



1 **Understanding divergent Brown Carbon Photobleaching Rates from**

2 **Molecular Perspective**

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17 **ABSTRACT**

18 The global radiative effect of brown carbon (BrC) remains highly uncertain. BrC's
19 photobleaching, which significantly alters its radiative effect, has been still poorly constrained.
20 This study investigates photobleaching rates of laboratory-synthesized secondary BrC (aq-BrC),
21 biomass burning-derived BrC (b-BrC), and ambient PM_{2.5}-derived BrC (p-BrC). Our results reveal
22 a source dependence in BrC photobleaching rates. The highest photobleaching rate constant (k_{BrC})
23 is observed for aq-BrC ($1.13 \pm 0.08 \text{ h}^{-1}$), followed by p-BrC ($0.12 \pm 0.02 \text{ h}^{-1}$) and b-BrC ($0.05 \pm$
24 0.01 h^{-1}), indicating the stable light-absorption capacity of b-BrC in the atmosphere. The OH
25 oxidation of imidazole-2-carboxaldehyde (2-IC) and methylglyoxal oligomers, nitrophenols
26 (including phenols), and lignin derivatives governs the photobleaching of aq-BrC, p-BrC, and b-
27 BrC, respectively. The high k_{BrC} of aq-BrC is attributed to the high reactivity of the chain structures
28 in 2-IC and methylglyoxal oligomers. In contrast, the highly conjugated structures of lignin
29 derivatives in b-BrC impart stability against OH oxidation, resulting in a low k_{BrC} . Our findings
30 reveal the significant differences in the photobleaching behavior of BrC originated from different
31 sources, underscoring the crucial need to account for source differences in assessments of BrC's
32 global radiative forcing effect.

33



34 1. Introduction

35 Brown carbon (BrC) is a significant contributor to atmospheric warming (Saleh, 2020; Chung et al., 2012;
 36 Brown et al., 2021) and Earth's radiative budget (Bond, 2001; Kirchstetter et al., 2004; Wang et al., 2018; Wang
 37 et al., 2022) due to its ultraviolet-visible (UV-Vis) light absorption. The BrC's direct radiative effect (DRE),
 38 accounting for 20%–40% of total DRE caused by light-absorbing carbonaceous aerosols (Heald et al., 2014; Jo
 39 et al., 2016; Saleh et al., 2015; Wang et al., 2018; Wang et al., 2014), remains highly uncertainty. BrC with global
 40 average DRE values varying from +0.03 to +0.57 W/m² is considered as the least understood warming agents in
 41 the atmosphere (Zhang et al., 2020; Zeng et al., 2020; Corr et al., 2012).

42 BrC comprises abundant chemically reactive species that react readily with atmospheric reactive gases and
 43 radicals (Aiona et al., 2017; Fleming et al., 2020; Hems et al., 2021; Liu et al., 2020), modifying its light-
 44 absorption properties. Both laboratory experiments (Borduas-Dedekind et al., 2019; Fleming et al., 2019; Hems
 45 and Abbatt, 2018; Hems et al., 2020; Schnitzler and Abbatt, 2018; Schnitzler et al., 2022) and field campaigns
 46 (Dasari et al., 2019; Qiu et al., 2024; Wang et al., 2014; Xu et al., 2024; Zeng et al., 2020) consistently found
 47 that BrC's photochemical aging process reduced its mass absorption coefficient (MAC), which is also known as
 48 photobleaching. In current models, the evaluation of BrC photobleaching on its radiative forcing mainly depends
 49 on the temporal evolution of BrC's light-absorption capacity (Schnitzler et al., 2022; Wang et al., 2018; Xu et
 50 al., 2024), i.e., the photobleaching rate, which is substantial variability across different laboratory experiments
 51 and field observations. Notably, BrC generated from aqueous-phase reactions often exhibits negligible
 52 absorbance in the UV-Vis range after less than 2 hours of radiation (Aiona et al., 2017). Field observations reveal
 53 that atmospheric BrC undergoes a MAC reduction exceeding 60% after 1 day of sunlight exposure (Müller et
 54 al., 2023; Qiu et al., 2024). In comparison, over the same 1 day of solar radiation, the reduction in BrC derived
 55 from biomass burning is much lower than atmospheric BrC (Fan et al., 2019; Fleming et al., 2020; Müller et al.,
 56 2023; Wong et al., 2019). Overall, accurately determining BrC's photobleaching rate is paramount for
 57 quantifying the impact of BrC photobleaching on its radiative forcing effect in models.

58 Furthermore, BrC's chemical composition fundamentally governs its photochemical reactivity (Hems et al.,
 59 2021; Hems and Abbatt, 2018; Dalton and Nizkorodov, 2021; Liu et al., 2025), thereby modulating its
 60 photobleaching rate. However, the relationship between changes in BrC's molecular composition and light
 61 absorption properties during the photobleaching is still poorly constrained. This knowledge gap leads to
 62 significant uncertainty in evaluating the evolution of BrC's light absorption capacity during atmospheric
 63 transportation. For instance, BrC emitted from biomass burning has been observed to retain strong absorption
 64 after several days of transport to the Arctic (Yue et al., 2022). In contrast, model simulations estimate BrC is
 65 nearly complete photobleaching over this timescale (Wang et al., 2018; Xu et al., 2024), which contradicts the
 66 observational evidence. Elucidating the linkage between BrC molecular composition and photobleaching rates
 67 would improve the accuracy of global-scale assessments of BrC's dynamic light absorption properties during
 68 long-range transport.

69 In this work, we performed photochemical aging experiments on three types of BrC, including synthesised
 70 by aqueous phase reaction (referred to "aq-BrC"), and extracted from ambient (referred to "p-BrC") and biomass



71 burning (referred to “b-BrC”) PM_{2.5} samples. We found that the photobleaching rates of BrC from different
 72 sources vary significantly. High-resolution mass spectrometry (HRMS) analysis was adopted to provide
 73 molecular-level understanding of this source-dependent BrC photobleaching rate. In addition, OH oxidation was
 74 determined as the dominate pathway of BrC photobleaching. These findings highlight the importance of
 75 considering differences in BrC sources when assessing its global radiative forcing effect.

76 2. Methodology

77 2.1 Preparation of BrC

78 Methylglyoxal (MG, 40 wt%, Merck) and ammonium sulfate (AS, 98%, Aladdin) solutions (1 mol/L) was
 79 mixed in darkness at room temperature for 2 h (pH 3.7–4.2) (Yang et al., 2024) to generate aq-BrC. Residual
 80 MG/AS were removed by solid-phase extraction (SPE) (Lin et al., 2010; Wang et al., 2017). The eluent was
 81 dried under gentle stream of ultrapure N₂ (>99.99%) and reconstituted in ultrapure water (18.2 MΩ·cm).

82 PM_{2.5} samples emitted from corn straw combustion in household stoves was collected on quartz fiber filters
 83 (25 × 20 cm, Whatman) using a high-volume sampler (TH-1000H, Wuhan Tianhong; 1.05 m³/min). Figure S1
 84 shows a schematic of sample collection process of biomass burning PM_{2.5} samples, and Table S1 summarizes
 85 the concentrations of organic carbon (OC), element carbon (EC), the concentration of total organic carbon (TOC)
 86 of each sample. This approach closely represents real-world biomass burning emissions (Chen et al., 2019; Shan
 87 et al., 2017). For each sample, background aerosols were collected for 1 hour both before to and after the
 88 sampling period.

89 Atmospheric PM_{2.5} samples were collected at Peking University Changping Campus (40°8′N, 116°6′E)
 90 from 15 December 2020 to 15 January 2021 using a high-volume sampler. The sampling period of each sample
 91 was 23 hours. Gaseous pollutants, PM_{2.5} mass concentrations, non-refractory submicron particles (NR-PM₁)
 92 chemical composition, and meteorology were monitored (Figure S2; details in Table S2).

93 For those PM_{2.5} samples, twenty 1.5-cm² filter sections were ultrasonically extracted in LC-MS grade
 94 methanol (Merck Inc.) for twice (20 min) to extract p-BrC and b-BrC. Extracts were dried gentle stream of
 95 ultrapure N₂ (>99.99%) and redissolved in ultrapure water.

96 2.2 Laboratory aqueous-phase photochemical aging setup

97 Photochemical aging experiments were conducted in a temperature-controlled (20 ± 0.1 °C) chamber,
 98 equipped with 20 UV bulbs (40 W; λ_{max} = 370 nm). The TOC of all BrC solutions were adjusted to 50 mgC/L.
 99 BrC solutions were irradiated in quartz bottles (10 mL) with added H₂O₂ (30 wt%, Aladdin) to generate OH
 100 radicals. Steady-state OH concentrations ([OH]_{ss}) were quantified via pseudo-first-order decay of benzoic acid
 101 (methods detailed in Text S1) (Hems and Abbatt, 2018). The average [OH]_{ss} was (3.17 ± 0.47) × 10⁻¹⁴ mol/L,
 102 slightly below typical cloud water maxima. Laboratory irradiation times were converted to equivalent solar
 103 durations using a spectral flux density ratio (scaling factor: 3.86) (Qiu et al., 2024). Samples were collected at 0,
 104 2, 4, 8, and 16 hours of equivalent solar radiation.



Furthermore, a series of control experiments were performed under H₂O₂ free and dark conditions. In both cases, the same BrC solutions (50 mgC/L TOC) were used for different types of BrC. The former consisted of 16 hours of radiation in the absence of H₂O₂. The dark experiments involved storing the solutions in complete darkness for an identical period (16 hours) to account for any non-photochemical changes.

These experiments were configured to delineate the contributions of different pathways to BrC photobleaching. The H₂O₂ added photochemical aging experiments elucidates the synergistic effect of OH oxidation, direct BrC photolysis, and other light-independent reactions occur in dark conditions. The H₂O₂ free experiments considered the sole contribution of the direct photolytic pathway. In addition, the dark control experiments served to investigate any potential influence of chemical reactions in dark conditions on the BrC's light-absorption capacity.

2.3 Light-absorption properties and molecular composition analysis of BrC

UV-Vis absorption spectra (250–700 nm) were recorded using a spectrophotometer (UV-1780, Shimadzu Inc.). MAC normalized absorption to TOC concentrations. In eq.(1), $A_{10}^{abs}(\lambda)$, b, and c denotes the measured base-10 absorbance, optical path length (1 cm), and measured TOC, respectively.

$$MAC(\lambda) = \frac{A_{10}^{abs}(\lambda) \times \ln(10)}{b \times c} \quad (1)$$

BrC's molecular composition was analyzed via HRMS (Orbitrap FusionTM TribridTM, Thermo Scientific) with electrospray ionization (ESI) in negative mode (ESI(-); m/z 70–700). Samples were directly injected into HRMS at a flow rate of 10 μ L/min. Triplicate analyses ensured reproducibility, with only peaks detected in all replicates retained for assignment. Molecular formulas ($C_xH_yO_zN_wS_n$; x = 1–90, y = 1–200, z = 0–20, w = 0–3, n = 0–1) were assigned with m/z error lower than ± 0.005 Da. The H/C, O/C, N/C, and S/C atom ratios for each assigned formula were constrained to 0.3–3.0, 0–3.0, 0–0.5, and 0–2.0, respectively (Wang et al., 2017). Signal-intensity-weighted average elemental compositions (eq.(2)), O/C atom ratios, and N/C atom ratios (eq.(3)) were calculated as:

$$Y = \frac{\sum_i x_i Y_i}{\sum_i x_i}, \text{ where } Y = C, H, O, N, \text{ and } S \quad (2)$$

$$Y/Z = \frac{\sum_i x_i Y_i}{\sum_i x_i Z_i}, \text{ where } Y/Z = O/C \text{ or } N/C \quad (3)$$

where x_i in eqs. (2) and (3) indicated the signal-intensity-weighted factor.

3. Results and Discussion

3.1 BrC Photobleaching rates

Given that BrC's light-absorption properties at shorter wavelengths are a better indicator of its photobleaching kinetics (Aiona et al., 2017), we focused our analysis on the variation in the MAC at 300 nm



(MAC₃₀₀). Figure 1(a) shows the decay of MAC₃₀₀ for different BrC types, plotted as the natural logarithm of the ratio ($\ln(\text{MAC}_{300,t}/\text{MAC}_{300,0})$) versus irradiation hours. Here, MAC_{300,t} and MAC_{300,0} denote the MAC₃₀₀ after t hours of irradiation and its initial value, respectively. The observed linear decay demonstrates that BrC undergoes photobleaching regardless of its source. The aq-BrC has been almost completely bleached in the initial 2 hours. The MAC₃₀₀ decreased by 77.2% after 16 hours of radiation. In comparison, for b-BrC, the MAC₃₀₀ only decreased by 56.1%. Even after extending to 32 hours of radiation, b-BrC still kept strong light-absorption capacity (see Figure S3). The differences in the variation in MAC₃₀₀ for different types of BrC demonstrate that aq-BrC undergoes rapid photobleaching in the presence of solar radiation, while b-BrC exhibits stable light-absorption capacity during photobleaching.

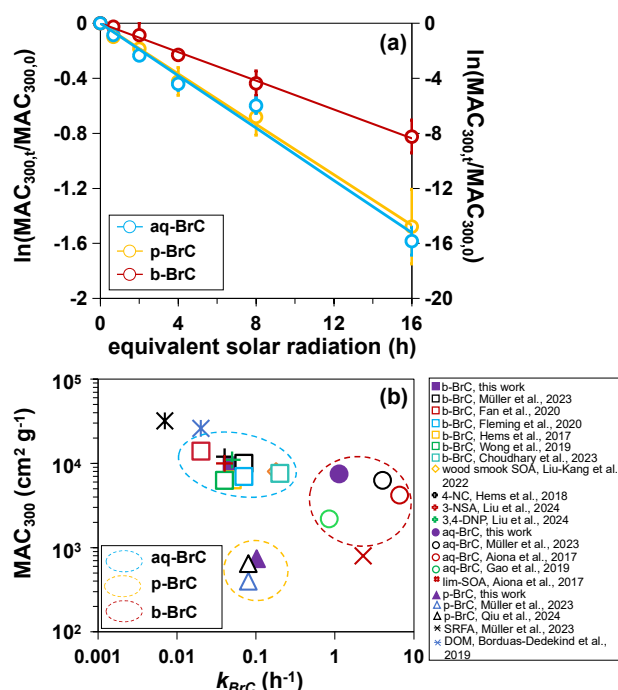


Figure 1 (a) Variations in the $\ln(\text{MAC}_{300,t}/\text{MAC}_{300,0})$ of aq-BrC (blue plots, right axis), p-BrC (yellow plots, left axis), and b-BrC (red plots, left axis) as a function of radiation hours. Blue, yellow, and red lines indicate fitted k_{BrC} of aq-BrC, p-BrC, and b-BrC. Error bars represent one standard deviation of datapoints derive from three (aq-BrC and b-BrC) or five (p-BrC) parallel experiments. (b) The relationship between MAC₃₀₀ and calculated k_{BrC} of different types of BrC in this work and other studies. 4-NC, 3-NSA, 3,4-DNP, SRFA, and DOM represents 4-nitrocateol, 3-mitrosalicylic acid, 3,4-dinitrophenol, Suwannee River fulvic acid standard, and dissolved organic matters, respectively. Blue, yellow, and red circles marked aq-BrC (including BrC generated via aldehyde-amine condensation reactions), p-BrC, and b-BrC (including representative compounds emitted from biomass burning). The exact MAC₃₀₀ values are obtained from figures in corresponding literatures, and k_{BrC} are calculated based on the MAC₃₀₀ and hours of radiation.



155 The liner decline in $\ln(\text{MAC}_{300,t}/\text{MAC}_{300,0})$ indicates that BrC photobleaching fitted the pseudo-first order
 156 kinetic, which was represented by the negative slope of $\ln(\text{MAC}_{300,t}/\text{MAC}_{300,0})$ vs. radiation hours:

$$157 \quad \text{MAC}_{300,t} = \text{MAC}_{300,0} \times \exp(-k_{\text{BrC}}t) \quad (4)$$

158 where k_{BrC} is the apparent photobleaching rate constant. The calculated k_{BrC} of aq-BrC, p-BrC, and b-BrC were
 159 $(1.13 \pm 0.08) \text{ hour}^{-1}$, $(0.12 \pm 0.02) \text{ hour}^{-1}$, and $(0.05 \pm 0.01) \text{ hour}^{-1}$, respectively. Correspondingly, the lifetime of
 160 BrC (τ_{BrC} , indicated by BrC's absorption decreased to 1/e of its initial value) (Schnitzler et al., 2022) were 1.05
 161 hours, 10.91 hours, and 19.16 hours, respectively. Longer τ_{BrC} of b-BrC indicates its absorption largely retains
 162 during atmospheric transportation process.

163 Figure 1(b) presents a comparison of MAC_{300} versus k_{BrC} for BrC from various sources, including data from
 164 this study and other studies (data and references in Table S3). Based on their sources, we classified aq-BrC
 165 (including BrC generated via aldehyde-amine condensation reactions), p-BrC, and b-BrC (including
 166 representative compounds emitted from biomass burning) in three different clusters. Our results are comparable
 167 to those of previous studies. The characteristic of aq-BrC is strong absorption, rapid photobleaching. This
 168 demonstrates that the light-absorption capacity of BrC generated by atmospheric aqueous phase reactions (De
 169 Haan et al., 2017) decrease rapidly in the presence of solar radiation. In stark contrast, b-BrC shows strong
 170 absorption but slow photobleaching, implying its stable strong absorption in the atmosphere. p-BrC exhibits
 171 weak absorption and slow photobleaching, which has also been observed in different urban areas, including
 172 Eastern China (Qiu et al., 2024), India (Dasari et al., 2019), Northern Europe (Müller et al., 2023), and the United
 173 States (Chen et al., 2021).

174 Having investigated the differences in photobleaching rates across different BrC types, we further
 175 elucidated the underlying mechanisms by assessing the contribution of each photobleaching pathway. Previous
 176 studies suggest that BrC photobleaching occurs via three different pathways, including direct photolysis, OH
 177 oxidation, and other light-independent reactions occur in dark conditions (Gao and Zhang, 2019; Liu-Kang et
 178 al., 2022; Wong et al., 2019). Choudhary et al. (2023) (Choudhary et al., 2023) have demonstrated that in the
 179 presence of H_2O_2 , the k_{BrC} can be expressed as a linear sum of the pseudo first-order effective photobleaching
 180 rate constant due to OH oxidation ($k_{\text{BrC},\text{OH}}$), BrC direct photolysis ($k_{\text{BrC},\text{pho}}$), and other light-independent reactions
 181 occur in dark conditions ($k_{\text{BrC},\text{ctrl}}$), see eq. (5):

$$182 \quad k_{\text{BrC}} = k_{\text{BrC},\text{OH}} + k_{\text{BrC},\text{pho}} + k_{\text{BrC},\text{ctrl}} \quad (5)$$

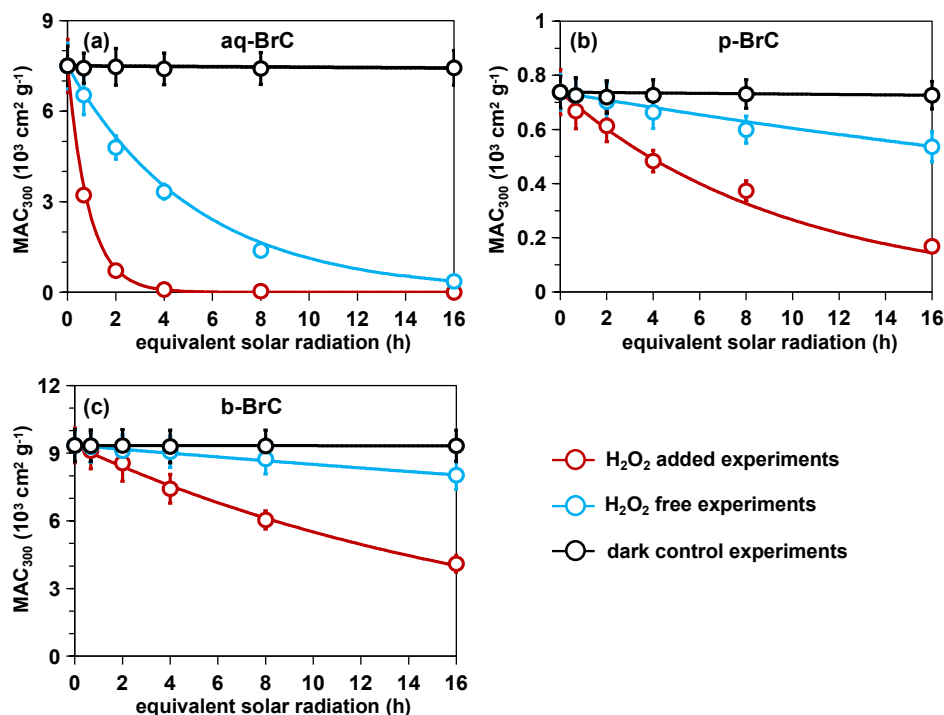
183 The $k_{\text{BrC},\text{pho}}$ and $k_{\text{BrC},\text{ctrl}}$ are derived from the pseudo-first-order decay of MAC_{300} in H_2O_2 free experiments
 184 and dark control experiments, respectively, using the calculation approach identical to eq. (4) and the
 185 experimental procedures detailed in Section 2.2. Therefore, although $k_{\text{BrC},\text{OH}}$ cannot be measured directly, it can
 186 be indirectly determined by calculating the difference between the k_{BrC} and the sum of $k_{\text{BrC},\text{pho}}$ (from H_2O_2 free
 187 experiments) and $k_{\text{BrC},\text{ctrl}}$ (from dark control experiments).

188 Figure 2 displays the variations in MAC_{300} in photochemical aging experiments (with H_2O_2 , red line,
 189 representing the synergistic effect of OH oxidation, direct BrC photolysis, and other light-independent reactions
 190 occur in dark conditions), H_2O_2 free experiments (blue line, representing the contribution of BrC direct
 191 photolysis), and dark control experiments (black line, representing the contribution of other light-independent



reactions occur in dark conditions). It is noted that for dark control experiments, the x-axis denotes the time the BrC solution is kept under dark conditions. MAC₃₀₀ variations were negligible in dark controls, rendering the calculated $k_{BrC,ctrl}$ was close to 0. For all BrC types, $k_{BrC,pho}$ was substantially lower than $k_{BrC,OH}$. Specifically, the $k_{BrC,pho}$ was $(0.19 \pm 0.03) \text{ h}^{-1}$, $(0.02 \pm 0.005) \text{ h}^{-1}$, and $(0.01 \pm 0.004) \text{ h}^{-1}$ for aq-BrC, p-BrC, and b-BrC, respectively. The calculated $k_{BrC,OH}$ was $(0.94 \pm 0.06) \text{ h}^{-1}$, $(0.08 \pm 0.02) \text{ h}^{-1}$, and $(0.04 \pm 0.01) \text{ h}^{-1}$ for aq-BrC, p-BrC, and b-BrC.

For all types of BrC, the overwhelming contribution of $k_{BrC,OH}$ (over 80% of k_{BrC}) unambiguously identifies OH oxidation as the dominant mechanism governing BrC photobleaching. Consequently, the observed differences in k_{BrC} among different types of BrC can be fundamentally attributed to the molecular-composition-dependent reactivity of BrC components toward OH radicals. Hence, understanding the underlying molecular composition that determine a BrC molecule's susceptibility to this OH oxidation.



202

Figure 2 The variations in MAC₃₀₀ in photochemical aging experiments (with H₂O₂, red line, representing the synergistic effect of OH oxidation, direct BrC photolysis, and other light-independent reactions occur in dark conditions), H₂O₂ free experiments (blue line, representing the variation in MAC₃₀₀ contributed by BrC direct photolysis), and dark control experiments (black line, representing the variation in MAC₃₀₀ contributed by other light-independent reactions occur in dark conditions) for (a) aq-BrC, (b) p-BrC, and (c) b-BrC, respectively. For dark control experiments (black line), the x-axis represents the time the BrC solution is kept in the absence of light.



210 3.2 Chemical investigation of different BrC photobleaching rates

211 To elucidating the differences in BrC photobleaching rates, we investigated BrC's molecular composition
 212 before and after photobleaching, which was represented by BrC's molecular composition after 16 hours of
 213 radiation.

214 Table 1 summarizes the signal-intensity-weighted average elemental compositions, molecular weights, O/C
 215 ratios, and N/C ratios of different types of BrC before and after 16 hours of radiation. The observed decrease in
 216 average molecular weights upon photobleaching across all BrC types, alongside an elevated O/C ratio, points to
 217 the production of more oxidized, lower-mass species. Additionally, we found the mass concentrations of low-
 218 molecular-weight organic acids (formic acid, acetic acid, and oxalic acid; see Figure S4) significantly increased
 219 with longer radiation hours, demonstrating carbon backbone fragmentation (Borduas-Dedekind et al., 2019).
 220 Figure S5 demonstrates that more products with lower C atom numbers are detected after photobleaching, further
 221 proves the carbon backbone fragmentation. Considering OH oxidation dominates BrC photobleaching, the
 222 molecular-level evolution aligns with OH-mediated degradation pathways (Hems et al., 2020; Qiu et al., 2024;
 223 Liu et al., 2025): (i) electrophilic addition to unsaturated bonds (like C=C bonds) and (ii) subsequent carbon
 224 backbone fragmentation.

225 In addition, the reduced N/C atom ratios (Table 1) further confirmed degradation of N-containing
 226 compounds, which are key UV-Vis absorbers contributing significantly to BrC absorption (Lin et al., 2017; Wang
 227 et al., 2019; Zeng et al., 2021; Li et al., 2025). The formation of compounds with higher oxidation states and the
 228 degradation of N-containing compounds reduce the conjugation degrees of BrC, diminishing $\pi \rightarrow \pi^*$ electronic
 229 transitions and weakening light-absorption capacity.

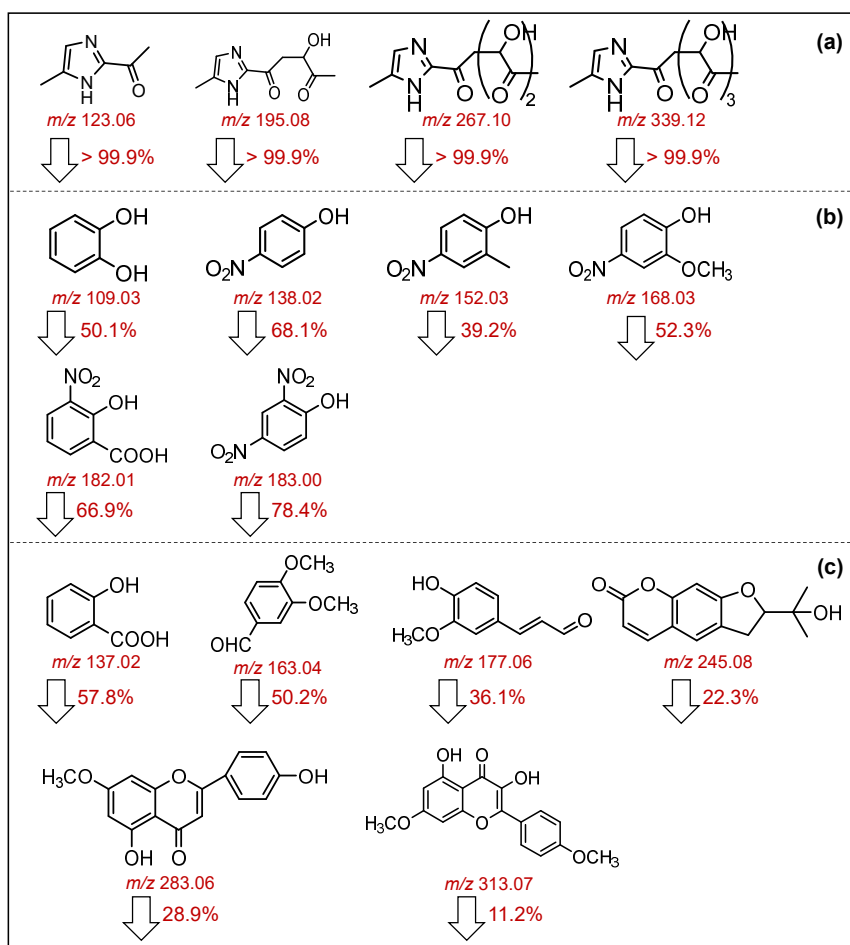
230 **Table 1** Variations in the signal-intensity-weighted average elemental compositions, molecular weights,
 231 O/C ratios, and N/C ratios of different types of BrC before and after photobleaching

aq-BrC	elemental composition	molecular weight	O/C	N/C
before	C _{8.94} H _{10.22} O _{4.64} N _{1.33}	210.36	0.52	0.15
after	C _{8.22} H _{10.17} O _{4.66} N _{1.07}	198.35	0.57	0.12
p-BrC	elemental composition	molecular weight	O/C	N/C
before	C _{7.88} H _{8.38} O _{2.47} N _{2.13} S _{0.65}	193.08	0.31	0.28
after	C _{7.21} H _{9.17} O _{2.52} N _{1.82} S _{0.68}	183.25	0.35	0.25
b-BrC	elemental composition	molecular weight	O/C	N/C
before	C _{15.92} H _{16.68} O _{6.05} N _{1.18} S _{0.31}	330.96	0.38	0.08
after	C _{15.42} H _{16.92} O _{6.11} N _{1.07} S _{0.41}	327.82	0.40	0.07

232 Figure S6 shows the differential HRMS spectra, the negative peaks represent species consumed after
 233 photobleaching. The photooxidation of MG and imidazole-2-carboxaldehyde (2-IC) oligomers (Aiona et al.,
 234 2017) with formula C_{3n}H_{4n}N₂O_{2n-3} (n = 2-5; including *m/z* 123.06, 195.08, 267.10, and 339.12), phenols and
 235 nitrophenols (Hems and Abbatt, 2018; Magalhães et al., 2017) (including *m/z* 109.03, 138.02, 152.03, 168.03,
 236 182.01, and 183.00), and lignin derivatives (Fleming et al., 2020) (including *m/z* 137.02, 163.04, 245.08, 283.06,
 237 and 313.07) were identified as the dominant reactions in the photobleaching of aq-BrC, p-BrC, and b-BrC,



238 respectively. Compounds identified as products after photobleaching for aq-BrC, p-BrC, and b-BrC are
 239 respectively summarized in Tables S4-S6.



240

241 **Figure 3** The chemical structures of (a) MG and 2-IC oligomers detected in aq-BrC; (b) phenols and nitrophenols
 242 detected in p-BrC, and (c) lignin derivatives detected in b-BrC. The detected m/z for these compounds are marked
 243 under their structures. The ratios of decline in signal intensities for these compounds after 16 hours of radiation
 244 are indicated next to the bottom arrows.

245 Figure 3 displays the chemical structures of these MG and 2-IC oligomers, phenols and nitrophenols, and
 246 lignin derivatives, as well as the declines in their signal intensities after 16 hours of radiation. Variations in
 247 HRMS signal intensities represent the changes in the mass concentrations, though specific mass concentrations
 248 cannot be derived. Figure 3(a) illustrates the predominant molecular species and the corresponding decline in



MG and 2-IC oligomers during the photobleaching of aq-BrC. Following photobleaching, these oligomers became undetectable (signal-to-noise ratio < 3), experiencing a reduction in signal intensity of >99.9% (Figure 3(a)). This observation is consistent with the nearly complete bleaching of aq-BrC. The high reactivity of the chain structures of MG and 2-IC oligomers with OH radicals (Aiona et al., 2017) explains the elevated k_{BrC} value observed for aq-BrC.

As shown in Figure 3(b), a greater number of electron-withdrawing substituents on the benzene ring correlates with a more significant decline of signal intensity after photobleaching. For instance, 2-methyl-4-nitrophenol (m/z 152.03), which bears an electron-donating methyl group in addition to the electron-withdrawing nitro substituent present in 4-nitrophenol (m/z 138.02), demonstrates a comparatively lower signal attenuation of 39.2% after photobleaching, whereas 4-nitrophenol exhibits a signal decrease of 68.1%. The electron-withdrawing substituents exert both inductive and resonance effects, effectively diminishing the electron density of the π -system of the aromatic ring. This electron-deficient state enhances the susceptibility of these species to nucleophilic attack by OH radicals, thereby facilitating their photooxidation and contributing to the observed photobleaching behavior. However, the reactivity of these species remains lower than that of MG and 2-IC oligomers, which explains lower k_{BrC} for p-BrC compares to aq-BrC.

In comparison, the signal intensities of the majority of lignin derivatives only decreased by below 50%, corresponding to the lowest k_{BrC} for b-BrC (Figure 3(c)). Specifically, nodakenetin and flavonoids (m/z 245.08, 283.06, and 313.07) exhibit highly stabilized molecular structures, wherein the aromatic rings conjugated with oxygen-containing heterocyclic moieties significantly reduce their chemical reactivity, resulting in low k_{BrC} for b-BrC. The chemical stability of lignin derivatives has been also proved by previous laboratory experiment (Wong et al., 2019).

4. Conclusion and Atmospheric Implication

This work demonstrates that BrC exhibits source-dependent photobleaching rates. Specifically, aq-BrC shows the highest k_{BrC} , followed by p-BrC and b-BrC, indicating that light-absorption capacity of b-BrC keep stable during photobleaching. In addition, considering the overwhelming contribution of $k_{BrC,OH}$ to k_{BrC} , the OH oxidation is determined as the predominate pathway of BrC photobleaching. The OH oxidation leads to the formation of compounds with higher oxidation state and lower molecular weights, which decreases the conjugation degree of BrC and therefore results in photobleaching. The oxidation of MG and 2-IC oligomers, phenols and nitrophenols, and lignin derivatives dominate the photobleaching of aq-BrC, p-BrC, and b-BrC, respectively.

Source-dependent k_{BrC} values necessitate explicit consideration of source variability when evaluating BrC's climate effect. Current models often assume constant k_{BrC} based on simplified representative compounds or field data (Hems and Abbatt, 2018; Schnitzler et al., 2022; Wang et al., 2018), yet substantial discrepancies exist. For instance, the reported k_{BrC} values differ by approximately an order of magnitude between field observations and laboratory simulations (Schnitzler et al., 2022; Wang et al., 2018). Spatiotemporal heterogeneity in BrC sources (Xiong et al., 2022) (fossil fuel/biomass combustion, secondary formation) drives global regional variations in k_{BrC} . To further reduce the uncertainties in assessing BrC's global DRE, it is imperative to systematically quantify



286 the impact of BrC's source and integrate source-dependent data into regional and global climate models.

287 In addition, the low k_{BrC} for b-BrC explains unexpectedly high BrC absorption in remote regions (e.g., the
 288 Arctic). Field data confirm biomass burning contributes 57% of Arctic BrC while < 10% for secondary BrC (Yue
 289 et al., 2022). Previous studies have confirmed that biomass burning aerosols contains substantial amounts of
 290 lignin derivatives (Wong et al., 2019), which exhibit strong light-absorbing properties and chemical stability in
 291 OH oxidation. Therefore, during the long-range transport (Zhang et al., 2025), b-BrC is expected to retain strong
 292 absorption, leading to enhancement of the aerosol DRE over large spatial scales. On a global scale, one major
 293 source of b-BrC is wildfire events (Wang et al., 2025; Shen et al., 2024; Bond et al., 2004). With the increasing
 294 frequency of wildfires (Yue et al., 2013; Hurteau et al., 2014; Cunningham et al., 2024; Jain et al., 2022), the
 295 emission of b-BrC is expected to rise correspondingly. In all, b-BrC would exert a significant influence on BrC's
 296 global radiative effect.

297 Associate Content

298 Code and Data Availability

299 The data used in this study is available upon request from the corresponding author (zhijunwu@pku.edu.cn).

300 Author Contributions

301 Y.Q. and Z.W. designed this work. Y.Q., T.Q., Y.G., and D.L. collected and analyzed the measurement data.
 302 Y.Q., T.Q., Y.L. and R.M. analyzed UHPLC-MS data. Z.W. and M.H. edited the manuscript. All authors have
 303 read and agreed to submit this manuscript.

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308 Competing Interests

309 The authors declare that they have no conflict of interest.

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