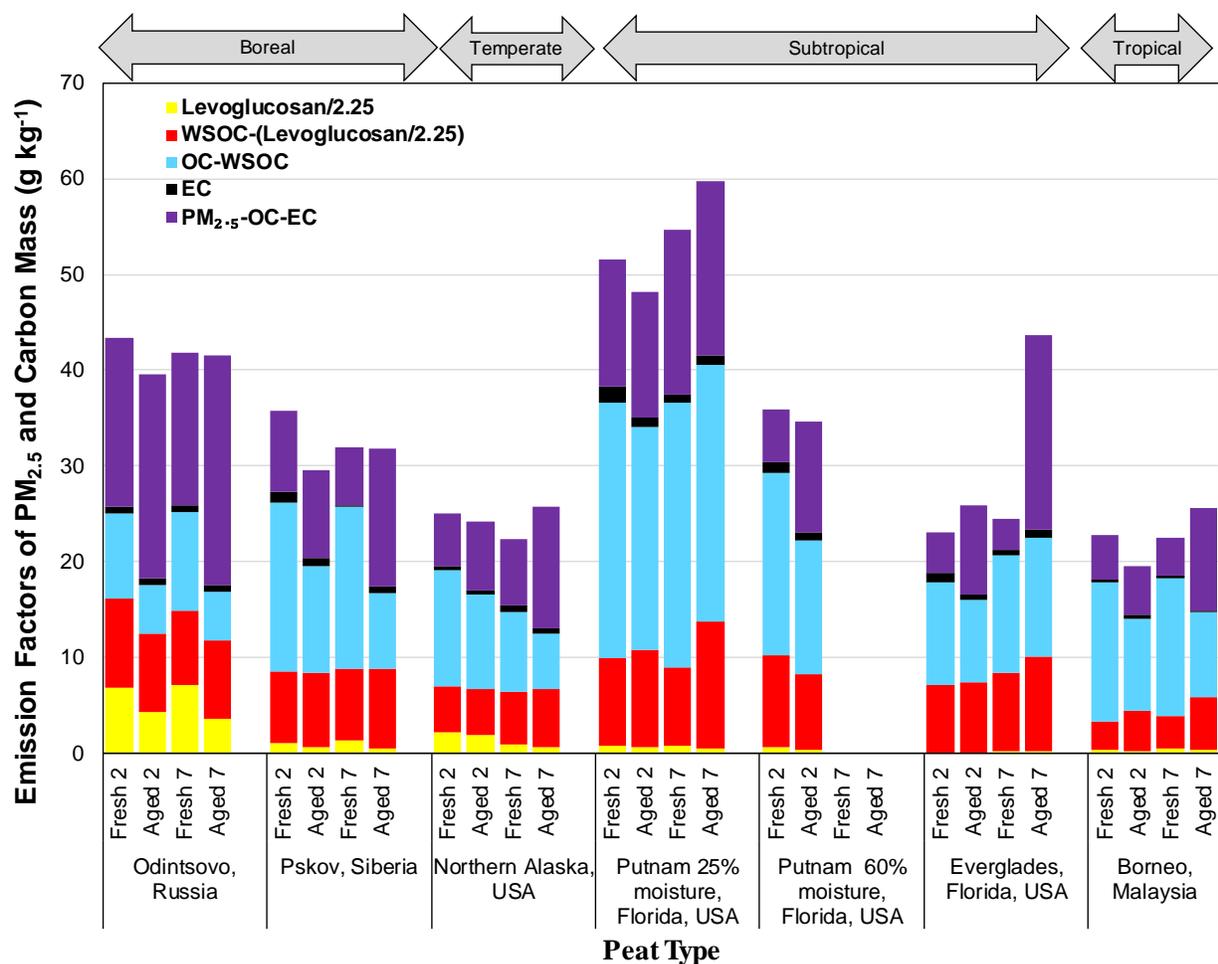


Figure S10. Comparison of fresh and aged emission factors of PM_{2.5} mass and carbonaceous compounds. All peats were tested with 25% fuel moisture content except that the Putnam County Lakebed peat was also tested with 60% fuel moisture content.

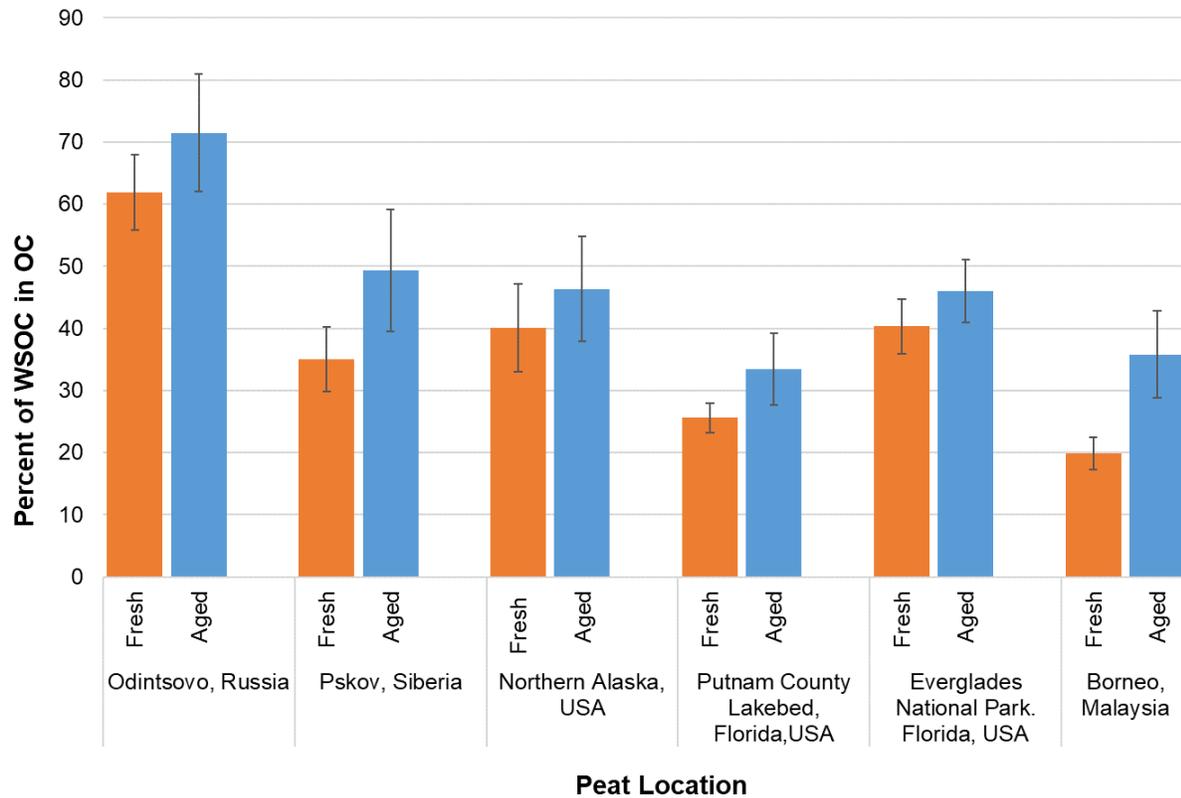


Figure S12. Percentage of water-soluble organic carbon (WSOC) in organic carbon (OC) for six types of peat samples (Fresh denotes all Fresh 2 and Fresh 7 samples; Aged denoted all Aged 2 and Aged 7 samples).

Responses to Reviewer #2 Comments

This manuscript presents measurement results from a laboratory combustion study of peat from different regions around the world. Detailed chemical speciation of both gas-phase and particulate smoke components is provided as a function of fresh vs simulated aged emissions, with a special focus on nitrogen species. The results presented in this paper are helpful to better understand the properties of biomass combustion source emissions and improve emissions inventories. Specifically, the authors presented a very nice detailed, yet concise description of the experimental set-up and procedures. The manuscript is well suited for publication in ACP, and only a few comments and suggestions are given below for the authors to consider in their revised version of the paper.

Specific comments:

1. *Line 148–151: What was the rationale for heating the fuels to such a relatively high temperature and reducing the moisture content to such low levels? As the authors point out, some volatile fuel components may have gotten lost during this preparatory step. Has the chosen heating temperature been applied in other similar studies as well?*

Response:

- Peat fuels were heated to 90°C to achieve a stable dry mass with ~0.16% moisture by weight content. The prescribed heating of peat samples to high temperature sterilization for non-US origin soil/peat fuels is required by the U.S. Department of Agriculture (USDA) under Animal and Plant Inspection Service for Plant Protection and Quarantine (USDA, 2010). We recognize that this is not ideal; however, most studies performed in the US with peat samples from non-US sources likely will have the same issue with potential changes in fuel properties by heating. The following sentences are revised to clarify this (Lines 110–113):

Import and export regulations (USDA, 2010) require high temperature heating of soil/peat fuels as part of the sterilization process. Peat fuels were heated to 90°C and weighed every 24 hours to achieve a stable dry mass with ~0.16 % moisture by weight content (after ~96 hours of heating).

2. *Lines 151–155: The authors may want to add a cautionary statement regarding the re-hydration procedure. According to studies conducted by the USFS Fire Science Lab, it is very difficult to re-hydrate biomass fuels once most of the water has been removed. And even after adding a certain amount of moisture back to the fuel, the physio-chemical properties are not the same as the original ones prior to the drying procedure. Would a potential alternative method be gradual drying to the desired moisture level, and thus maintaining the original water bonding structure?*

Response:

- We agree that this and several other aspects of laboratory-based biomass burning should be subject to caveats: these include collection and handling of samples (which causes issues with bulk density); nearly all aspects of sample storage; and sample combustion

conditions, as the Reviewer rightly indicates. Taken together these shortcomings all make the extrapolation of laboratory results to in-situ peat fires difficult. We suspect that the research community for peat and/or biomass combustion are aware of this and are exploring approaches to reduce uncertainties. To the Reviewer's point, it would be ideal to begin with fresh fuels collected at ambient (field) moisture, and then dried down to the desired moisture content. As pointed out above, import and export regulations by the USDA unfortunately prevented this strategy. Further research is needed to examine the influence of multiple drying-rewetting process on laboratory emissions of peat and other biomass fuels.

3. *Lines 378–381: Aside from lower OC emission factors, an increase in the WSOC fraction is expected due to the higher degree of oxygenation of the aged organic smoke components, isn't it?*

Response:

- Yes, the Reviewer is right that atmospheric aging will result in a higher degree of oxygenation of the aged organic smoke components. However, OC water-solubility varies by peat type. The companion paper by Chow et al. (2019) shows an increase of organic mass (OM) to organic carbon (OC) ratio from 1.1–1.7 to 1.3–2.2 for fresh and aged source profiles, respectively. An additional 14–21% increase in OM/OC ratios were found from 2- to 7-days of aging.

As WSOC is part of the OC, the WSOC/OC ratio can be used to illustrate atmospheric aging. As shown in Table S7, WSOC/OC ratios increased by 6–16 % after aging, this is also shown in new Figure S12. The following paragraph is added to clarify this (Lines 440–445):

As WSOC is part of the OC, the WSOC/OC ratio can be used to illustrate atmospheric aging. Figure S12 shows that WSOC/OC ratios increased by 6–16 % after aging. This is attributed to a combination of oxygenation of the aged organic emissions and the reduction of EF_{OC} (Table S7). The increase in WSOC/OC ratios may also be due to photochemical transformation of primary OC to WSOC and/or formation of water-soluble secondary organic aerosol during atmospheric aging (Agarwal et al., 2010; Aggarwal and Kawamura, 2009).

4. *Lines 410–411: Could the authors comment on possible degradation pathways that might occur during OFR treatment and potential reaction products of levoglucosan?*

Response:

- Degradation pathways for levoglucosan are complex as particle chemistry and mass transfer rate that dictate the reaction mechanisms are mostly unknown. Past studies show that the extent of levoglucosan degradation depends on OH exposure in the OFR, organic

aerosol composition, and vapor wall losses (e.g., Bertrand et al., 2018a; 2018b; Hennigan et al., 2010; Hoffmann et al., 2010; Lai et al., 2014; May et al., 2012; Pratap et al., 2019).

A modeling study by Hoffmann et al (2010) pointed to the atmospheric instability of levoglucosan. A smog chamber experiment conducted by Hennigan et al (2010) confirmed the degradation of levoglucosan as biomass burning emissions were exposed to OH radicals. However, large variabilities are found among the experiments, the extent of decay ranged from ~20–90 %. These tests suggested an atmospheric lifetime of ~0.7–2.2 days when biomass burning emissions are exposed to 1×10^6 molecule cm^{-3} of OH radicals.

The following paragraph is added to explain changes in $\text{EF}_{\text{Levoglucosan}}$ (Lines 472–479):

Past studies show that the extent of levoglucosan degradation depends on OH exposure in the OFR, organic aerosol composition, and vapor wall losses (e.g., Bertrand et al., 2018a; 2018b; Hennigan et al., 2010; Hoffmann et al., 2010; Lai et al., 2014; May et al., 2012; Pratap et al., 2019). Potential chemical pathways for the formation of organic species in biomass combustion emissions were proposed by Gao et al. (2003) that suggested the fragmentation of levoglucosan to C3–C5 diacids, followed by oxalic acid, acetic acid, and formic acid. This is consistent with the increases in $\text{EF}_{\text{organic acids}}$ after atmospheric aging, as shown in Table S6. However, detailed chemical mechanisms need to be further investigated.

5. *Lines 433–442: As the authors point out, higher fuel moisture content usually results in lower MCE, and consequently often increases PM emissions. However, the opposite pattern was observed in this study. Could this possibly be related, at least partly, to the re-hydration procedure which may not have restored the original conditions of the wet fuel (see comment No. 2 above)?*

Response:

- The observation of increased MCE from 0.65 ± 0.04 to 0.72 ± 0.01 and ~30 % reduction in $\text{EF}_{\text{PM}_{2.5}}$ as fuel moisture contents increased from 25 % to 60 % is indeed contrary to expectations and intuition. Unfortunately the re-hydration procedure does not seem to offer an explanation for this observation, since the implication would be that additional moisture would alter the chemical or physical properties of the fuels to permit more efficient combustion. While also not appealing to intuition, one explanation that appears possible is that at higher moisture values, combustion residence time is slowed enough so that radiant heat transfer from ignited particles to uncombusted areas of peat can be greater, thus increasing overall efficiency. However, as with putative chemical or physical transformations resulting from higher moisture, there are no data to support possible explanations. Besides the uncertainties associated with the small number ($n=3$) of samples submitted for the tests, it may be that our range of moisture values (25–60 %) was not high enough to reveal the expected relationship between moisture and MCE. Therefore, this explanation makes the most intuitive sense; even a moisture content of 60 % is quite low compared to many areas where peat fires can be observed.

The following sentence is added to the revised text (Lines 511–513):

It is hypothesized that at higher fuel moisture contents, combustion residence time is slowed enough so that radiant heat transfer from ignited particles to uncombusted areas of peat can be greater, thus increasing the combustion efficiency.

6. *Lines 487-489: Can the authors add some speculations regarding the "missing" nitrogen, i.e., whether it's due to unidentified nitrogen species, measurement uncertainties, or other reasons?*

Response:

- The unaccounted nitrogen may result from a combination of unidentified nitrogen species and measurement uncertainties. Examples of potential N-containing compounds in biomass burning emissions include nitrogen gas (N₂), isocyanic acid (HNCO) (Roberts et al., 2011); acetonitrile (CH₃CN), acetamide (CH₃CONH₂), benzonitrile (C₆H₅CN), and pyridine + pentadienenitriles (C₅H₅N) (Koss et al., 2018); as well as alkaloids, dissolved organic nitrogen, and nitroaromatic compounds (Benitez et al., 2009; Kopacek and Posch, 2011; Koppmann et al., 2005; Kuhlbusch et al., 1991; Laskin et al., 2009; Stockwell et al., 2015) are exemplified in Section 3.4. However, these specific nitrogen-containing species are not commonly measured in biomass/peat combustion emission sources.

Technical corrections:

1. *Please use consistent spacing between temperature numbers and the degree symbol throughout the manuscript.*

Response:

- Corrected
2. *Line 67-69: Additional field studies by Behera et al. (2014) and Engling et al. (2014) specifically address the effects of peat burning emissions in Southeast Asia.*

Response:

- Yes, many past studies characterize the emissions from peat fires in Southeast Asia. The following sentence is added to acknowledge this (Lines 69–73):

For Southeast Asia, fire-related regional air pollution and its effects on atmospheric visibility, forest ecosystem, and human health have been addressed in many studies (e.g., Behera et al., 2014; Betha et al., 2013; Bin Abas et al., 2004; Dall'Osto et al., 2014; Engling et al., 2014; Fujii et al., 2017; Heil and Goldammer, 2001; Hu et al., 2019; Kundu et al., 2010; Levine, 1999; Tham et al., 2019).

3. *Line 178: Omit the indefinite article at the beginning of the sentence.*

Response:

- This section (Lines 199–201) has been revised as:

A portion (0.5 cm²) of the quartz-fiber filter was submitted for organic, elemental, and brown carbon (OC, EC, and BrC) analyses following the IMPROVE_A thermal/optical reflectance (TOR) protocol (Chow et al., 2007; 2015).

4. *Line 215: The degree symbol is not needed for temperatures expressed in K units.*

Response:

- Corrected

5. *Line 258: When the authors state the "concentrations were high", it would be helpful for the reader to know a reference point, i.e., "compared to".*

Response:

- The sentence is revised as the following (Lines 299–300):

PM_{2.5} mass concentrations, in the range of 328–2277 µg/m³, are one to two orders of magnitude higher than those commonly measured at ambient monitoring sites.

6. *Lines 313-314: Isn't the low correlation coefficient indicating different emissions characteristics of the fresh vs. aged smoke, and not just a variability between tests?*

Response:

- The Reviewer is correct. The sentence has been revised as the following (Lines 363–365):

A low correlation coefficient ($r = 0.67$) between the downstream and upstream EF_{NO_x} suggests the changes of NO/NO₂ ratios between the fresh and aged emissions as well as variabilities among tests.

7. *Line 453: Add "volatile" before "carbon".*

Response:

- Corrected

8. *Lines 495-496: The statement regarding the nitrogen content in this sentence is not clear.*

Response:

- The sentence is revised as the following (Lines 568–570):

Dried peat fuel contains 44–57 % carbon (C), 31–39 % oxygen (O), 5–6 % hydrogen (H), 1–4 % nitrogen (N), and <0.01 % Sulfur (S). The nitrogen to carbon ratios are low, in the range of 0.02–0.08, consistent with peat compositions reported in other studies.

9. *Lines 510-511: What do the authors mean with “average Russian peat”?*

Response:

- As average EFs are presented in the study. The sentence is revised as follows (Lines 480–481):

The highest $EF_{\text{LevoglucoSan}}$ is found for the fresh Russian peats ($15.8 \pm 2.9 \text{ g kg}^{-1}$), and this is diminished by 45 % after 7-day aging ($8.8 \pm 2.1 \text{ g kg}^{-1}$).

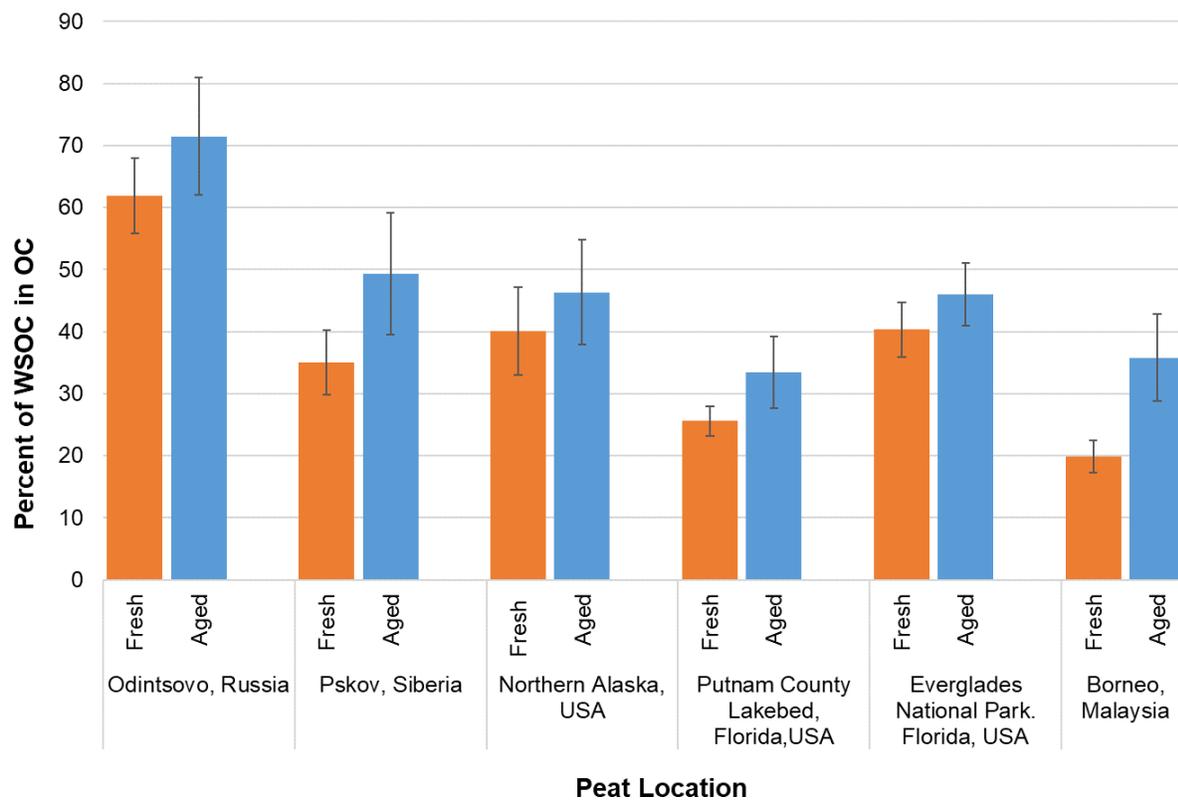


Figure S12. Percentage of water-soluble organic carbon (WSOC) in organic carbon (OC) for six types of peat samples (Fresh denotes all Fresh 2 and Fresh 7 samples; Aged denoted all Aged 2 and Aged 7 samples).

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