

Brown carbon emissions from laboratory combustion of Eurasian arctic-boreal and South African savanna biomass

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Abstract:

Warming climate is predicted to increase forest fires which can be a major source of black and brown carbon (BC and BrC) into the atmosphere. Unlike North American forest fires, very limited studies have characterized North Eurasian biomass burning (BB) emissions. In this work, we determined the emission factors (EF) of carbonaceous aerosols and characterized light absorption of BrC emitted from boreal and peat burning through offline filter extraction method. The results were compared to African savanna emissions. Effects of atmospheric dilution and oxidative aging on BrC absorptivity were investigated for selected BB emissions sampled into an environmental chamber. Organic carbon (OC) and elemental carbon (EC) EFs of fresh BB emissions ranged between 1.30-89.9 g kg⁻¹ and 0.01-4.80 g kg⁻¹ respectively. Methanol soluble OC (MSOC) represented more than 92% of fresh BB emissions, irrespective of fuel type, and consisted of weakly absorbing BrC with imaginary refractive index at 550 nm ($k_{\text{MSOC}_{550}}$) ranging from 0.002 to 0.011. Water soluble OC (WSOC) fractions varied among fresh BB emissions but overall exhibited higher mass absorption efficiencies at 365 nm (MAE_{365}) than MSOC. Dilution-related evaporative loss in environmental

38 chamber resulted in less volatile OC, making them less soluble in methanol. Photochemical and dark
39 oxidative aging further increased the low volatility OC fractions of the organics along with its oxidation
40 state. Our estimated OC-EC emission factors and k_{MSOC} for fresh BB emissions can be used for future
41 modelling purposes. Further online measurements are needed to account for non-soluble strong BrC in aged
42 BB emissions.

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45 **Introduction:**

46 Biomass burning (BB) emission is one of the largest anthropogenic sources of black (BC) and brown carbon
47 (BrC) in the atmosphere (Bond et al., 2004; Kirchstetter et al., 2004). Multiple studies have predicted that
48 open BB emissions such as wildfires will become more prevalent in the future with the warming climate,
49 with increasing boreal forest and peatland fires threatening also the Eurasian regions (Krawchuk et al., 2009;
50 Costa et al., 2020; Feyen et al., 2020). Along with the expected overall increase in the frequency of “high-to-
51 extreme” wildfires in Europe (de Rigo et al., 2017) an expected shift in vegetation distribution from Southern
52 towards Northern Europe may further amplify fire hazard of Fenno-Scandinavian biomass (Costa et al.,
53 2020). Only limited laboratory and field studies exist describing aerosol emissions from boreal forest and
54 peatland fires in the Eurasian area (Wilson et al., 2015; McCarty et al., 2021; Zhong et al., 2024 ; Schneider et
55 al., 2024a), compared to the more studied North-American boreal forest and peat fires (Aurell and Gullett,
56 2013; Urbanski, 2013; Stockwell et al., 2014; Black et al., 2016; Andreae, 2019; Phillips et al., 2022; Zhao et
57 al., 2021). However, the fire regimes in Eurasia are known to be dominated by surface burning, that likely
58 emits carbonaceous aerosol of different characteristics in comparison to more crown-fire dominated fires in
59 North America (De Groot et al., 2013; Rogers et al., 2015). Another important source of global carbon
60 emissions is savanna surface fires, which has been estimated to account for almost 50% of total carbon
61 emissions from open BB (van der Werf et al., 2017). Further, roughly 30% of the total wildfire-induced
62 carbon emissions originate solely from Southern African savanna fires (van Wees et al., 2022). Yet, relatively
63 limited studies have been carried out to characterize these emissions in recent years (Vakkari et al., 2014,
64 2018; Desservettaz et al., 2017; Wu et al., 2021; Vernooij et al., 2022, 2023).

65 Chemical and optical properties of BC have been studied extensively over the last two decades and it has
66 been established as an important climate warming agent (Jacobson, 2000, 2001; Bond et al., 2013). On the
67 other hand, the properties of combustion emitted BrC have still not been fully characterized due to variability
68 in combustion conditions (Martinsson et al., 2015; Wang et al., 2020; Saleh, 2020), fuel chemical
69 composition (Saleh et al., 2014; Smith et al., 2020; Moschos et al., 2024; Navinya et al., 2024) and
70 secondary transformation of BrC in the atmosphere (Laskin et al., 2015; Brown et al., 2018; Hems et al.,
71 2021). BB derived BrC consists of different light-absorbing organic precursors of BC, such as polyaromatic
72 molecules, which are not transformed to fully ordered BC during the combustion process (Saleh et al., 2018).
73 Since BC formation is strongly temperature-dependent, low combustion temperatures may favour BrC
74 formation, whereas high combustion temperatures increase oligomerization/polymerization of polyaromatic
75 carbon structures to form BC (Faccinnetto et al., 2011; Desgroux et al., 2013; Solum et al., 2001; Wang,
76 2011). Consequently, the light-absorbing primary organic aerosols (POA) emitted from BB emission are
77 chemically diverse and distinct for different fuel types and combustion conditions. Additionally, they evolve
78 in the atmosphere through oxidation and functionalization reactions, driven by the formation of secondary
79 organic aerosol (SOA) and the evaporation and chemical fragmentation of organic aerosol (OA), resulting in
80 a complex mixture of organic chromophores with variable absorptivity.

81 BB is a dynamic and variable process that strongly depends on fuel moisture content, fuel
82 composition and combustion conditions. A common parameter to characterize BB is the modified
83 combustion efficiency (MCE), which describes the share of carbon dioxide to the sum of CO and CO₂
84 emissions. However, studies have typically found only weak correlations between BrC light absorption and
85 MCE (Pokhrel et al., 2016; McMeeking et al., 2014). The ratio of the emitted BC (mainly composed of
86 elemental carbon or EC structures) to the organic carbon (OC) has been suggested as a more suitable
87 parameter for correlating the aerosol optical properties with combustion conditions (McClure et al., 2020;
88 Stockwell et al., 2016; Saleh, 2020). For instance, flaming dominated combustion processes generally lead to
89 relatively high EC/OC ratios and more absorptive BrC than smouldering emissions (Saleh et al., 2014; Xie et
90 al., 2018; McClure et al., 2020; Kumar et al., 2018).

91 In climate models, the light absorption strength of the material is described by the imaginary
92 part of the refractive index (k). For pure BC, k - are constrained close to unity and exhibit very little to no
93 wavelength dependence, especially at UV and shorter visible wavelengths (Bond and Bergstrom, 2006).
94 However, the k for BB emitted BrC seem to vary across several orders of magnitude depending on the
95 biomass type and combustion conditions as well as the measurement techniques (Chakrabarty et al., 2010;
96 Bluvstein et al., 2017; Saleh et al., 2018; Saleh et al., 2020; Navinya et al., 2024). Therefore, characterizing
97 the k for individual BrC compounds is a daunting and, in some cases, impossible task due to instrumental
98 limitations. A more comprehensive approach has been adapted in the last decade where the k values for BB
99 derived BrC has been shown to fall in a “brown-black carbon continuum” (Saleh et al. 2018). BB derived
100 BrC generally exhibit progressively higher k values with increasing temperature and has been termed as
101 “dark BrC (d-BrC)” (Hoffer et al., 2017; Adler et al., 2019; Atwi et al., 2022; Chakrabarty et al., 2023) or
102 “strongly absorptive BrC (s-BrC)” (McLure et al., 2020; Saleh et al., 2020) when the combustion
103 temperatures approach the BC formation regime. In contrast, the BrC derived from BB combustions in
104 progressively decreasing combustion efficiency and temperature has been termed as “moderately absorptive
105 (M-BrC)”, “weakly absorptive (w-BrC) and “very weakly absorptive (VW-BrC)”, respectively (Saleh et al.,
106 2020; Moschos et al., 2024), as the k values decrease with lower burning temperature.

107 One of the most convenient and widely used methods for the characterization of optical
108 properties of bulk BrC is the collection of aerosol particles on filters and subsequent extraction of the organic
109 fractions with suitable solvents followed by filtration (Chen and Bond, 2010; Liu et al., 2013; Mo et al.,
110 2017; Shetty et al., 2019; Li et al., 2020, Yan et al., 2020). The light absorption of solvent extracted OA is
111 measured using a UV-vis spectrophotometer which provides high precision spectral data over a wide
112 wavelength range. In previous studies water has been one of the primary solvents for extraction of BrC from
113 filters (Bosch et al., 2014; Kirillova et al., 2014; Mukherjee et al., 2020) because of the atmospheric
114 relevance of the water soluble organic (WSOC) fraction (Hallar et al., 2013, Taylor et al., 2017).
115 Quantification of WSOC can be done with Total Organic Carbon (TOC) analysis (Li et al., 2016), and the
116 loss estimation of the extraction process has very little uncertainty. On the other hand, the water insoluble
117 BrC fraction is generally extracted from aerosol particles using methanol (MeOH) as a solvent (Chen and

Bond, 2010) but it has been recently discovered that some highly light-absorbing, extremely low volatility organic (ELVOC) compounds may not be efficiently extracted by this procedure (Saleh et al., 2014; Liu et al., 2013). Quantifying the dissolved organic mass in MeOH is also challenging as organic solvents interfere with TOC measurements and indirect methods are used instead (Chen and Bond 2010; Cheng et al., 2017; Huang et al., 2018; Yan et al., 2020) to estimate MeOH soluble organic carbon (MSOC) which leads to additional potential sources of uncertainties (Yan et al., 2020). Nevertheless, MeOH has exhibited very high organic extraction efficiencies (Chen and Bond, 2010; Xie et al., 2017) and the optical properties of MSOC have agreed well with OC extracted by more polar solvents like Dimethylformamide (DMF) for BB emissions and coal combustion (Xu et al., 2022). Therefore, MeOH should be used in parallel to water to extract BB emitted organic compounds with a broader range of polarities and gain more information on their light-absorbing properties. Although filter based solvent extraction has its limitations, such as lack of information on size-dependent absorption of extracted organics (Moosmüller et al., 2011; Liu et al., 2013; Washenfelder et al., 2015), this analytical method is low-cost, easily accessible, and excludes the interference of BC (or EC) and other light-absorbing species from OA absorption.

This work aims to define the emission factors of carbonaceous aerosols and characterize OA optical properties for emissions originating from different open BB sources. We used a laboratory open burning setup with the objective to create conditions representing natural Eurasian forest surface fires, in which the combustion temperatures likely remain low and burning is dominated by smouldering (Rogers et al., 2015; Walker et al., 2020). Finnish boreal peat and forest surface samples, commercially available boreal peat samples, and permafrost peat from arctic Russia and Svalbard (Norway) were burned in the laboratory setup. To assess the effects of burning conditions and biomass materials on carbonaceous emission factors and their optical properties, we replicated conditions ranging from smouldering to flaming, with clearly distinct combustion behaviour and MCEs for each fuel. Furthermore, we extended the study samples to include South-African savanna biomass and North European wood stove emissions, thereby encompassing a larger range of combustion conditions. Finally, we investigated the impact of atmospheric dilution and aging on the chemical and optical properties of the organic aerosols by conducting environmental chamber experiments either under photochemical or dark aging conditions for selected biomasses. The results were

145 used to derive imaginary refractive indices (k) for OAs, allowing for their classification within the black-
146 brown carbon continuum. The results are essential to accurately estimate the direct radiative forcing effects
147 of biomass burning emissions in climate models.

148

149 **2. Methodology:**

150

151 **2.1 Combustion setup and fuels**

152 The combustion experiments were conducted in the ILMARI laboratory of the Kuopio campus of University
153 of Eastern Finland (<https://sites.uef.fi/ilmari>) using an in-house designed open combustion appliance. The
154 appliance consisted of a steel cage, a concave plate, a metal/steel mesh and a metal/steel biomass holder. The
155 details of the combustion setup and the specific biomass holders used for each fuel type are illustrated in
156 Supplementary Fig. S2. Combustion was initiated using an electric resistor of which the power was adjusted
157 to generate exclusively flaming or smouldering emissions, similar to previous studies (Pokhrel et al., 2021;
158 McRee et al., 2025). With this open combustion setup (Fig. S2), we allowed instant dilution of the emissions
159 in an aim to simulate real world forest surface fires.

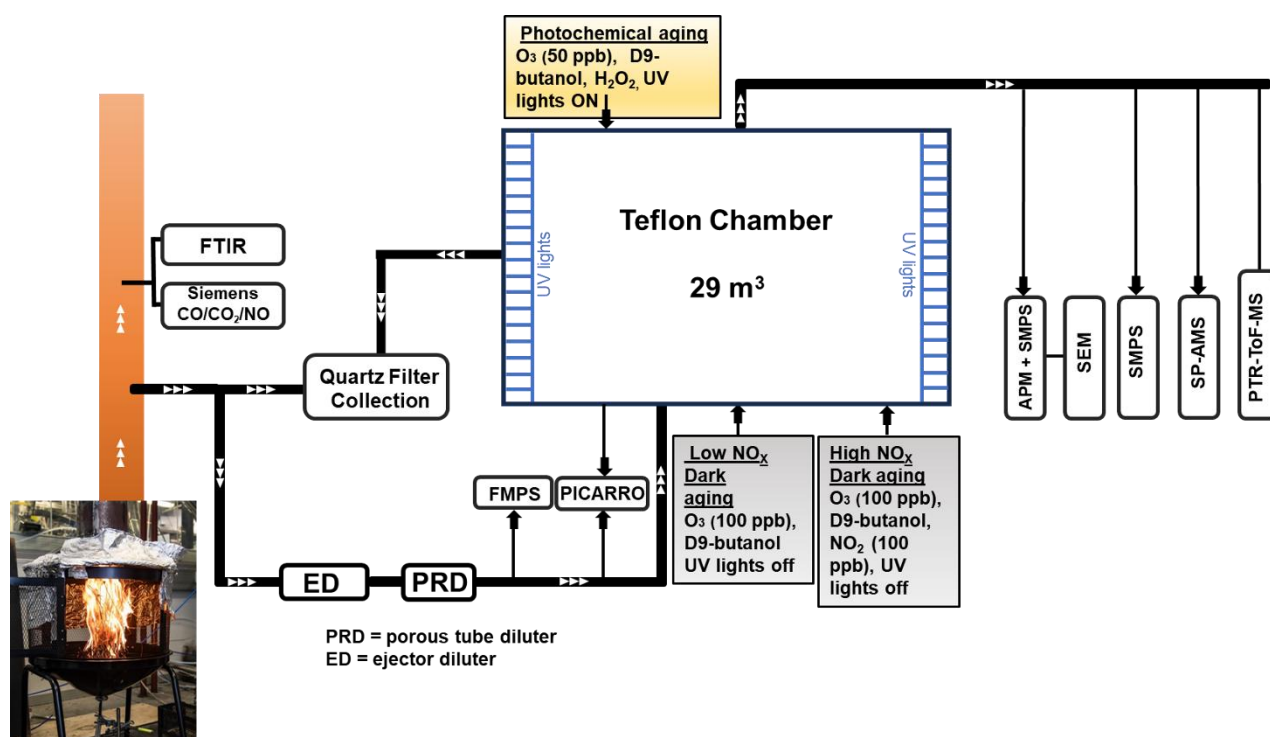
160 Eight different types of biomasses were used as fuel samples in the combustion experiments. Namely,
161 commercially available peat samples (CP), Finnish boreal forest surface (BFS, including vegetation, litter,
162 and the soil organic layer), peat from two Finnish boreal peatlands (FIA and FIB), peat from arctic
163 permafrost regions of Russia (RUS) and Norway (NOR), savanna wood and grass from South Africa (SW
164 and SG respectively) were selected as fuels for this study. The origins and properties of the combusted
165 biomasses are available in the Supplementary Information (Section S1, Table S1). The savanna biomass
166 included in this study was part of the BASFAA (Boreal and savanna fire aerosol aging) measurement
167 campaign that took place from May to June 2022 at the ILMARI laboratory as described in Vakkari et al.
168 (2025). For this study specific combustion phases were selected for sampling representing either solely
169 flaming, pre-flame smouldering, or full combustion consisting of pre-flame smouldering, flaming and post-
170 flame smouldering periods. Therefore, a subset of Savanna biomass (SG and SW) combustions presented in
171 Vakkari et al. (2025), which included sampling of separate combustion phases (either flaming or pre or post

172 flame smouldering), were chosen to be included in this study. The sampling periods were selected based on
 173 careful visual inspection of the combustion process and the concentrations of CO and CO₂, monitored online
 174 as described below (Section 2.2).

175 2.2. Sampling setup and gas phase measurements

176 The emitted smoke was sampled using the setup presented in Fig. 1. We measured the gaseous

177



178

179 **Figure 1: A schematic presentation of the experimental setup for combustion experiments involving the**
 180 **environmental chamber**

181

182 compounds from the fresh, undiluted BB emission using an online multicomponent FTIR analyzer (Gasmeter
 183 Technologies Inc.), and the measured compounds are listed in Table S2. The fresh BB emissions were
 184 sampled through a PM₁₀ pre-cyclone and a heated probe (180 °C) before being diluted using an Ejector
 185 Dilutor (Titta et al., 2016) and porous tube dilutor. A dilution ratio (DR) of 4-20 was achieved during this
 186 two-stage dilution with clean synthetic air (Woikoski N50) in ambient temperature. A gas concentration

187 analyzer (Picarro G2401) measured the concentrations of CO₂, CO, CH₄ and water vapor (H₂O) from the
188 diluted exhaust (Fig. 1).

189 MCEs were calculated from the average increase in CO₂ and CO concentrations during the sampled
190 combustion period relative to the background concentrations (Eq. 1)

$$191 \quad MCE = \frac{\Delta[CO_2]}{\Delta[CO_2] + \Delta[CO]} \quad (1)$$

192 MCE was estimated from CO and CO₂ concentrations measured in both undiluted and diluted BB emission.
193 Burns with average MCE values smaller than 0.9 were defined as smouldering-dominated combustion, while
194 the combustions with average MCE values larger than 0.9 (Table 1) were classified as flaming dominated
195 (Yokelson et al., 1996; Stockwell et al., 2014).

196

197 **2.3 Environmental chamber and aging of the emissions**

198 Different burning phases of Finnish BFS, and Finnish commercial peat (CP) samples were used in the
199 chamber experiments to study the effects of dilution and oxidative aging on the organic aerosol optical
200 properties. Additionally, samples of Savanna burning chamber experiments, described by Vakkari et al.
201 (2025), were analyzed as part of this study. For peatland samples FIA, FIB, NOR and RUS, only fresh
202 emissions were characterized in this study and no chamber feeding was performed. In each of the chamber
203 experiments, diluted primary BB emissions were sampled into a 29 m³ TeflonTM chamber (Leskinen et al.,
204 2015) which was pre-filled with purified air (Model 737-250, Aadco Instruments Inc.). For each experiment,
205 the sampling period to the environmental chamber was set based on the targeted mass concentration of 20-50
206 µg m⁻³ as estimated from the fresh particle size distribution measured online by a fast mobility particle sizer
207 (FMPS, model 3091, TSI Inc.). The relative humidity (RH) inside the chamber was set at 20% for SG and
208 SW emissions and at around 50% for BFS and CP emissions to reproduce typical daytime RH in
209 corresponding environments during the fire active seasons, using a humidification setup described in
210 Leskinen et al. (2015). The temperature in the chamber was kept constant at approximately 22°C. The
211 sample was first allowed to mix and homogenise for 20 minutes, which was an adequate duration for the
212 mass concentrations of particulate matter (PM) and organics inside the chamber to stabilize, as observed

213 respectively by a scanning mobility particle sizer (SMPS) and a soot particle aerosol mass spectrometer (SP-
214 AMS, Aerodyne Research Inc, USA) connected to the chamber. After sampling of the fresh exhaust, Picarro
215 was switched to monitor the gaseous components inside the chamber. Total DRs inside the chamber were
216 135-4045 compared to the two-stage diluted fresh emission (Table 1), based on the ratio of CO concentration
217 measured by Picarro in the diluted exhaust and from the chamber.

218 After the stabilization period, chamber diluted primary BB emissions were monitored for
219 additional 45 mins before adding reactants to the chamber to induce oxidative aging. Oxidative reactants
220 were injected into the chamber after the BB emissions, because we first wanted to measure the characteristics
221 of primary aerosols as well as, to study the effect of dilution on the partitioning of the POA fraction without
222 inducing any aging pathways, similar to previous studies (Kodros et al., 2020, 2022). The oxidative reactants
223 were allowed to interact with the fresh BB emission in the chamber for 4.5 hours. For each experiment, 2 μ L
224 of d9-butanol (~25 ppb) was injected into the chamber and the concentration was probed throughout the
225 experimental duration using a proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS).
226 Four distinct aging conditions were simulated in the environmental chamber to evaluate the impact of aging
227 on the physio-chemical and optical properties of different BB emissions. Firstly, photochemical aging
228 conditions were induced inside the chamber for both flaming and smouldering combustion. In the
229 photochemical experiments, UV-light in presence of externally fed O₃ (50 ppb) and H₂O₂ (0.5 ml of 30% v/v
230 solution) led to the formation of hydroxyl radicals (OH $\cdot$). The OH exposure and equivalent photochemical
231 age were determined from the decay of d9-butanol (Barmet et al., 2012). In our experiments, equivalent
232 photochemical age ranged between 1-1.6 days (Table 1) with an assumed ambient OH concentration of
233 $1.5 \times 10^6 \text{ cm}^{-3}$. In addition, two different dark aging conditions were simulated for two distinct sets of
234 biomasses. The primary emissions from SG and SW underwent dark aging in the presence of externally
235 added 100 ppb of O₃ and no additional NO_x. We classified this aging condition as “*low-NO_x dark aging*”, as
236 the nitrate radical (NO₃) formation was limited by the lack of NO_x. CP and BFS burning emissions were
237 aged in relatively ‘*high-NO_x*’ conditions in the dark chamber with 100 ppb NO₂ followed by 100 ppb O₃
238 directly injected to chamber. It should be clarified that even though we termed this aging condition as “*high-*
239 *NO_x*”, the NMVOC/NO_x ratio was still relatively high (Seinfeld and Pandis, 2006), in the range of 4-6 in

these cases (Table 1). Furthermore, the addition of O₃ within a small span of time (~10 minutes) after the addition of NO₂ in the chamber might have led to competition between the expected NO₃ radical chemistry and ozonolysis pathways, although the reaction rates of ozonolysis are much slower than nitrate oxidation pathways. Another method for simulating aerosol aging with NO₃ radicals would be to precondition the chamber with NO₂ and O₃ before adding the BB emissions (McRee et al., 2025). However, by doing this, we would not have been able to study both fresh and oxidatively aged aerosol during the same chamber experiment. Seven additional experiments were conducted in this study without any added oxidants in the chamber to evaluate the impacts of chamber dilution, estimate wall loss and other processes on the exhaust emissions. Out of these seven experiments, three were conducted by sampling emissions from the whole combustion of BFS, while the other four experiments were constituted with distinct smouldering and flaming dominated burns of CP and BFS.

2.4 Particle size distribution, density and morphology:

The particle size distributions were measured from the environmental chamber for the whole duration of each experiment by a scanning mobility particle sizer (SMPS) consisting of a differential mobility analyzer (DMA, model 3080; TSI Inc.) and a condensation particle counter (CPC, model 3775; TSI Inc.). In addition, an aerosol particle mass analyzer (APM, model 3602; Kanomax Inc.) in tandem with another SMPS (with *DMA 3081* and *CPC 3750*, TSI Inc.) was used to measure the size distribution of mass classified particles to estimate the size-resolved effective densities of the particles (Leskinen et al., 2023; Mukherjee et al., 2024). Particle densities of primary emission from the chamber were measured before the application of any additional reactants to the chamber. The density of the aged particles in the chamber were measured 4.5 hours after the addition and stabilization of the reactants (for dark aging) and/or turning on of the UV lights (for photochemical aging; Table S4). Simultaneous to the density measurements, particles were collected on holey-carbon grids (S147-4 Holey carbon film 400 Mesh Cu; Agar Scientific Inc.) from the chamber using an aspiration sampler at a flowrate of 0.3 litres per minute (lpm). Subsequently, we performed Scanning Electron Microscopy (SEM, Sigma HD/VP; Carl Zeiss NTS) to investigate the morphology of both chamber diluted primary and aged BB emitted particles.

2.5 Offline optical analyses:

267 Biomass burning emissions were collected on precombusted 90 mm Quartz microfiber filter (Pallflex™
268 Tissuquartz™ 7203, Pall Corporation) from the fresh emissions and at the end of each chamber experiment.
269 First, the freshly emitted particles from the raw exhaust without any additional dilution were deposited on the
270 filter at a flow rate of 90 lpm. For experiments with subsequent chamber study, the sampling was done for
271 the same duration as the chamber feed (Fig.1). Approximately 4.5 hours after the chamber feeding, another
272 filter sample was collected from the chamber with the same flow rate for 120 minutes. For the boreal (FIA
273 and FIB) and arctic peat (RUS and NOR) samples, the primary emission was deposited on the filter
274 throughout the whole combustion period, and no chamber studies were performed.

275 Thermal–optical carbon analysis was carried out with the IMPROVE-A protocol (Chow et al., 2007) in a
276 carbon analyzer (Lab OC-EC Aerosol Analyzer; Sunset Laboratory Inc.) by placing 1.5 cm² punches of the
277 Quartz fiber filters (QMA) containing deposited aerosol particles. The details of the measurement protocol
278 have been described in the supporting information (Section S2). Additionally, two separate 1.5 cm² filter
279 punches from each experiment were extracted, one with ultrapure Mili-Q water (>18.2 MΩ) and the other
280 with MeOH (Fisher Scientific, Analytical Reagent Grade ≥99.9% pure). After solvent extraction, the two
281 filters were dried under gentle airflow in a clean room for 12 hours before being analysed with the same OC-
282 EC analyzer. This setup (Fig. S4) enabled us to measure the dissolved organic concentration in the solvent
283 using Eq. 4 (see section 2.8) and therefore estimate the MAE_{OC} (Eq. 3).

284 **Table 1: List of chamber experiments with detailed parameterization of combustion and aging conditions (mean $\pm$ standard deviation)**

No.	Fuel	Combustion Condition (no. of replicates)	MCE	OC EF (fresh emission) (g kg ⁻¹)	EC EF (fresh emission) (g kg ⁻¹)	Final DR (chamber)	SMPS PM ₁ EF (g kg ⁻¹) (chamber primary)	GMD (nm)	Aging condition (in chamber)	NMVOC/NO _x (ppbC/ppb)	OH exposure (x 10 ¹¹ molecules.cm ⁻³ . s ⁻¹)	eqv. age (days)
1a	CP	flaming (n = 2)	0.971 $\pm$ 0.004	5.34 $\pm$ 2.32	0.29 $\pm$ 0.08	247 $\pm$ 106	6.98 $\pm$ 4.01	52.4 $\pm$ 1.60	photo aged	21.4 $\pm$ 4.40	2.09 ($\pm$ 0.33)	1.61 $\pm$ 0.26
1b		flaming (n = 1)	0.979	1.85	0.40	229	3.86	48.0	high NO _x dark aged	5.45	NA	
1c		flaming (n = 1)	0.986	1.39	1.81	273	8.05	54.4	no aging	6.40	NA	
1d		smouldering (n = 2)	0.619 $\pm$ 0.147	58.3 $\pm$ 31.5	0.24 $\pm$ 0.07	764 $\pm$ 118	152 $\pm$ 89	48.3 $\pm$ 5.20	photo aged	88.6 $\pm$ 5.01	1.15 ($\pm$ 0.63)	0.88 $\pm$ 0.48
1e		smouldering (n = 1)	0.793	26.8	0.16	429	33.6	45.5	high NO _x dark aged	4.20	NA	
1f		smouldering (n = 1)	0.479	73.7	0.49	666	228	59.7	no aging	108	NA	
2a	BFS	flaming (n = 3)	0.922 $\pm$ 0.003	3.20 $\pm$ 0.94	0.43 $\pm$ 0.15	309 $\pm$ 199	6.07 $\pm$ 1.05	51.0 $\pm$ 4.10	photo aged	63.9 $\pm$ 41.7	1.80 ($\pm$ 0.34)	1.39 $\pm$ 0.26
2b		flaming (n = 3)	0.952 $\pm$ 0.005	3.18 $\pm$ 0.70	0.39 $\pm$ 0.08	387 $\pm$ 140	4.79 $\pm$ 0.82	51.6 $\pm$ 0.70	high NO _x dark aged	4.78 $\pm$ 1.23	NA	
2c		flaming (n = 1)	0.961	3.06	0.73	833	5.95	53.6	no aging	20.0	NA	
2d		smouldering (n = 3)	0.759 $\pm$ 0.020	23.9 $\pm$ 15.7	0.08 $\pm$ 0.07	276 $\pm$ 100	48.3 $\pm$ 29.5	84.7 $\pm$ 41.4	photo aged	133 $\pm$ 104	1.57 ($\pm$ 0.11)	1.21 $\pm$ 0.09
2e		smouldering (n = 3)	0.761 $\pm$ 0.017	24.0 $\pm$ 10.5	0.07 $\pm$ 0.06	287 $\pm$ 123	26.6 $\pm$ 21.1	73.2 $\pm$ 22.8	high NO _x dark aged	23.5 $\pm$ 19.6	NA	
2f		smouldering (n = 1)	0.774	15.3	0.33	135	23.96	51.2	no aging	163	NA	
3a	SW	flaming (n = 1)	0.935	8.04	0.97	922	12.4	60.5	photo aged	26.1	1.41	1.09
3b		flaming (n = 1)	0.950	5.93	0.60	244	10.5	48.0	low NO _x dark aged	43.9	NA	
3c		smouldering (n = 1)	0.589	87.0	5.18	389	61.1	81.0	photo aged	53.2	1.29	0.99
3d		smouldering (n = 1)	0.892	22.5	1.36	416	31.1	62.6	low NO _x dark aged	69.7	NA	
4a	SG	flaming (n = 1)	0.943	4.83	0.31	512	6.24	48.1	photo aged	124	1.46	1.13
4b		flaming (n = 1)	0.973	2.87	0.28	1974	5.44	50.3	low NO _x dark aged	2.73	NA	
4c		smouldering (n = 1)	0.803	31.8	1.19	4045	28.6	60.5	photo aged	19.3	1.49	1.15
4d		smouldering (n = 1)	0.843	14.0	1.25	902	23.2	58.5	low NO _x dark aged	108	NA	

For filters containing fresh emissions collected from the raw exhaust, 40 mL of solvent was used for the extraction of the 1.5 cm² filter area. The volume of solvent used for extraction of highly loaded filters containing fresh emissions can be a limiting factor for dissolved OC concentration, especially for BB emitted particles that have been shown to be dominated by non-polar molecules (Lin et al., 2017) that are less soluble in water. We chose 40 ml of solvent, as this volume provided a dilute micromolar solution in which the light absorption by the dissolved organics would be linearly proportional to their concentration, in accordance with Beer-Lambert's law (Huang et al., 2018). The utilized solvent volume was also sufficient to dissolve the sparingly soluble low volatility strongly light-absorbing fraction of OC. On the other hand, for the chamber diluted fresh emissions and aged particles, two 1.5 cm² filter punches (total of 3 cm²) were extracted in 20 mL water or MeOH (Cao et al., 2021; Fan et al., 2018) to obtain solution phase organic concentrations necessary for analytically significant S/N ratio in the UV-vis spectrophotometry.

In parallel to the deposition of primary particles from the raw exhaust on quartz fiber (QMA) filters for emission factor calculation, a fraction of the emitted particles was fed to the TeflonTM chamber to create real life atmospheric dilution. It is important to study these particles in a diluted system to understand their chemical and optical evolution in the atmosphere. The particle size distribution, effective density, and morphology of the primary BB emission were studied after feeding them in the chamber and letting them mix homogeneously with clean air. Subsequently, different oxidants were added to the chamber to initiate photochemical or dark oxidation reactions. After 4.5 hours of feeding the oxidants, the particles from the chamber were deposited on another QMA filter. In addition to that, five separate experiments were conducted for different BB emissions, where no oxidants were directly injected to the chamber and the primary particles were on QMA filter 4.5 hours after the chamber feed. These samples are referred to as "chamber primary" samples. The QMA filters containing chamber aged and chamber primary particles were produced to undergo OC-EC analyses and solvent extraction similar to the QMA particles collecting raw exhaust primary particles.

The filter punches were submerged in the solvents and sonicated for 10 mins in three separate intervals in an optimized ultrasonicator (SONOREX Digitec, Bandelin Inc.). The optimization enabled a much gentler sonication as compared to regular commercial ultrasonicators (Huang et al., 2018; Li et al., 2020), resulting

313 in high extraction efficiencies without damaging the filter punch and thereby allowing subsequent OC-EC
314 analyses. Such gentle ultrasonication also minimised the dislodging of insoluble organics or EC from the
315 filter to the solution (Phillips and Smith, 2017). During the intervals between the 10-minute sonication
316 windows, the samples were kept in a refrigerator to keep the effects of any thermal disintegration of OC or
317 chemical transformation through reactions with the solvent (Bateman et al., 2008; Chen et al., 2022) due to
318 the added kinetic and thermal energy during the sonication at minimal.

319 After the sonication, the elute was passed through 0.2 µm hydrophilic PTFE syringe filters (Fisherbrand) to
320 remove any insoluble particles from the solution. Aliquots of 3 mL from the filtered solutions were taken in a
321 quartz cuvette with 1 cm path length and UV-vis spectra were recorded for the wavelength range of 250-700
322 nm using a spectrophotometer (UV-2401 PC, Shimadzu). The overall contact time between the solvent and
323 the extracted organics in the solution never exceeded 2 hours before the recording of the optical spectra.
324 These were much smaller time intervals compared to the reaction rates between MeOH and carboxyl groups
325 for instance (McIntyre and McRae, 2005), enabling us to disregard any potential artifact caused by the
326 solvent (MeOH) to the chemical composition of the original OC deposited on the filter (Lin et al., 2012).

327 **2.6. Residential wood combustion experiments**

328 In addition to different open BB emissions, filter samples from experiments performed with modern
329 European logwood-fired chimney stove (Aduro 9-3) operated with beech logs from two separate
330 experimental campaigns were included in the study. This enabled us to extend the analysed sample set to
331 conditions representing high-temperature biomass combustion, and to test the validity of the BrC-BC
332 continuum. The general experimental setups of these two sets of experiments have been presented in our
333 previous works (Ihalainen et al., 2019a, Mukherjee et al., 2024). Each experiment consisted of four
334 (Ihalainen et al., 2019a) or six (Mukherjee et al., 2024) 45 min batches of wood log burning, ignited with
335 wood sticks as kindling. Photochemical aging of the residential wood combustion (RWC) exhaust was
336 conducted by the Photochemical Emission Aging flow tube reactor (PEAR, Ihalainen et al., 2019b), in
337 similar conditions as the low-dilution aging in Hartikainen et al. (2020). Briefly, exhaust samples were
338 collected onto quartz fiber filters before and after the PEAR in a flow rate of 10 lpm for 60 min, with each
339 sample consisting of one full batch and 20 min of the subsequent batch, either from the cold stove (consisting

of 1st and 2nd batch of wood) or in the warm stove (consisting of 3rd and 4th batches of wood) combustion.
The filter samples were extracted similarly to the open biomass burning samples for optical analyses.

2.7. Optical data processing:

The raw absorption (A) of the extracted solution measured by the UV-vis spectrophotometer was converted to the light absorption coefficient (β_{abs} , Mm⁻¹) in the sampled air using Eq. 2 (Hecobian et al., 2010):

$$\beta_{abs}(\lambda) = \frac{(A_{\lambda} - A_{700}) \times V_l \times \ln(10)}{V_a \times l} \quad (2)$$

where A_{λ} is the raw absorbance measured by the spectrophotometer in base-10 at wavelength λ and A_{700} is the absorption at 700nm (which was subtracted from A_{λ} to correct for baseline drift caused by the absorption), V_l is the volume of the extract, V_a (m³) is the volume of the air sampled through the quartz filter during emission measurements, l is the path length of light through the solution, which is equivalent to the width of the quartz cuvette (0.01 m):

The mass absorption efficiency (MAE) of the extracted BrC at a particular wavelength λ is given by Eq. 3 (Liu et al., 2013):

$$MAE(\lambda) = \frac{\beta_{abs}(\lambda)}{C} \quad (3)$$

where C is the concentration of the dissolved organic in a particular solvent (g m⁻³). C for MeOH and water soluble organics (C_{MSOC} and C_{WSOC} respectively) were calculated as:

356

$$C_{MSOC} = \frac{(\text{OC in original filter}) - (\text{OC in MeOH extracted filter})}{\text{volume of methanol used for extraction}} \quad (4a)$$

and,

$$C_{WSOC} = \frac{(\text{OC in original filter}) - (\text{OC in water extracted filter})}{\text{volume of water used for extraction}} \quad (4b)$$

360

C_{WSOC} was also directly measured from the aqueous filter extracts using a total organic carbon (TOC/TN) analyzer (TOC-L, Shimadzu) to compare with the estimates obtained using Eq. 4b. Overall the two methods yielded comparable C_{WSOC} values ($R^2 = 0.98$, Fig. S5.), so for consistency we have used the filter based C

values for both MSOC and WSOC in calculating MAE. The solubility of organics in each solvent were estimated as:

$$\text{solubility} = \frac{(\text{OC in original filter}) - (\text{OC in extracted filter})}{(\text{OC in original filter})} \quad (5)$$

The imaginary part of the refractive index, k , was related to MAE at any given wavelength λ according to Jennings et al. (1979):

$$MAE(\lambda) = \frac{4\pi k(\lambda)}{\rho \lambda} \quad (6)$$

where ρ is the density of the dissolved organic (g cm^{-3}). For fresh, photochemically aged and dark aged emissions different mean density values were used for calculating k as mentioned in Table S4. For the RWC experiments, particle densities measured for fresh and PEAR-aged beech combustion exhaust particles from the same appliance (Mukherjee et al., 2024). were applied for the calculation of k . Specifically, density values of $1(\pm 0.1) \text{ g cm}^{-3}$ and $1.6(\pm 0.1) \text{ g cm}^{-3}$ were used for the primary and aged RWC exhaust particles, respectively. It should be noted that the MAE values obtained in this study for soluble OC will need to be divided by an OA/OC factor (typically in the range of 1.8-2.2 for fresh emissions, Hartikainen et al., 2020) to obtain the MAE of MeOH or water-soluble OA before comparing with previous literature reporting light absorption properties of BB emitted OA.

Absorption Ångström exponent (AAE) of MSOC and WSOC were estimated between a pair of wavelengths λ_1 and λ_2 as the power law exponent of the ratio between the absorption coefficients at the two wavelengths (Moosmüller et al., 2009), such as:

$$AAE(\lambda_1, \lambda_2) = \frac{\ln\{\beta_{abs}(\lambda_1)/\beta_{abs}(\lambda_2)\}}{\ln(\lambda_2/\lambda_1)} \quad (7)$$

For this study, AAE between wavelengths 300-550 nm were used to describe the optical behaviour of BrC emitted from different biomass sources. The wavelength dependence of the imaginary refractive index k is denoted as w (Saleh et al., 2014) which we derived for fresh and chamber samples using a similar power law relationship as Eq. 7 but involving k at two different wavelengths. In literature, w has also been estimated using AAE (Saleh et al., 2014, McClure et al., 2020) as they are related by:

$$AAE \approx w + 1 \quad (8)$$

2.8 Emission Factor Calculation

Emission factors from the combustion experiments were calculated by the carbon mass balance method such (Eq. 9; Yokelson et al., 1999)

$$EF_X = F_C \cdot 1000 \cdot \frac{ER_X}{\frac{\sum \Delta C}{\Delta CO}} \quad (9)$$

where F_c is carbon fraction in the combusted sample and the summation $\sum \Delta C$ is the total carbon released during the combustion. For savanna biomasses (SG and SW) $\sum \Delta C$ was estimated as (Vakkari et al., 2025):

$$\sum \Delta C = \Delta CO_2 + \Delta CO + \Delta CH_4 + \Delta OC + \Delta rBC + \sum \Delta C_{VOC} \quad (10 a),$$

ΔCO_2 , ΔCO and ΔCH_4 were their respective concentrations in ppm subtracted by background and synthetic air concentration, ΔOC was measured by AMS, rBC was measured by SP2 and VOCs were measured by VOCUS. For commercial peat (CP), boreal forest surface (BFS) and natural peatland (FIA, FIB, RUS and NOR) emissions, $\sum \Delta C$ was estimated as:

$$\sum \Delta C = \Delta CO_2 + \Delta CO + \Delta CH_4 + \Delta OC + \Delta EC + \sum \Delta C_{VOC} \quad (10 b),$$

For CP and BFS, CO_2 , CO and CH_4 were measured using Picarro and non-methane VOCs (NMVOC) were estimated from FTIR (Table S2). For natural peatland samples CO , CH_4 and NMVOCs were all measured by FTIR, while CO_2 was measured by a CO_2 analyzer (Siemens). OC and EC for all of these samples were estimated from $OC-EC$ analyses of filters.

ER_x in Eq. 9 is the ratio of measured concentration in $\mu g m^{-3}$ relative to the CO carbon concentration in $\mu g C m^{-3}$. ER_x was calculated by Eq. 11, where the CO concentration measured in ppm is converted to CO carbon concentration in $\mu g C m^{-3}$ using the following equation:

$$\Delta ER_X = \frac{X}{\frac{12.01 g/mol \cdot 101325 Pa}{8.31451 J / mol \cdot K \cdot (295.15 K + T)} \cdot \Delta CO} \quad (11)$$

Where X is the concentration of the measured parameter in $\mu g m^{-3}$, ΔCO is the CO concentration in the emission in ppm compared to background, T is measured temperature in the chamber in Kelvin (295.15 K or

412 22°C), 12.01 is the molecular weight of carbon (g mol^{-1}), 101325 standard atmospheric pressure in Pa,
413 8.31451 is gas constant in SI unit.

414 **2.9 FT-ICR MS analyses:**

415 Chemical compositions of the biomass burning emission samples collected on quartz filters were
416 characterized by means of ultrahigh-resolution mass spectrometry. All experiments were performed using a
417 12-T Bruker solariX XR Fourier transform ion cyclotron resonance mass spectrometer (FT-ICR MS) (Bruker
418 Daltonics GmbH, Bremen, Germany), equipped with a dynamically harmonized ICR cell (ParaCell®). The
419 mass spectrometer was coupled to an Apollo-II atmospheric pressure chemical ionization (APCI) source,
420 fitted with a direct insertion probe (DIP) accessory. This set-up enabled chemical characterization of the
421 filter samples directly with minimal sample preparation. Five layers of each quartz filter sample were packed
422 into a prebaked glass capillary (Hirschmann melting point tube) and inserted into the ion source vaporizer,
423 held at 370 °C. The capillary voltage was set to 4500 V and corona current to 4000 nA. Dry nitrogen was
424 used as the drying (3.5 Lmin^{-1} , 220 °C) and nebulizing (2.0 bar) gas. After an induction time of 10 s, the MS
425 data were recorded until the total ion current plateaued (i.e number of scans ranged from 15 to 35), indicating
426 that the entire aerosol sample was completely desorbed at the given temperature.

427 All DIP-APCI FT-ICR measurements were conducted in a positive ion mode. The generated ions were
428 accumulated in the hexapole ion trap and transferred to the ICR cell for trapping, excitation and detection.
429 The instrument control and data acquisition were performed by Bruker fimsControl 2.1 software. For each
430 spectrum, a broadband frequency excitation and detection were carried out with 4 MWord time-domain
431 transients (transient time 1.1 s), which were full-sine apodized and zero-filled once to provide the final 8
432 MWord magnitude-mode data spanning m/z range of 100–2000. The time-of-flight and ion accumulation
433 time settings were 1.0 ms and 0.30 s, respectively.

434 The FT-ICR instrument was externally m/z -calibrated prior to the measurements using a polystyrene (PS)
435 standard covering a wide mass range and reaching accuracies generally below 1 ppm. The initial spectral
436 post-processing was done with Bruker DataAnalysis 5.1 software, including the internal re-calibration of the
437 m/z -axes with a custom-made mass list for organic aerosol samples. For the peak picking, a signal-to-noise

(S/N) ratio was set at ≥ 6 . PetroOrg IS 18.0.3 software (Omics LLC, Tallahassee, FL, USA) was used for the molecular formula assignments with the following constraints: double bond equivalent (DBE) 0–40; mass error ± 1.0 ppm; atomic formula $^{12}\text{C}_{1-100}^{1}\text{H}_{1-200}^{14}\text{N}_{1-4}^{16}\text{O}_{1-15}^{32}\text{S}_{1-4}$. The elemental composition boundaries for the annotation were chosen based on careful manual inspection of spectra identifying the edges of the observed chemical space. The time-resolved spectral information of the DIP experiment had been summed to an average spectral read back for each measurement.

For data interpretation and visualization of the complex lists of attributed sum formulae, we used established data grouping and fingerprint diagrams (Schneider et al., 2024a; 2024b). Visualization and pre-processing, calculating molecular properties and diagnostic measures from the sum formulae were performed via MATLAB (MATLAB R2023a, MathWorks Inc., MA). Characteristic molecular properties encompassed double bond equivalents (DBE), aromaticity index (AI), saturation vapor pressure (C^*) and average carbon oxidation state (OS_C) frequently used in ultra-high resolution mass spectrometric studies of complex environmental sample materials (Koch and Dittmar, 2006; Kroll et al., 2011; Li et al., 2016)

2.10: SP-AMS analyses:

This study employed a soot particle aerosol mass spectrometer (SP-AMS; Aerodyne Research Inc., Billerica, MA, USA; Onasch et al., 2012) to analyze the concentration levels, mass spectral signatures, and size distributions of non-refractory (organics, sulfate, nitrate, ammonium, and chloride) and refractory (e.g. metals, rBC) components. The SP-AMS enhances the capabilities of the standard AMS by incorporating a laser vaporizer, which enables the analysis of refractory aerosol components. While the instrument can function using only the laser vaporizer, both the laser and tungsten vaporizers were used in this study to ensure comprehensive detection of mentioned chemical species. Size-resolved measurements were obtained using particle time-of-flight (PToF) mode, with the SP-AMS aerodynamic lens enabling detection of particles ranging from roughly 50 nm to 1 μm . The instrument operated with a time resolution of 60 seconds, with about half of the time measuring in mass spectrum mode and the other half in PToF mode. A calibration of the SP-AMS based on particle mass was carried out using size-selected, dried particles of ammonium nitrate and ammonium sulfate. This approach allowed for the determination of an effective nitrate response factor, as well as the relative ionization efficiencies (RIEs) for ammonium (RIE_{NH_4}) and sulfate (RIE_{SO_4}),

by converting the instrument signals into nitrate-equivalent mass concentrations. The determined RIE value for NH_4 was 3.4 while the one for sulfate was 0.9. A default RIE value of 1.4 was used for organics. SP-AMS data processing was conducted using the AMS analysis tools SQUIRREL (version 1.63B) and PIKA (version 1.23B) within Igor Pro 8 (Wavemetrics, Lake Oswego, OR).

3. Results

3.1 Emission factors of OC and EC in fresh emission

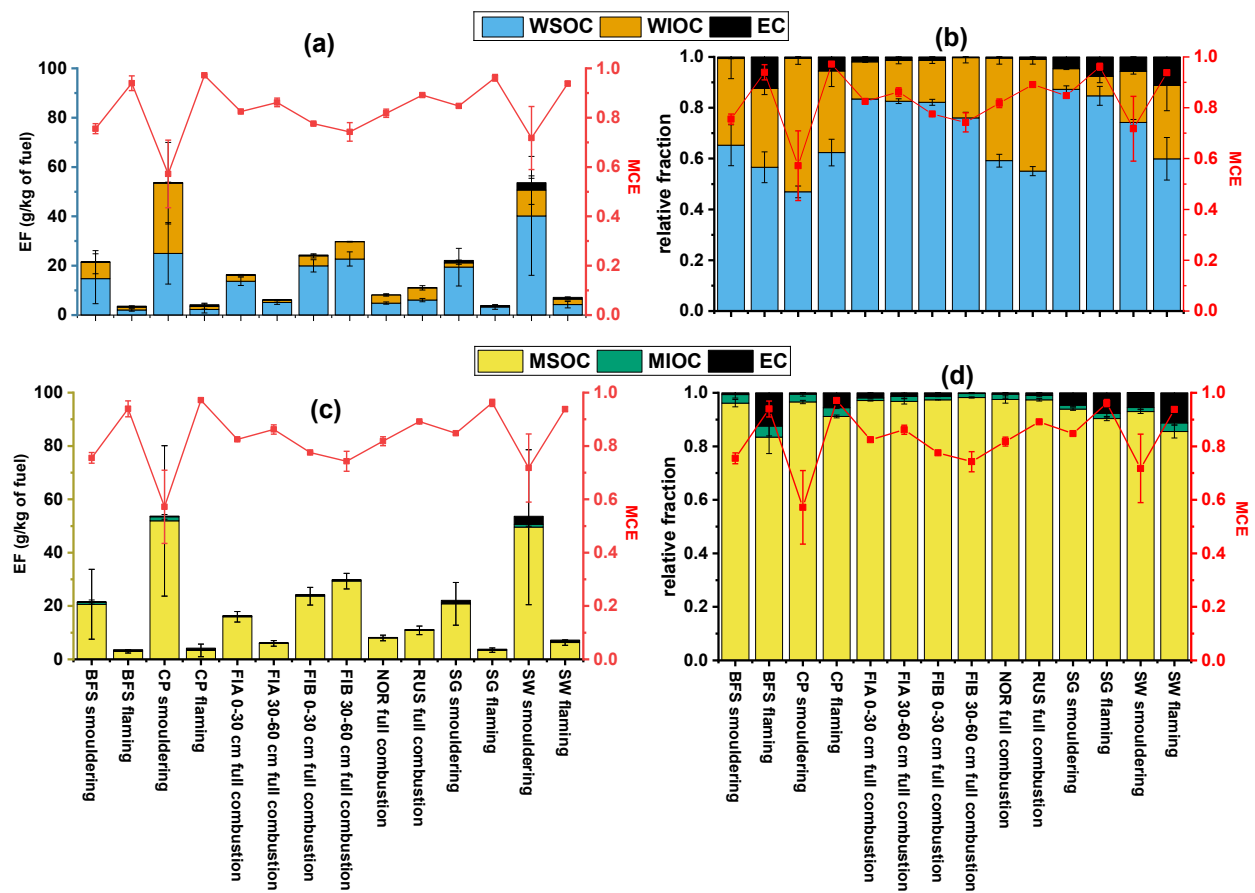
The emission factors of OC (EF_{OC}) and EC (EF_{EC}) were determined based on thermal-optical carbon analyses of filters collected from fresh exhaust aerosol (Fig. 2, Fig. S6). Generally, we observed higher EF_{OC} for smouldering combustions with low MCE, as expected. For EC however, the trend was reversed, with flaming combustion having higher MCE producing more EC per kg of fuel (Fig. S6). The combustion emissions in our experiments were OC rich, most likely due to the open setup of the burner, which resulted in lower combustion temperature compared to wood stove emissions, due to high air-to-fuel ratios and because there is no combustion chamber around the fire to retain the heat in the surroundings. However, there were significant variations in EF_{OC} obtained from different combustion conditions and for different biomasses, ranging from 1.39-7.44 g kg^{-1} for flaming dominated combustions and 6.12-89.9 g kg^{-1} for smouldering dominated combustions. CP and SW samples had the largest EF_{OC} of 53.4(± 29.0) g kg^{-1} and 50.6(± 29.8) g kg^{-1} respectively, during the smouldering combustion phase. In contrast, full combustion of different natural peat samples yielded EF_{OC} in the range of 5.06-32.7 g kg^{-1} , with FIA (at depth of 30-60 cm) having the smallest (6.09 ± 1.2 g kg^{-1}) and FIB (at depth of 30-60 cm) having the largest (29.8 ± 2.92 g kg^{-1}) average EF_{OC} among peat samples. Interestingly, the average MCE values and EF_{OC} reported for field measurements of subtropical Indonesian peat fires (Stockwell et al., 2015; Jayarathne et al., 2018) and laboratory studies of North American (Black et al., 2016; Chakrabarty et al., 2016), West European (Iinuma et al., 2007) and South-east Asian peat fires (Christian et al., 2003; Iinuma et al., 2007) fell inside the range of our reported EF_{OC} for laboratory combustion of Finnish (boreal) and arctic peat samples. In our study, flaming and smouldering combustion of BFS samples had average EF_{OC} of 3.17(± 0.77) g kg^{-1} and

21.5($\pm$ 13.8) g kg⁻¹ respectively, while previously reported EF_{OC} for North American boreal forest fires were in similar range of 5.9($\pm$ 2.5) g kg⁻¹ (Andreae, 2019). Savanna wood and grass (SW and SG) samples burnt under flaming conditions in this study were estimated to have average EF_{OC} of 6.47($\pm$ 0.98) g kg⁻¹ and 3.56($\pm$ 0.91) g kg⁻¹ respectively, which were somewhat higher in comparison to previously reported organic emission factor of 3.0($\pm$ 1.5) g kg⁻¹ (Andreae, 2019) and 2.62($\pm$ 1.24) g kg⁻¹ (Akagi et al., 2011) from high MCE savanna fires.

Emissions from savanna biomass had higher average EF_{EC} compared to the other biomasses, with an average EF_{EC} of 3.03($\pm$ 1.77) g kg⁻¹ and 0.94($\pm$ 0.16) g kg⁻¹ from the smouldering emissions of the woody (SW) and grassy (SG) fuels respectively. The carbonaceous fraction of flaming dominated emissions from the woody savanna samples (MCE $\sim$ 0.94 $\pm$ 0.01) consisted of 12($\pm$ 2) % of EC while savanna grass had an EC content of 7.4($\pm$ 1.5)% (MCE $\sim$ 0.96 $\pm$ 0.01). Previously reported average EF_{EC} values for savanna and grassland fires ranged from 0.37($\pm$ 0.20 g kg⁻¹) (Akagi et al., 2011) to 0.53($\pm$ 0.35) g kg⁻¹ (Andreae, 2019) while Vakkari et al. (2018) reported EF_{EC} of 0.67 g kg⁻¹ for South African savanna grass burning. These values fell in between our estimated EF_{EC} from flaming burn of savanna grass (0.27 $\pm$ 0.01 g kg⁻¹) and wood (0.72 $\pm$ 0.17 g kg⁻¹). Full combustion (MCE 0.74-0.89) of the Finnish peat samples (FIA and FIB) collected at the depth of 30-60 cm from the surface level and arctic permafrost peat samples from Svalbard (NOR) and Russia (RUS) had the lowest measured average EF_{EC} (in the range of 0.02-0.13 g kg⁻¹), which matched well with previously reported EF_{EC} from field and lab studies of subtropical Indonesian (Stockwell et al., 2015; Christian et al., 2003) and laboratory replicates of North American peat fires (Black et al., 2016). Flaming and smouldering combustions of CP had average EF_{EC} of 0.70($\pm$ 0.65) g kg⁻¹ and 0.28($\pm$ 0.14) which resembled EF_{EC} reported from laboratory studies of Western European and South-East Asian peat fires (Iinuma et al., 2007). Burning of the boreal forest surface sample (BFS) resulted in higher EC emissions than any of the peat samples with an average EF_{EC} of 0.45($\pm$ 0.16) g kg⁻¹ for high MCE ($\sim$ 0.94 $\pm$ 0.01) combustion and an average EF_{EC} of 0.11($\pm$ 0.09) g kg⁻¹ for low MCE ($\sim$ 0.76 $\pm$ 0.02) smouldering burns. For North American boreal forest fires, Andreae (2019) reported EF_{EC} of 0.43($\pm$ 0.21) g kg⁻¹ with the fire having an average MCE of 0.89($\pm$ 0.04), which resembles Finnish BFS EF_{EC} for flaming dominated burns. Thus, it seems that our experiments with simulated surface fire conditions at comparably lower combustion

518 temperature typical for Eurasian wildfires yield similar EC emissions as reported for North American
 519 wildfires that are commonly high temperature crown fires. The estimated EC/OC ratios for fresh BB
 520 emissions in this study ranged between 0.003-1.3, which roughly corresponds to BC/OA range of 0.002-0.7,
 521 with the lowest values obtained during smouldering and higher values during flaming dominated emissions
 522 respectively.

523 In comparison, for residential wood combustion (RWC) of North European beech wood logs
 524 (Mukherjee et al., 2024) the respective average OC and EC emission factors were $0.64(\pm 0.31) \text{ g kg}^{-1}$ and
 525 $0.28(\pm 0.04) \text{ g kg}^{-1}$, respectively. Therefore, the OC emission factors for the open burning in the current study
 526 were approximately 4-140 times higher for smouldering emissions and 2-13 times higher for flaming
 527 emissions than our previous estimates of RWC emission. In comparison, we did not observe such a
 528 significant difference between the EC emission factors of RWC and flaming BB emissions. This implies that
 529 surface fires have the potential to be important sources of specifically organic pollutants and BrC in forest
 530 fire prone environments, whereas the BC emissions would not be particularly strong from these fires.



531

532 **Figure 2. Emission factors of water-soluble (WSOC), MeOH soluble (MSOC), water-insoluble (WIOC), MeOH**
533 **insoluble organic carbon (MIOC) and elemental carbon (EC) in absolute scale (a,c) and relative to total EF (b,d).**

534 **3.2 Chemical composition and solubility of fresh organic aerosol**

535 Chemical characterization of the fresh emission samples from whole combustions of peat (FIA, FIB, RUS and
536 NOR) and Finnish BFS have been previously reported by Schneider et al. (2024a) in which electrospray
537 ionization (ESI) was utilized for the FT-ICR mass spectrometry on extracts, targeting the polar constituents.
538 For this study, the filters containing specifically flaming and smouldering emissions of CP and BFS were
539 analyzed directly from the filters by using 12-T DIP-APCI FTICR-MS to identify differences in the organic
540 matter compositions due to both fuel and combustion conditions. The advantage of the DIP-APCI technique
541 used in this study is the ability to detect analytes with low solvent extraction recoveries due to minimal sample
542 preparation required (Rüger et al., 2021). Additionally, APCI was able to address a wide chemical space
543 covering polar up to apolar analytes. Therefore, inspite of the lower mass loadings on the filters for specific
544 combustion phases compared to full biomass combustions as reported in Schneider et al. (2024a), we could
545 assign up to 2000-4000 monoisotopic elemental compositions. Overall, CHO, CHNO and CH compound
546 classes were the most abundant in the fresh BB emissions of CP and BFS. The low temperature smouldering
547 combustions have been shown to emit mostly direct thermal degradation products from biomass pyrolysis
548 (Chakrabarty et al., 2016) compared to flaming combustion (Engling et al., 2006, Popovicheva et al., 2019),
549 with similar chemical composition and molecular structures to that of the fuel (Kourtchev et al., 2011). Similar
550 to the smouldering dominated peat burning emissions reported in Schneider et al. (2024a), we also observed
551 that the CP smouldering emissions were chemically most diverse with the highest number of sum formulae
552 unique to this burning condition (in total 774), thus reflecting the complex inherent compositional variability
553 of the peat samples (Figure 3). CP smouldering emissions also had clearly higher CHN and CHNO fractions
554 than all the other biomasses and combustion conditions. This observation can be attributed to the fact that the
555 CP samples contained only below-surface peat that have decomposed for longer time periods and experienced
556 more microbial activity than the BFS samples, leading to higher N containing species. Flaming combustion of
557 CP emitted the second highest number of unique sum formulae (in total 297) but had much lower CHN class
558 content compared to smouldering conditions, suggesting a considerable effect of the burning condition / MCE
559 on the POA chemical composition (Figure 3).

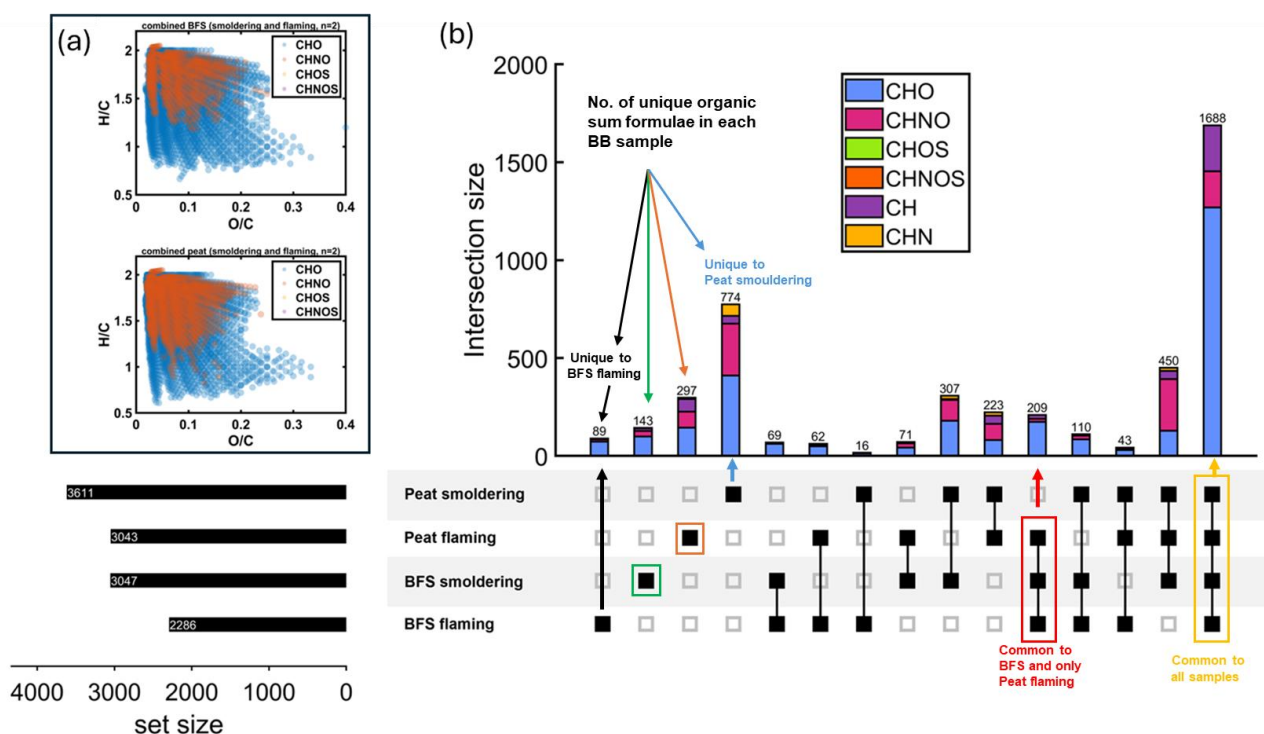
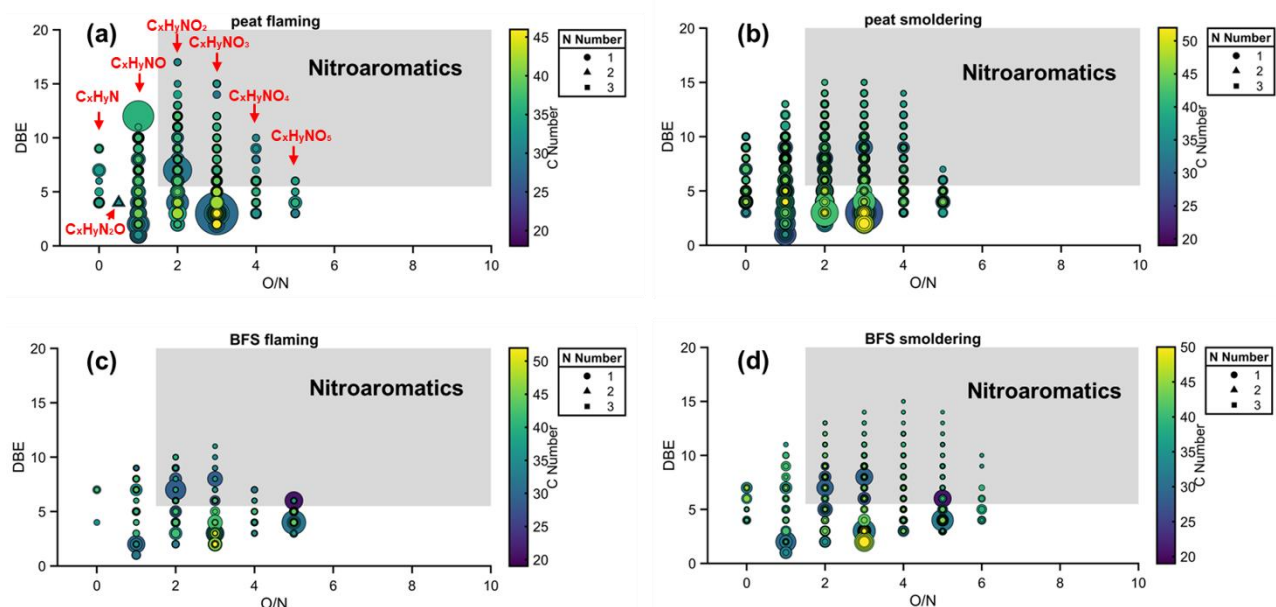


Figure 3: (a) Van-Krevelen diagrams of oxygen-containing compounds and (b) upset plot intersections (combined CP and BFS) with compound class indicated by colour. Samples were analysed by positive-ion DIP-APCI FT-ICR mass spectrometry. Upset plot of four organic aerosol datasets (flaming and smouldering emissions of CP and BFS) with the number of measured molecular formulae in each dataset (bottom left) and the individual intersections (top right) indicated by colour (CHO-light blue, CHOS-green, CH-purple, CHNO-magenta, CHNOS-orange and CHN-yellow). The compared samples are shown in the lower right corner and the common organic sum formulas they contain in the panel on the upper right.

On the other hand, the BFS smouldering and flaming emissions exhibited chemically similar compounds, with nearly 1900 identified molecular formulae shared among all the analysed BFS samples. Almost none of the molecular formulae were unique to BFS smouldering or flaming samples, as the variability among replicates themselves was high. This indicates that although different combustion conditions of BFS strongly influence the OC emission factor, it does not lead to prominent chemical differences in POA. Similar to CP, BFS emissions were also dominated by CHO, CHNO and CH classes, although a much lower fraction of CHN was present in the BFS smouldering emission compared to CP. Furthermore, a more detailed inspection of the identified formulae of the nitrogen-containing molecules in the fresh CP and BFS emissions reveals that they mostly belonged to the CHNO class, contained one nitrogen substitution, and consisted of

both aliphatic and aromatic nitro-organic compounds (Figure 4). Specifically, nitroaromatics were more abundant in the peat samples, which is relevant, as nitroaromatics are known to be prominent BrC chromophores absorbing light in the wavelength range of 360-600 nm (Fleming et al., 2020).



581

Figure 4: Detailed chemical characterization of CHN and CHNO compounds in (a) CP flaming, (b) CP smouldering, (c) BFS flaming and (d) BFS smouldering fresh emissions. The x-axis shows O:N ratio and y-axis corresponds to double-bond equivalence (DBE). Nitroaromatic compounds fall into the shaded (grey) area in the figures.

Due to their different chemical compositions, OC from various fresh BB emissions exhibited variable solubility in water and MeOH (Fig. 2, Fig. S11). While MeOH extracted almost all the OC (~95%) from the filter irrespective of fuel or combustion type, the water-soluble fraction of OC (WSOC) exhibited fuel dependent behaviour. For example, both FIA and FIB emissions exhibited higher WSOC fractions (76-83%) in the total organic carbon (TOC) compared to arctic permafrost peat samples NOR (~59% of TOC) and RUS (~55% of TOC). This has been previously elaborated by Schneider et al. (2024a), who found that in comparison to FIA and FIB samples, NOR and RUS peat combustion emissions were constituted by less oxidized primary organic carbon (POC), which explains the diminished solubility in a polar solvent like water (Budisulistiorini et al., 2017). Among the biomass samples used in this study, SG showed the highest WSOC fractions for both flaming (~92% of TOC) and smouldering (~91% of TOC) combustion (Fig. 2c-d), suggesting abundance of more oxidized POC in these emissions, while smouldering burning of CP consisted

of the smallest WSOC fraction (~46%). These observations point towards the presence of non-polar organic moieties, which are soluble in MeOH but not in water, as previously reported in the literature (Lin et al., 2017).

3.3 Optical properties of Fresh organic aerosol:

Due to the variation in chemical composition and solubility of the fresh BB emissions, we observed substantial diversity in the optical properties of the freshly emitted organic particles. Since MSOC corresponded to >92% of the total OC in our fresh BB emissions, our estimated MSOC concentrations can be used as a proxy for bulk OC. The absorption angstrom exponent ($AAE_{300-550}$, measured in the range of 300-550 nm) of the particles sampled from the fresh emission for different fuels ranged between 4.4-5.3 and 4.7-6.2 for WSOC and MSOC, respectively (Fig. S7). In general, for all samples, AAE_{MSOC} was found to be marginally higher than the respective AAE_{WSOC} . These values fall in the ranges of previously reported AAE for BB emitted as well as urban MSOC and WSOC (Cao et al., 2021; Fan et al., 2018; Yan et al., 2015; Mukherjee et al., 2020). However, our estimated $AAE_{300-550}$ values were higher than reported values from western-US wildfires for similar wavelength range (1.6-1.8, Chakrabarty et al., 2023) and $AAE_{401-870}$ reported from laboratory burns of canopy (2.69 ± 0.36), litter (1.86 ± 0.20) and mixed (2.26 ± 0.36) coniferous ecosystem from western USA (Selimovic et al. 2018). Interestingly, our reported $AAE_{300-550}$ values matched well with lab burning of rotten logs (4.60 ± 3.73) of Douglas fir (*Pseudotsuga menziesii*) and ponderosa pine (*Pinus ponderosa*) from Western USA and generally fell in the range of previously reported AAE values from FIREX campaigns (FIREX 2016: McClure et al., 2019; FIREX 2019: Zeng et al., 2022) for emissions with OA/BC ratios ranging from 6.6-143 (McClure et al., 2020) or EC/OC ratios of 0.01-0.3, assuming an average OA/OC ratio of 2. All the flaming dominated and full combustion emissions, as well as some smouldering dominated emissions reported in our work, had EC/OC ratios that fell in this range. The largest variability in AAE_{MSOC} in our study was observed among BFS smouldering (5.5-6.2) and flaming (5.3-6.0) samples, which was likely a result of the difference in combustion and heterogeneous vegetation distribution in the burnt BFS samples. The Svalbard peat (NOR) emission displayed AAE_{WSOC} in the range of 4.6-5.2, while its AAE_{MSOC} was constrained in the range of 4.9-5.2. In comparison to open BB, fresh RWC emissions having high soot contents (Mukherjee et al., 2024; wood stove 1 in Figure S7) exhibited much lower AAE for both WSOC and MSOC (2.2-2.4), while the wood

combustions from the previous campaign (Ihalainen et al., 2019a; wood stove 2 in Fig. S7) displayed larger
 AAE_{WSOC} (5.8-6.8) and AAE_{MSOC} (5-5.6). The lower AAE values of RWC emissions indicates that BrC from
 high EC/OC emissions absorb light throughout the measured spectral range, as shown earlier by Saleh et al.
 (2018).
 The MAE values obtained from UV-vis spectroscopy at 365 nm for fresh BB emitted MSOC (MAE_{MSOC_365})
 were constrained between 0.46-1.48 m² g⁻¹ for the experiments. In comparison, MAE_{WSOC_365} was found to
 be in the range of 0.51-1.78 m² g⁻¹. These MAE₃₆₅ values fall within the same range as observed for BBOA in
 previous literature (Park and Yu, 2016; Huo et al., 2018; Moschos et al., 2018; Cao et al., 2021). In general,
 flaming dominated BB emissions had higher MAE_{MSOC_365} values compared to smouldering emissions (Fig.
 5a). Among open BB experiments, the lowest MAE_{MSOC_365} were generally observed for fresh emissions from
 BFS smouldering combustions (0.56±0.10 m² g⁻¹), while flaming combustion of CP (1.31±0.13 m² g⁻¹) and
 SW (1.06-1.38 m² g⁻¹) were estimated to have the highest MAE_{MSOC_365} (Table S4). The MAE_{MSOC_365} for
 smouldering SG (0.68-0.94 m² g⁻¹) and SW (1.05-1.11 m² g⁻¹) emissions were in a similar range to the
 MAC_{OA_370} estimated by Vakkari et al. (2025) and agreed well with of MAC₃₇₀ from African residential
 biomass burning emissions (0.24 – 2.2 m² g⁻¹) as reported by Moschos et al. (2024). Previously estimated
 MAC_{BrC} (at 405 nm) from In-Situ observations made during WE-CAN campaign (2018) in the US and
 ORACLES-2016 and CLARIFY (2017) campaigns over Southern Africa ranged from 0.9 to 1.6 m² g⁻¹
 (Carter et al., 2021). However, it should be noted that our estimated MAE values need to be divided by a
 suitable OA/OC ratio before comparing with MAC_{OA}. In addition, MAE values for high temperature wood
 log combustion in the modern European stove were significantly higher than for open BB. Fresh RWC
 emissions had MAE_{MSOC_365} values in the range of 3.33-16.4 m² g⁻¹, while the MAE_{WSOC_365} values ranged
 from 0.32-4.96 m² g⁻¹. The trend between the EC/OC ratios obtained for different experiments to the
 corresponding MAE_{MSOC} also suggests that EC rich emissions contribute to stronger BrC light absorption
 (Fig. 5a). Formation of BrC internally mixed with soot particles in high temperature BB emissions has been
 shown to contribute to enhanced light absorption in past studies, due to lensing effect (Jacobson et al., 2001;
 Liu et al., 2015; Liu et al., 2017; Zhang et al., 2025), which supports our observation.

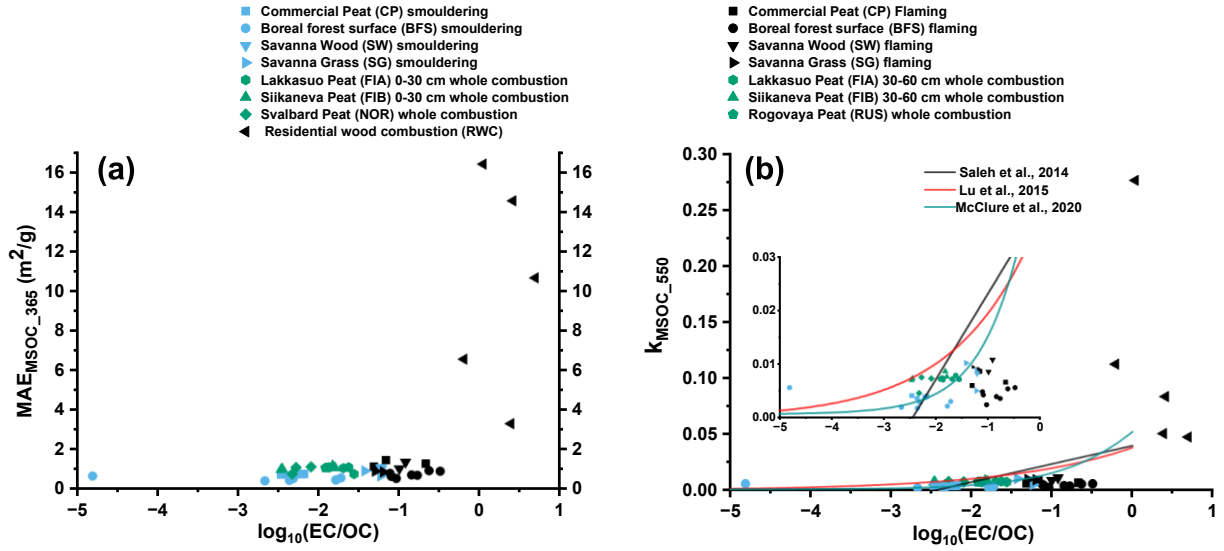


Figure 5: Relationships between MAE_{MSOC_365} (a) and k_{MSOC_550} (b) with EC/OC of open BB and RWC emissions. We observe increasing MAE and k for high temperature combustions with higher EC/OC ratio

The imaginary refractive index measured at the middle of the visible light spectrum at 550 nm (k_{550}) for the BB MSOC varied across orders of magnitude and ranged between 0.002-0.011 (Fig. 5b, Table S4) in our experiment. Similar to MAE, the smouldering emissions exhibited lower k_{MSOC_550} than the flaming dominated burns. As shown in previous studies (Saleh et al., 2018; Saleh et al., 2020) there seem to exist a continuum between the ratio of EC/OC in the fresh BB emission to the BrC light absorption, as we again observed enhanced k_{MSOC_550} for EC rich wood stove emissions (fig. 5b). Most of the fresh BB MSOC fell in the weak BrC regime in the “ k - w ” space (Fig. 6a), proposed by Saleh et al. (2018), while some of the wood stove combustion generated particles, in contrast, were in the strong BrC domain. These results fall in line with previous observations of high temperature (and high MCE) biomass combustions generating “darker BrC”, which exhibit much stronger light absorption compared to lower temperature open BB emissions (Saleh et al., 2018). The data points in k - w space obtained in this study matches reasonably well with previous literature, as shown in Figure 6a.

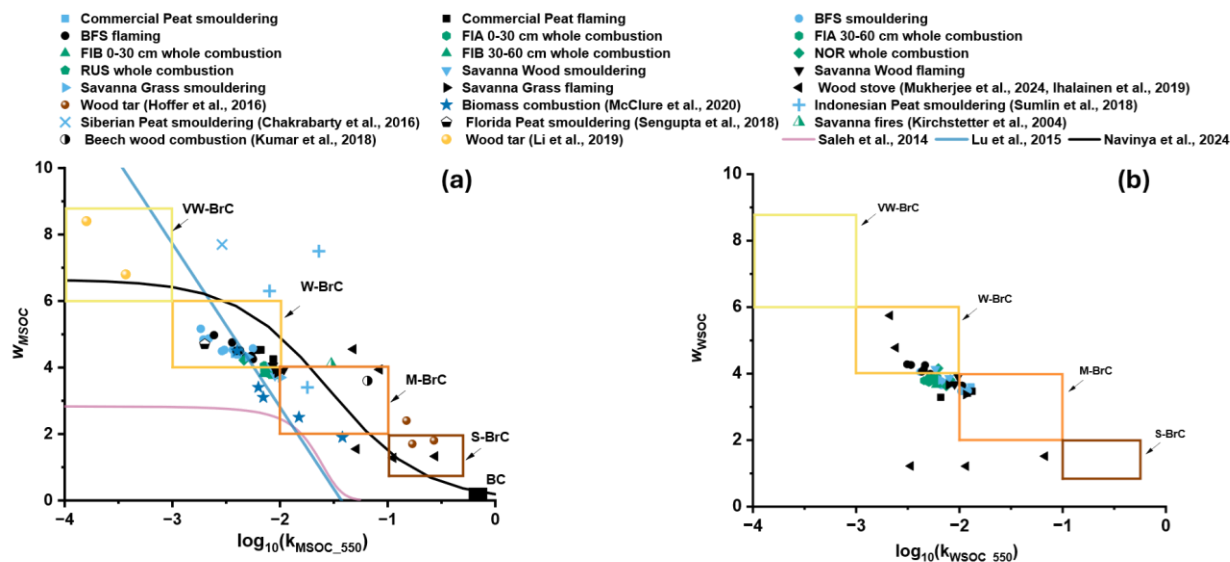


Figure 6: Relationship between w and k_{550} for (a) MSOC and (b) WSOC in fresh emissions from smouldering (light blue points), flaming (black points), full combustion (green points) of open BB performed in this study. Rectangles denote the different light absorbing BrC classes. Several data points from previous literatures (Hoffer et al., 2016; McClure et al., 2020; Sumlin et al., 2018; Chakrabarty et al., 2016; Sengupta et al., 2018; Kirchstetter et al., 2004; Kumar et al., 2018; Li et al., 2019; Saleh et al., 2014; Lu et al., 2015; Navinya et al., 2024) have been added in (a) for reference.

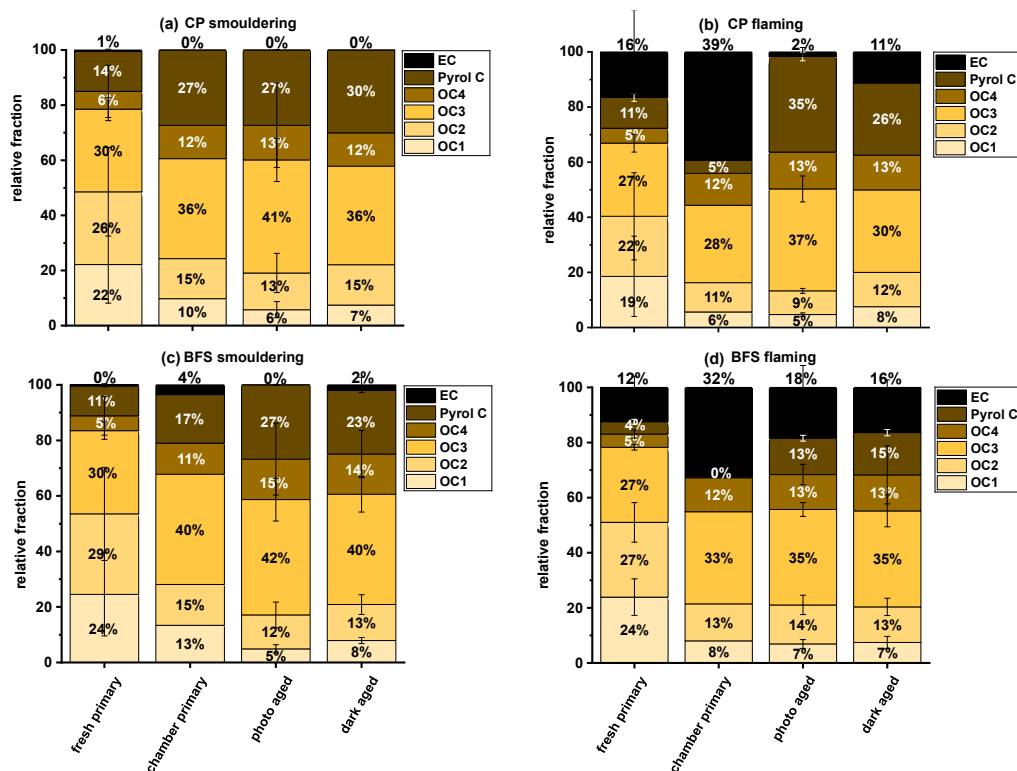
3.4 Particle size distributions and morphologies of chamber diluted primary emissions

Particle size distribution, morphology and effective density were measured for chamber diluted primary emissions. Smouldering combustion of BB generally emitted more particles on average when compared to flaming combustion (Fig. S8). Overall, there was little variability in the particle size distributions for flaming combustion experiments, irrespective of the biomass type. The average particle geometric mean diameters (GMD) ranged between 48 and 85 nm (Table 1). However, we observed clear variation in the emitted particle size distribution among the smouldering combustions, likely due to the unique combustion behaviour of individual samples with varying distributions of different types of biomasses, such as surface vegetation, litter and woody branches. Smouldering combustion of BFS gave rise to distinct bimodal size distributions with the lower mode having a GMD of 30-40 nm, and the larger mode peaking around 100-175 nm. Some replicates of smouldering emissions from CP and SW burning also exhibited bimodal size distribution.

The effective densities of primary particles inside the chamber varied between experimental replicates (Fig S9), which can again be explained by the variability in the combustion and emissions between experiments. The effective densities were largely independent of the particle diameter for all fuel type and combustion conditions (Fig. S9), unlike soot-rich RWC emissions that exhibited size-dependent behaviour in our previous work (Leskinen et al., 2014; Mukherjee et al., 2024). Due to the size-independent behaviour, we averaged over all experimentally obtained density values and estimated average particle densities for each BB emissions (Table S4). While CP and BFS combustion emitted particles had average densities of 1.1 g cm^{-3} and $1.20(\pm 0.05) \text{ g cm}^{-3}$ for both smouldering and flaming dominated combustions, SW and SG emitted particles were denser with average densities of 1.4 g cm^{-3} and $1.65(\pm 0.05) \text{ g cm}^{-3}$ for smouldering combustion respectively. For flaming combustions, SW emitted primary particles had an average density of $1.45(\pm 0.05) \text{ g cm}^{-3}$ and SG particle density was $1.75(\pm 0.05) \text{ g cm}^{-3}$. Little to no size-dependency for particle density was observed in our experiments suggesting near-spherical particle morphology with mobility exponents close to 3 (Leskinen et al., 2023; Corbin et al., 2023). SEM images of chamber diluted primary emissions (Fig. S10) support these results, since mostly round organic particles were observed from the collected grids. These findings agree with the measured chemical compositions that indicated that freshly emitted particles consisted mostly of POC and very little EC, leading to formation of particles with a spherical morphology, so-called tarballs. Tarballs have been previously observed in BB emissions (Chakrabarty et al., 2010; China et al., 2013; Hoffer et al., 2016; Adachi et al., 2019) and atmospheric dilution and aging have been shown to aid the formation of spherical viscous tarball BrC (Hennigan et al., 2011; Sedlacek et al., 2018). In chamber diluted CP and BFS emissions, we observed mostly tarballs (Fig. S10a-d) but SG and SW emissions (Fig. S10e-h) also had soot agglomerates that were partially coated or embedded with organics, as previously reported in China et al. (2013). Notably, the morphology of fresh particles emitted from open BB was very different to those emitted from a wood stove (Mukherjee et al., 2024), where the high temperature combustion formed mostly chain-like fractal soot agglomerate structures which were mostly bare or with some organic inclusions (Fig. S10i; China et al., 2013).

3.5 Effect of chamber dilution on the properties of fresh emission aerosols

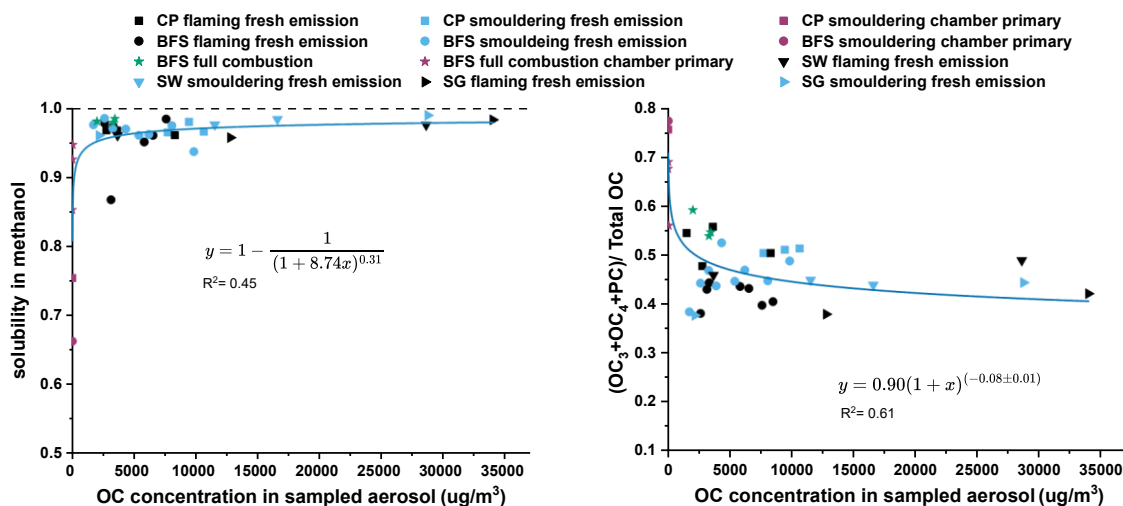
711 The relative fractions of OC1, OC2, OC3, OC4, PC and EC in fresh emissions differed before and after
 712 dilution in the chamber (Fig.7; Table S3), highlighting the effect of dilution on the overall chemical
 713 composition of BB emissions. The relative fraction of the intermediate volatility (IVOC) and semi-volatile
 714 organics (SVOC), namely OC1 and OC2 obtained from IMPROVE-A protocol (Ma et al., 2016) were lower
 715 in the diluted chamber samples than in the fresh emissions, while the low volatile fractions from OC3 to PC
 716 increased (Fig. 7, Table S3). We might have over-estimated the OC1 fraction in fresh emission due to gas
 717 phase VOC adsorption on the filter surface causing positive artifact (Kirchstetter et al., 2001). However, the
 718 significant increase in low volatile OC fractions (OC3, OC4 and PC) in chamber samples indicate dilution
 719 related evaporative loss of the volatile OC fractions in the chamber due to partitioning between gas and
 720 particle phase (Calderon-Arrieta et al., 2024).



721
 722 **Figure 7: The relative abundance of different OC fractions and EC measured in BB emissions from (a) fresh**
 723 **emission, (b) fresh emission after dilution in chamber, (c) photochemically and (d) dark aged emission in the**
 724 **chamber. Error bar indicates the variability among experimental replicates**

725 The partitioning of OC1 and OC2 fractions between gaseous and condensed phases is largely influenced by
 726 the prevailing concentrations in the collected aerosol. Thus, a clear trend is seen between the share of low

727 volatile OC fractions (OC3+OC4+PC) to the total OC and the collected aerosol OC concentration (Fig. 8).
 728 The dilution and consequent evaporation of the most volatile OC also influence the solubility of the POC of
 729 the fresh emission. MeOH extraction efficiency significantly decreased for the more diluted POC in the
 730 chamber (Fig. S11b). This is in line with the change in volatility as the low volatile OC fraction, including
 731 tarball BrC, is generally less soluble in solvents (Chakrabarty et al., 2023; Saleh et al., 2020).



732 **Figure 8: Effect of OC concentration on the partitioning of the OC fractions in BB emissions. Higher dilution in**
 733 **the chamber resulting in lower OC concentration result into larger fraction of low volatile OC fractions and**
 734 **lower solubility in MeOH**

736
 737 The increment in the low volatile OC fractions in chamber diluted primary emissions could also be seen from
 738 the abundance of spherical tarballs in the SEM images (Fig. S15). We postulate, based on indirect evidence
 739 and previous knowledge, that the fresh BB emission collected on the filters probably had poorly formed
 740 tarballs (consisting of higher OC1 and OC2 fractions) and higher MeOH extractable OC. After the dilution
 741 related change in volatility distribution of the OC in the chamber (Calderon-Arrieta et al., 2024), MeOH
 742 insoluble, highly viscous and spherical tarballs were formed in the chamber (Heninghan et al., 2011;
 743 Sedlacek et al., 2018; Adachi and Buseck, 2011). Interestingly, we observed a slight increase in the solubility
 744 of WSOC for chamber primary samples (Fig. S11a). This might be the effect of low OC amount filters
 745 collecting chamber diluted samples, leading to higher water to OC ratio during the extraction. Thus, the

746 volume of water used for extracting chamber primary filter punches allowed the dissolution of sparingly
747 water-soluble organics.

748

749 The change in particle volatility and solubility due to dilution in the chamber did not significantly affect the
750 light absorption wavelength dependency of MSOC and WSOC. The AAE_{MSOC} and AAE_{WSOC} of chamber
751 diluted samples ranged from 5.5-6.1 and 5-5.9, respectively. At the same time, the AAE_{MSOC} and AAE_{WSOC}
752 for the corresponding freshly emitted particles were 5.3-5.6 and 4.6-5.3, respectively. However, we observed
753 a 17% decrease in MAE_{MSOC_365} (Fig. S12) and a 25% decrease in MAE_{WSOC_365} (Fig. S13) in the chamber
754 primary samples compared to fresh emissions for BFS full combustion samples. This suggests that the
755 fraction of OC extracted by water and MeOH in chamber diluted primary samples had smaller mass
756 absorption efficiency compared to those extracted from the fresh emissions. This can again be explained by
757 the formation of non-soluble dark BrC through the phenomenon termed as “darkening by volatilization” as
758 the low volatile fraction consists of stronger chromophores (Calderon-Arrieta et al., 2024).

759

760 **3.6. Effects of simulated atmospheric aging on properties of particulate organic matter**

761 **3.6.1 Effects of aging on volatility and solubility of OC**

762 The relative OC fractions of chamber diluted primary particles and aged particles seemed to be similar (Fig.
763 7), suggesting that when it comes to the relative distribution of the OC volatility in BB emissions, the effect
764 of dilution in the environmental chamber outweighed the impact of photo and dark oxidative aging we
765 achieved in our experiments. After oxidative aging there were, however, further increments of the PC, which
766 is the lowest volatility organic fraction. Oxidative aging is known to increase the oxidation state and decrease
767 the volatility of aged particles, making them more insoluble in organic solvents (Saleh et al., 2020).
768 Expectedly, we observed that both photochemical and dark oxidative aging decreased the fraction of MSOC
769 significantly (Figure S11b). On the other hand, the more oxygenated aged OC exhibited slightly higher
770 WSOC compared to POC in fresh emissions, and we observed a medium correlation ($R = 0.5$) between water
771 solubility and the atomic O:C ratio obtained from SP-AMS (Fig. S14b). The only exception to this were the
772 smouldering emissions of CP and BFS, which did not exhibit any significant change in water solubility after

773 dark or photochemical aging. This suggests that the chamber diluted POA emitted from smouldering burns
774 are more resistant to photochemical or dark oxidation, inadvertently hinting towards the highly viscous
775 tarball BrC. Tarballs are known to be resistant to chemical oxidation (Chakrabarty et al., 2023) and we also
776 observed the formation of more stable dark tarballs, especially in smouldering emissions, due to oxidative
777 aging in the chamber (Fig. S15).

778 **3.6.2 Effects of aging on organic matter density**

779 Oxidative aging, both in photochemical and high NO_x dark conditions, led to a marginal increment in particle
780 densities for BFS emissions. For BFS smouldering emission, the effective density after photochemical aging
781 increased to 1.30 ± 0.05 g cm⁻³ while no significant change was observed between primary and dark aged
782 particle densities. For flaming dominated combustion, the particle densities increased under both
783 photochemical (1.30 ± 0.09 g cm⁻³) and dark aging conditions (1.30 ± 0.07 g cm⁻³). For CP, the effective
784 density of high NO_x dark aged particulate emissions remained unaltered at 1.1 g cm⁻³, while photochemically
785 aged emissions exhibited higher density. For SW emissions, both photochemical aging and low NO_x dark
786 aging appeared to have a negligible impact on overall particle effective densities (Table S4). In contrast,
787 particle effective densities for flaming and smouldering combustion of SG emissions seemed to be slightly
788 lower after undergoing photochemical and dark aging (Table S4), although the effects of statistical errors
789 couldn't be discarded due to lack of repetitions of the experiments.

790

791 **3.6.3 Effects of aging on organic matter chemical composition**

792 Due to low OC loading on the filters collected from the chamber, we were only able to assign roughly 120-
793 600 unique elemental formulae of the most abundant chemical compounds from FT-ICR MS analyses of the
794 photo and dark aged samples of CP and BFS, while signals arising from other compounds were below the
795 detection threshold (S/N ≥ 6) (Fig. S16, S17). Therefore, we were unable to perform a one-to-one comparison
796 of the chemical compositions of the fresh emission with the chamber-aged samples. However, we could
797 conclude that CH and CHO were the most abundant compound classes in the chamber diluted primary and
798 aged samples for both CP and BFS. We obtained elemental ratios (O:C, H:C and N:C) of the chamber diluted

799 primary and aged samples from the SP-AMS measurement to characterize the variability in the bulk
800 composition of the OC. The resulting Van-Krevelen plots (Fig. S18) showed that the chamber diluted
801 primary emission had H:C values close to 2.0, while the O:C and N:C values ranged between 0.10-0.25 and
802 0.013-0.027, respectively. We observed an increment in the O:C ratios in the chamber aged emission while
803 the H:C ratio decreased, as in some previous studies (Lambe et al., 2011). On the other hand, after high NO_x
804 dark aging, the overall N:C ratio did not display a similar increase, probably due to the fast degradation of
805 the nitroaromatics formed inside the chamber at the time of filter collection at the end of the experiments.

806 3.6.4 Effects of aging on optical properties

807 Depending on the biomass type and the combustion conditions, photochemical aging either increased (fig.
808 S19b) or decreased (fig. s19a,c,d) the light absorption efficiency (MAE) at wavelengths below 350 nm, while
809 the MAE_{MSOC} at wavelength range 350-450 nm decreased for all cases. Overall, MAE_{MSOC_550} and k_{MSOC_550}
810 also decreased and in some cases remain unaltered (Fig. 9, Table S4) after photochemical aging, while w_{MSOC}
811 increased. The w_{WSOC} and w_{MSOC} varied between 3.3-3.7 and 4.3-4.5 respectively for fresh emission of CP,
812 while it increased to 4.0-5.1 and 4.0-6.4 after photochemical aging. The largest increase in w_{MSOC} was
813 observed for flaming combustion of SG, where w_{MSOC} increased from 3.9 to 6.3, while k_{MSOC_550} decreased an
814 order of magnitude. All the photochemically aged WSOC and MSOC samples in this study lied in the w-BrC
815 to vw-BrC region of the k-w space. This behaviour is in line with the effects of photobleaching and
816 photolysis during OH exposure in the presence of UV lights. Increasing photooxidation has been shown to
817 cause fragmentation reactions after functionalization (Kroll et al., 2015; Saleh et al., 2020). OH• has been
818 shown to cleave polyaromatic chains and give rise to short unsaturated chemical moieties that absorb light at
819 the UV range of the spectrum. On the other hand, photochemical aging of RWC emissions in oxidation flow
820 reactor either decreased (for samples from Ihalainen et al., 2019a) or increased (Mukherjee et al., 2024) the
821 k_{MSOC_550}, suggesting that chemical composition of the precursor organic molecules and the reaction pathways
822 are consequential to secondary BrC optical properties (Lambe et al., 2013; Sumlin et al., 2017; Hems et al.,
823 2020).

824 The effect of dark aging on the optical properties was more non-trivial in our study. We noted that for
825 flaming dominated emissions, the NO_x dominated dark aging led to higher k₅₅₀ values and decreasing w

values (fig. 9) in accordance with Saleh (2020). This suggests an increase in light absorption towards the visible wavelength range, which was also seen in the absorption spectra (fig. S19b,d). On the other hand, dark aging seemed to have minor effect on the smouldering emissions of BFS (Fig.9, Fig. S19a). One explanation for this observation could be the prevalence of tarballs in the smouldering dominated emissions of both peat and other biomass samples, which have been shown to be quite resistant to oxidative aging (Chakrabarty et al., 2023). Additionally, nitroaromatics are reactive, and certain fractions of them may have degraded at the end of the chamber experiments or during filter collection from the chamber, making it difficult to distinguish the effects on optical properties. Owing to very limited NO_x concentration in the chamber during the “low-Nox” dark aging, we observed very limited effects of aging on the savanna biomasses. In general, the MAE and k decreased after aging (Table S4), most likely aided by the ozonolysis pathways in the presence of externally injected O₃ into the chamber.

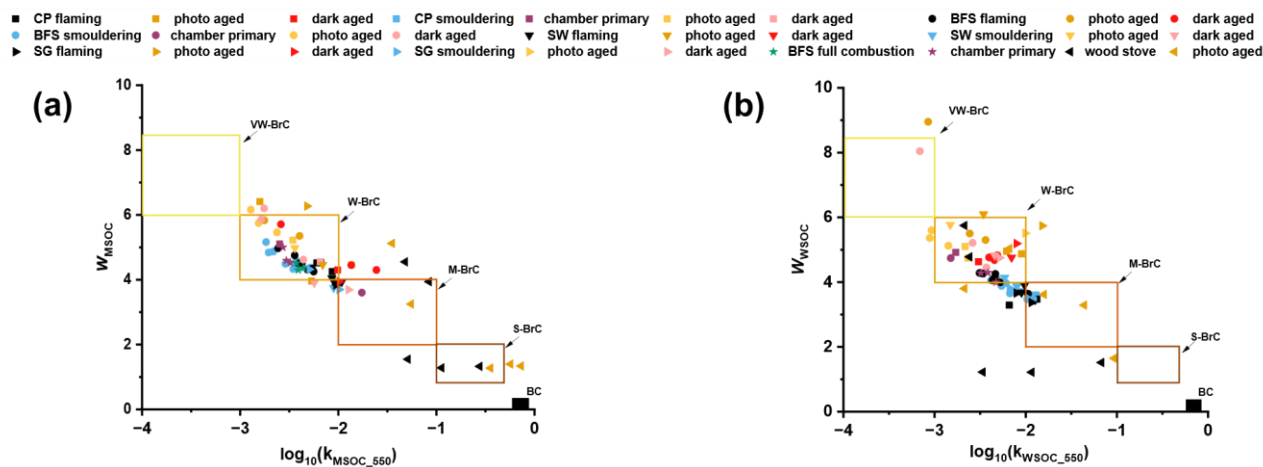


Figure 9: k-w space for all experimental data included in our study. Black, blue and green colours denote fresh emissions from flaming, smouldering and full combustion respectively. Purple dots denote primary emission in chamber without any aging (chamber primary). Dark yellow and red points denote photochemical and dark aging of flaming dominated emissions respectively, while light yellow and red dots represent respective photochemical and dark aging of smouldering dominated emissions.

WSOC light absorption in “k-w space” (fig. 9b) suggests that the water soluble BrC in fresh and aged emissions of RWC had lower k_{550} values and therefore less strongly absorbing chromophores than the corresponding MSOC samples. Certain dark aged samples, especially those for flaming BFS and SG emissions, also seemed to contain stronger chromophores in MSOC than in WSOC (Fig. 9). These observations suggest that less polar organics, which are soluble in MeOH but not in water, contribute to higher light absorption in these BB emissions. In addition, it should be noted that MSOC light absorption in the aged emissions might be underestimated, because fractions of the aged organics were non-soluble (Fig. S12b) and might have contained even stronger chromophores. In future, online measurements of OA absorption at different wavelengths is needed to confirm the observed effects of photochemical and dark aging.

855

856 **4. Conclusions:**

Our estimated OC and EC EFs suggest that OC generally dominates open BB emissions and specifically smouldering conditions emit higher OC than flaming combustion, in agreement with previously existing knowledge. However, we observed significant variation in EFs among experimental replicates, especially for smouldering burns of the same biomasses (fig. 2a,c). This clearly demonstrates how dynamic and variable open BB emissions are inherently, with many factors, including chemical compositions, fuel moisture content and combustion conditions, playing a crucial role in determining the emissions. Emission inventories for open BB from field and lab measurements are often presented as a single value averaged over experimental replicates (Andreae et al., 2019) for air quality or regional climate modelling purposes, and our findings highlight the importance of parameterizing EFs as a function of MCE (Fig. S6) or emitted OA/BC (or OC/EC) ratios for accurate modelling. The estimated high EF_{OC} for boreal and arctic peatland burns in our experiments also indicates that these vegetation types can become a major source for OA emissions in the region in response to increased forest fires, which can have drastic effects on local air quality and climate.

To include the effects of BrC in climate models, an important approach is to parameterize BrC k values as a function of the BC-to-OA (or alternatively EC/OC) ratio of the emissions. The estimated eBC/OA ratios for different fresh BB emissions in our study varied by orders of magnitude and ranged from 0.002 to

0.7. For different climate models, different BC/OA ratios such as 0.03-0.06 (Wang et al. 2018) and 0.08 (Brown et al., 2018) have been used, which don't account for the fuel or combustion phase dependent variations of BC/OA or EC/OC. The $k_{\text{MSOC}_{550}}$ for fresh BB emissions in our study varied between 0.002 and 0.011, and fell into the category of W-BrC, as classified by Saleh (2020). The k values of open BB emissions are very similar to those used in existing models, whereas BrC from high temperature combustion is currently underestimated. We also observed a different trend between $k_{\text{MSOC}_{550}}$ and EC/OC in this work (Fig. 5b) compared to previous literature, suggesting the need for an extensive fuel and combustion dependent emission inventory for better parameterization of BB emitted BrC k values for global climate models.

We observed a strong abundance of tarball morphologies of variable sizes for all open BB emissions, especially for smouldering burns. Characterization of the BB emissions in the environmental chamber revealed that the most volatile OC fractions, OC1 and OC2, decreased in the chamber compared to fresh emissions, due to particle-to-gas partitioning. This dilution-induced change of OC volatility distribution resulted in higher fractions of OC3, OC4 and PC (overlapping with SVOC, LVOC and ELVOC), potentially leading to more absorbing BrC chromophores (Calderon-Arrieta et al., 2024) as well as more defined tar balls in the chamber primary samples. We observed a slight decrease in MAE_{MSOC} for wavelengths higher than 365 nm. This contrasting finding is likely because our solvent extraction method cannot resolve the lowest volatility fraction of organics (with tar ball morphology) in the chamber primary samples.

We determined an increase in O:C ratio of the particles upon photochemical and dark oxidation, which resulted in an increase in ELVOC fractions (PC in particular), making it more insoluble in solvents. Dark aged CP and BFS smouldering emissions in particular had the lowest MeOH solubility, probably due to the observed high abundance of highly viscous, low volatility tarballs (Chakrabarty et al., 2023). For the dark-aged samples, MAE_{MSOC} increased for all wavelengths, suggesting the formation of stronger light-absorbing oxygenated compounds and/or nitroaromatics. For photochemically aged samples, $k_{\text{MSOC}_{550}}$ decreased, probably due to photobleaching and breaking down of BrC chromophores. After aging $k_{\text{MSOC}_{550}}$ values ranged from 0.0011 to 0.03 across all studied biomass types, which were significantly lower than the k_{550} values currently used in climate models. However, our estimated k_{550} values don't include the non-soluble and highly light absorbing s-BrC fractions. Consequently, in future it would be important to

899 combine online and offline optical measurements of open BB emitted OA to characterize also the non-
900 soluble organic fraction.

901 **Data availability:**

902 Essential data used in this study are available at FAIR-aligned data repository Zenodo via
903 <https://doi.org/10.5281/zenodo.15647186> with Creative Commons Attribution 4.0 International license
904 (Mukherjee et al., 2025). Detailed mass spectrometry data are available upon request from the corresponding
905 authors.

907 **Author contribution:**

908 VV, AnV, OS, AH, PYP, MI, KK and AB contributed in designing the study as well as supervising the whole
909 experimental campaign. KK and MR arranged for the arctic-boreal biomasses, whereas KJ, SJS, PGVZ and
910 VV collected, sorted and transported savanna biomasses to the study site in Finland. Measurements were
911 performed by AM, MaS, AH, MI, PYP, LMFB, DL, TuK, LV, AB, MuS and VV. Offline filter analyses,
912 including solvent extraction and thermal–optical analyses were performed by AM and VL, with supervisions
913 from NK. SEM images were captured by HK. FT- ICR MS analyses of filter samples and subsequent data
914 analyses and plotting were done by CPR, TiK, VHN and JJ. Other data analyses were performed by AM,
915 MaS, TuK, AH, LB and LV. The manuscript was prepared by AM with help from AH, OS, MR, KK, MaS,
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1494	Appendix:	
1495		
1496	APM	Aerosol Particle Mass Analyzer
1497	BB	Biomass Burning
1498	BC	Black Carbon
1499	BFS	Boreal forest Surface samples (from Finland)
1500	BrC	Brown Carbon
1501	CP	Commercial Peat samples (from Finland)
1502	DMF	Dimethylformamide
1503	DR	Dilution Ratio
1504	eBC	equivalent Black Carbon, approximated black carbon concentration by Aethalometer
1505	EC	Elemental Carbon
1506	EF	Emission Factor
1507	ELVOC	Extremely Low Volatility Organic Carbon
1508	FIA	Finnish boreal peatland (from Lakkasuo)
1509	FIB	Finnish boreal peatland (from Siikaneva)
1510	FTIR	Fourier-Transform InfraRed Spectrophotometer
1511	<i>K</i>	imaginary part of refractive inde
1512	MAE	Mass Absorption Efficiency
1513	MCE	Modified Combustion Efficiency
1514	MeOH	Methanol, an organic solvent
1515	MSOC	Methanol Soluble Organic Carbon
1516	NMVOC	Non-Methane Volatile Organic Carbon
1517	NOR	Arctic peatland samples (from Svalbard, Norway)
1518	OA	Organic aerosol
1519	OC	Organic Carbon, refers to only the carbon mass of OA
1520	PEAR	Photochemical Emission Aging flow-tube Reactor
1521	PM ₁	Particulate matter with aerodynamic diameter of 1 µm or less
1522	PM ₁₀	Particulate matter with aerodynamic diameter of 1 µm or less
1523	POA	Primary Organic Aerosol
1524	POC	Primary Organic Carbon

1525	PRD	Porous Tube Dilutor
1526	UV	Ultra-violet, spectral region consisting light radiation wavelength of 100-400 nm
1527	OHR _{ext}	external reactivity of hydroxyl radicals
1528	RUS	peatland samples (from Rogovaya, Russia)
1529	RWC	Residential Wood Combustion
1530	SG	Savanna Grass (grassy savanna samples from South Africa)
1531	SMPS	Scanning Mobility Particle Sizer
1532	SOA	Secondary Organic Aerosol
1533	SOC	Secondary Organic Carbon
1534	SW	Savanna Wood (woody savanna biomass from South Africa)
1535	TOC	Total Organic Carbon
1536	CPC	Condensation Particle Counter
1537	DMA	Differential Mobility Analyzer
1538	SEM	Scanning Electron Microscopy
1539	WSOC	Water Soluble Organic Carbon