



1 Contrasting absorption and transport regimes of black and brown carbon
2 across the western, central, and eastern Himalayas

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4 Ju-Lian Shi^{1,2,8}, Chong-Shu Zhu^{1,6*}, Yao Qu^{1,5}, Xiao Wang^{1,2}, Nan Wang⁴, Lu-Yao
5 Wang^{1,2}, Wen-Ting Dai^{1,6}, Hong Huang^{3#}, Jun-Ji Cao^{1,7}

6
7 ¹*State Key Laboratory of Loess Science, Institute of Earth Environment, Chinese*
8 *Academy of Sciences, Xi'an 710061, China*

9 ²*Xi'an Institute for Innovative Earth Environment Research, Xi'an, 710061, China*

10 ³*School of Resources and Environment, Nanchang University, Nanchang 330031,*
11 *China*

12 ⁴*Xi'an Meteorological Bureau, Xi'an 710016, China*

13 ⁵*University of Chinese Academy of Sciences, Beijing 100049, China*

14 ⁶*National Field Observation and Research Station (Shaanxi) for Ecological and*
15 *Environment Changes and Integrated Management in the Guanzhong Plain Region,*
16 *Xi'an 710061, China*

17 ⁷*Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China*

18 ⁸*School of Environmental and Municipal Engineering, Xi'an University of Architecture*
19 *and Technology, Xi'an 710055, China*

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*Corresponding author. Postal address: Institute of Earth Environment, Chinese Academy of Sciences, No. 97 Yan xiang Road, Yan-ta Zone, Xi'an 710061, Shaanxi, China. E-mail: chongshu@ieecas.cn

#Corresponding author. Postal address: School of Resources and Environment, NanChang University, Nanchang 330031, Jiangxi, China. E-mail: honghuang@ncu.edu.cn



21 **Key Points**

- 22 • Mineral dust, pollution-modified dust, and biomass burning emissions
23 characterized the aerosols in the western, central, and eastern Himalayas,
24 respectively.
- 25 • Orographic forcing modulated the spatial distribution of light absorption by
26 carbonaceous aerosols across the Himalayas.
- 27 • Primary brown carbon (BrC_{pri}) contributed substantially to light absorption
28 coefficients and aerosol radiative forcing in the central Himalayas.

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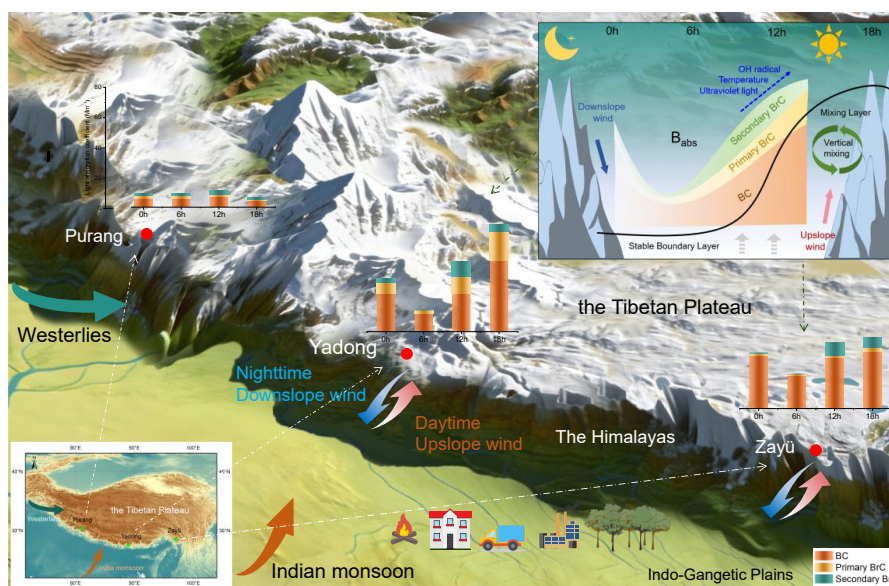
31 Abstract

32 Carbonaceous aerosols strongly influence atmospheric radiative forcing over the
33 Himalayas, yet their spatial variability and transport mechanisms remain poorly
34 constrained. Here we present high-time-resolution measurements of black carbon (BC),
35 primary brown carbon (BrC_{pri}), and secondary brown carbon (BrC_{sec}) at three high-
36 altitude sites across the western (Purang), central (Yadong), and eastern (Zayü)
37 Himalayas during the pre-monsoon period. Distinct spatial gradients in light absorption
38 and biomass burning (BB) tracers (levoglucosan and K^+) reveal pronounced regional
39 heterogeneity. Yadong exhibits the strongest absorption (B_{abs} : $53.1 \pm 18.5 \text{ Mm}^{-1}$ at
40 370 nm ; $14.2 \pm 5.5 \text{ Mm}^{-1}$ at 880 nm) and the highest BB influence, indicating a central
41 Himalayan hotspot of polluted aerosol accumulation. BC dominates light absorption
42 across all sites, with its contribution increasing eastward and reaching $\sim 95\%$ at 660 nm
43 in Zayü. In contrast, BrC contributions exhibit strong regional contrasts, with BrC_{pri}
44 dominating absorption in Yadong (8–28%) and BrC_{sec} enhanced in Purang (3–26%).
45 Trajectory analyses indicate that central Himalayan aerosol loading is strongly
46 modulated by orographic uplift and terrain-driven circulation. CALIPSO observations
47 further demonstrate a transition from mineral dust in the west, to pollution-modified
48 dust in the central Himalayas, and biomass-burning aerosols in the east. These results
49 improve our understanding of the sources, transport pathways, and atmospheric
50 transformations of BC, BrC_{pri} , and BrC_{sec} in the Himalayas.

51

52 **Keywords:** *the Himalayas, black carbon, primary brown carbon, secondary brown*
53 *carbon, radiative forcing*

54



Graph Abstract



59 1. Introduction

60 Carbonaceous aerosols influence Earth's radiative balance through the absorption
61 and scattering of solar radiation and by modifying cloud properties (Andreae and
62 Gelencser., 2006; Carslaw et al., 2010; Devaprasad et al., 2024; IPCC, 2013). Black
63 carbon (BC) is a key light-absorbing aerosol, mainly produced by the incomplete
64 combustion of fossil fuels, biomass, and biofuels (Fierce et al., 2020; T. C. Bond et al.,
65 2013; Zhang et al., 2025). In addition to BC, brown carbon (BrC), the light-absorbing
66 fraction of organic aerosol, absorbs radiation across the visible to near-ultraviolet
67 spectrum (approximately 300–700 nm) (Huang et al., 2025; Laskin et al., 2015; Laskin
68 et al., 2025; Rovira et al., 2026). According to its formation pathways, BrC can be
69 classified into primary BrC (BrC_{pri}) and secondary BrC (BrC_{sec}). BrC_{pri} is mainly
70 emitted from biomass burning (BB) and fossil fuel combustion, whereas BrC_{sec} is
71 formed through atmospheric chemical processes (Laskin et al., 2015; Nguyen et al.,
72 2012; Pang et al., 2019). Due to their complex chemical composition and structural
73 diversity, the constituents, optical properties, and radiative effects of BC and BrC
74 require further investigation.

75 Previous studies have demonstrated that BC contributes significantly to accelerated
76 glacial retreat across the Tibetan Plateau (TP). Moreover, the deposition of BC reduces
77 the albedo of snow and glacier surfaces, thereby influencing regional hydrological
78 cycles (Kopacz et al., 2011; Li et al., 2018; Qian et al., 2011; Qu et al., 2014;
79 Ramachandran et al., 2020; Zhu et al., 2021; Zhu et al., 2024). During the pre-monsoon
80 season, BC concentrations in the Himalayas typically reach their annual maxima,
81 largely due to enhanced BB emissions (Li et al., 2016; Ramachandran et al., 2020;
82 Zhang et al., 2020a). This seasonal enhancement is likely driven by favorable aerosol
83 transport conditions, as Westerlies and the Indian monsoon coincide with elevated
84 aerosol loading in the source regions during the pre-monsoon season (Xu et al., 2020).
85 In addition, atmospheric BrC over South Asia can be lifted to 5,000 m along the
86 southern slopes of the Himalayas (Bonasoni et al., 2010). The observations have further
87 shown that the light absorption by BrC increases with altitude (Zhang et al., 2017).



88 Previous studies have also shown that pollutant transport over the southern Himalayas
 89 is strongly influenced by mountain-valley circulation systems (Hindman and Upadhyay,
 90 2002; Yu et al., 2024; Zhang et al., 2020a).

91 Due to the harsh environment, the optical properties, transport pathways, and
 92 radiative forcing of BC, BrC_{pri}, and BrC_{sec} over the southern Himalayan slopes remain
 93 insufficiently characterized, particularly during the pre-monsoon season. This study
 94 presents high-time-resolution BC and BrC measurements conducted during three
 95 consecutive pre-monsoon seasons (March–May) from 2021 to 2023 at high-altitude
 96 sites along the southern slopes of the western, central, and eastern Himalayas.
 97 Concurrently, BB tracers were analyzed in total suspended particulate (TSP) samples
 98 collected at the same sites. Twenty-four-hour backward and forward trajectories were
 99 calculated to investigate aerosol transport pathways. The vertical distribution of
 100 aerosols during pollution events was characterized using Cloud-Aerosol Lidar and
 101 Infrared Pathfinder Satellite Observations (CALIPSO). Finally, the contributions of
 102 BrC_{pri} and BrC_{sec} to direct solar absorption relative to BC were estimated.

103

104 **2. Methods and Materials**

105 **2.1 Sampling and Measurement**

106 The field sampling campaign was conducted at three sites representing the western,
 107 central, and eastern Himalayas along the southern Himalayan slope: Purang (30.32°
 108 N, 81.17° E, 3,952 m a.s.l.), Yadong (27.52° N, 88.97° E, 3,400 m a.s.l.), and Zayü
 109 (28.50° N, 97.02° E, 1900 m a.s.l.) (Table S1). These sites were selected to investigate
 110 the spatial variability of carbonaceous aerosols across different sectors of the southern
 111 Himalayan slope. Sampling was carried out during the pre-monsoon season (March–
 112 May), with field campaigns conducted separately in 2021 at Purang, 2022 at Yadong,
 113 and 2023 at Zayü.

114 A seven-wavelength Aethalometer (model AE33, Magee Scientific Company,
 115 Berkeley, CA, USA) was used to measure the aerosol light absorption coefficient (B_{abs})
 116 at wavelengths of 370, 470, 520, 590, 660, 880, and 950 nm (Drinovec et al., 2015).



117 The spectral B_{abs} was obtained at these wavelengths, and the corresponding BC
 118 concentrations were calculated from the measured absorption. The aerosol B_{abs} at
 119 wavelength (λ) was determined from the corresponding BC concentration ($BC(\lambda)$) and
 120 the mass absorption cross-section ($MAC(\lambda)$), as described by equation (1) (Drinovec et
 121 al., 2015). In this equation, the values of MAC at 370, 470, 520, 590, 660, 880, and 950
 122 nm were 18.47, 14.54, 13.14, 11.58, 10.35, 7.77, 7.19 m^2g^{-1} , respectively.

$$123 \quad B_{\text{abs}}(\lambda) = BC(\lambda) \times MAC(\lambda) \quad (1)$$

124 TSP samples were collected on 47 mm Whatman quartz fiber filters (QM/A) using a
 125 custom-built high-volume sampler operating at a flow rate of 17 L/min. Sampling was
 126 performed continuously over a 7-day period. The concentrations of levoglucosan and
 127 potassium (K^+) in the TSP samples were quantified using established analytical
 128 methods (see Text S1 for details).

129

130 2.2 Data analysis methods

131 The B_{abs} of BC, BrC_{pri} , and BrC_{sec} can be derived according to the assumption that
 132 BrC absorption is zero at $\lambda = 880$ nm (Bahadur et al., 2012; Kirchstetter et al., 2012; Lu
 133 et al., 2015; Saleh et al., 2014). The $B_{\text{abs}}(\text{BC})$ at each wavelength was calculated by
 134 extrapolating the absorption at 880 nm using a absorption Ångström exponent (AAE_{BC})
 135 of 1.1 (Qu et al., 2022). BrC absorption at each wavelength was then estimated as the
 136 difference between the measured total aerosol absorption and the corresponding BC
 137 absorption. As BC is generated from combustion, a BC-tracer method was utilized to
 138 estimate the primary $B_{\text{abs}}(\lambda)$. The minimum R-squared method (MRS) was applied to
 139 derive the light absorption of BrC_{sec} across the full measurement wavelength range.
 140 This approach was adapted from the commonly employed BC-tracer technique
 141 originally developed for secondary organic carbon (SOC) quantification (Srivastava et
 142 al., 2018; Wang et al., 2019). Based on the negligible dust contribution, the MRS can
 143 separate $B_{\text{abs}}(\text{BrC}_{\text{sec}})$ versus the light absorption by BrC_{pri} ($B_{\text{abs}}(\text{BrC}_{\text{pri}})$) (Wang et al.,
 144 2019).

$$145 \quad B_{\text{abs}}(\text{BrC}_{\text{sec}})(\lambda) = B_{\text{abs}}(\lambda) - \left(\frac{B_{\text{abs}}(\lambda)}{BC} \right)_{\text{pri}} \times [BC] \quad (2)$$



[BC] represents the ambient BC concentration ($\mu\text{g m}^{-3}$). The parameter $\left(\frac{B_{\text{abs}}(\lambda)}{BC}\right)_{\text{pri}}$ denotes the ratio between the $B_{\text{abs}}(\lambda)$ and the BC mass concentration from primary emissions, with units of $\text{m}^2 \text{g}^{-1}$. $B_{\text{abs}}(\text{BrC}_{\text{pri}})(\lambda)$ is derived via the equation provided below:

$$B_{\text{abs}}(\text{BrC}_{\text{pri}})(\lambda) = B_{\text{abs}}(\text{BrC})(\lambda) - B_{\text{abs}}(\text{BrC}_{\text{sec}})(\lambda) \quad (3)$$

The total BrC absorption coefficient ($B_{\text{abs}}(\text{BrC})(\lambda)$) (Mm^{-1}) is derived by removing the BC contribution from the total absorption measured at $B_{\text{abs}}(\lambda)$. This calculation rests on the premise that BC constitutes the sole light-absorbing species at the 880 nm wavelength. A detailed description of the methodology is provided in Wang et al., (2019).

The direct solar absorption contributions of BrC_{pri} and BrC_{sec} relative to BC at the Purang, Yadong, and Zayü sampling sites were derived using a previously validated protocol described in previous studies (Huang et al., 2018; Kirillova et al., 2014). The light absorption by the species x (BrC_{pri} and BrC_{sec} , or BC) is computed in accordance with the Beer-Lambert's law, as shown in the equation below:

$$\frac{I_0 - I}{I_0}(\lambda) = 1 - e^{-b_{\text{ap},x,\lambda} \cdot h_{\text{ABL}}} \quad (4)$$

In this equation, the atmospheric boundary layer height (h_{ABL}) is assumed to be 1000 m. For a given wavelength (λ) and absorbing aerosol component (x), $b_{\text{ap},x,\lambda}$ denotes the component-specific aerosol absorption coefficient, representing either the BC absorption coefficient or the BrC absorption coefficient for BC and BrC, respectively. The relative contribution of BrC to solar radiation absorption compared with BC is then quantified using the following equation.

$$f = \frac{\int I_0(\lambda) \{1 - e^{-b_{\text{ap},\text{BrC}(\text{Primary/Secondary}),\lambda \cdot h_{\text{ABL}}}\} d\lambda}{\int I_0(\lambda) \{1 - e^{-b_{\text{ap},\text{BC},\lambda \cdot h_{\text{ABL}}}\} d\lambda} \quad (5)$$

where $I_0(\lambda)$ represents the clear sky Air Mass 1 Global Horizontal solar irradiance at wavelength λ (Levinson et al., 2010). The fraction f is derived via numerical integration



171 of the above expression across the entire sampling campaign, applied separately to three
172 distinct wavelength intervals spanning 300–400, 300–700, and 300–950 nm,
173 respectively (Huang et al., 2018).

174

175 **2.3 Application of software models**

176 The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was
177 used to assess air mass transport pathways to the sampling site, which was developed
178 by the National Oceanic and Atmospheric Administration's Air Resources Laboratory
179 (NOAA/ARL) (Draxler and Rolph, 2012). Using meteorological data from the Global
180 Data Assimilation System (GDAS, <ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/>)
181 with a spatial resolution of $1^{\circ} \times 1^{\circ}$, twenty-four-hour backward and forward trajectories
182 were produced on a six-hourly schedule, all terminating at 500 meters above ground
183 level. To identify dominant air mass transport pathways, trajectory clustering analysis
184 was performed by grouping trajectories according to their spatial similarity. The
185 Euclidean distance between corresponding trajectory coordinates was used as the
186 similarity metric.

187 The CALIPSO satellite, launched in April 2006, is a joint NASA–CNES mission
188 equipped with the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) as its
189 primary instrument. CALIOP is a dual-wavelength polarization-sensitive lidar
190 operating at 532 and 1064 nm (Hunt et al., 2009; Winker et al., 2007; Winker et al.,
191 2009). The sensor provides global vertical distribution information of aerosols and
192 clouds, making it particularly suitable for aerosol studies in regions with sparse ground-
193 based observations, such as the polar regions and the TP (Liu et al., 2008). This study
194 utilizes two CALIPSO Level 2 products to obtain vertical distribution information of
195 aerosols and clouds in the sites, namely the Aerosol Profile and the Vertical Feature
196 Mask, version V4.51, obtained from the NASA Langley Research Center Atmospheric
197 Science Data Center (<https://asdc.larc.nasa.gov/>). Detailed information is provided in
198 the supplementary material (Text S2).

199



200 **3. Results and discussion**

201 **3.1 Enhanced Carbonaceous Aerosol Light Absorption in the Central Himalayas**

202 Carbonaceous aerosol light absorption coefficients at 370 nm ($B_{\text{abs},370}$) and 880 nm
 203 ($B_{\text{abs},880}$) were simultaneously measured at three sites along the southern slopes of the
 204 Himalayas (Figure 1). At all sites, $B_{\text{abs},370}$ consistently exceeded $B_{\text{abs},880}$, suggesting a
 205 contribution from BrC in addition to BC. During the pre-monsoon season, the highest
 206 absorption coefficients were observed at Yadong, reaching $53.1 \pm 18.5 \text{ Mm}^{-1}$ at 370 nm
 207 and $14.2 \pm 5.5 \text{ Mm}^{-1}$ at 880 nm. The $B_{\text{abs},370}$ value was 2.8 times greater than $B_{\text{abs},880}$,
 208 indicating enhanced absorption by BrC in the near-ultraviolet wavelength range. In
 209 Zayü, the average absorption coefficients during the pre-monsoon were 31.5 ± 14.9
 210 Mm^{-1} at 370 nm and $13.1 \pm 6.1 \text{ Mm}^{-1}$ at 880 nm. In contrast, the lowest average
 211 absorption coefficients were observed in Purang, with values of $7.5 \pm 6.5 \text{ Mm}^{-1}$ at 370
 212 nm and $1.9 \pm 1.6 \text{ Mm}^{-1}$ at 880 nm. These results suggest that carbonaceous aerosols
 213 exhibited substantially stronger light absorption in the central Himalayas compared
 214 with those in the eastern and western Himalayan regions.

215 Previous studies have suggested that BB emissions from northern India can enhance
 216 shortwave aerosol absorption above clouds over the central Himalayas during spring
 217 (Kumar et al., 2011). The weekly averaged levoglucosan concentrations ranged from
 218 17.8 to 1688.9 ng m^{-3} at Purang, 280.8 to 2041.7 ng m^{-3} at Yadong, and 1.0 to 4.6 ng m^{-3}
 219 at Zayü. The seasonal mean levoglucosan concentrations were $518.5 \pm 567.6 \text{ ng m}^{-3}$
 220 at Purang, $812.0 \pm 457.8 \text{ ng m}^{-3}$ at Yadong, and $2.0 \pm 1.3 \text{ ng m}^{-3}$ at Zayü. Compared
 221 with previous studies, lower annual mean levoglucosan concentrations were reported
 222 for Ngari ($232.8 \pm 149.5 \text{ ng m}^{-3}$), Qinghai Lake ($65.5 \pm 39.6 \text{ ng m}^{-3}$), and Beiluhe (0.8
 223 $\pm 0.7 \text{ ng m}^{-3}$) (Zhu et al., 2022b), suggesting a stronger influence of BB emissions along
 224 the southern slopes of the Himalayas than in other regions of the TP.

225 The seasonal mean K^+ concentrations were $425.3 \pm 289.0 \text{ ng m}^{-3}$, $240.7 \pm 150.5 \text{ ng m}^{-3}$, and
 226 $151.4 \pm 89.1 \text{ ng m}^{-3}$ at Purang, Yadong, and Zayü, respectively. K^+ exhibited
 227 a wider concentration range in Purang (88.1 – 945.4 ng m^{-3}) than in Yadong (66.5 – 536.8
 228 ng m^{-3}) and Zayü (22.3 – 313.3 ng m^{-3}). Overall, the two BB tracers showed the lowest



concentrations in the eastern region, whereas substantially elevated concentrations were observed in the central and western regions. The enhanced optical absorption observed in the central Himalayas was likely associated, at least partly, with BB influences, as evidenced by elevated concentrations of levoglucosan and K^+ at sites along the southern Himalayan slopes.

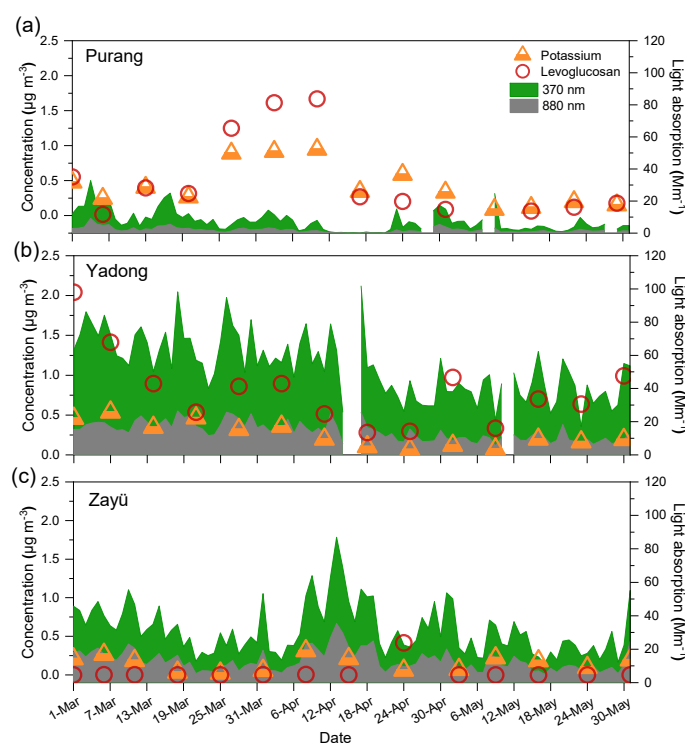


Figure 1. Variations of K^+ , levoglucosan, and light absorption coefficient (370 nm and 880 nm) in Purang (a), Yadong (b), and Zayü (c), respectively.

3.2 Primary Emissions Dominate Carbonaceous Aerosol Absorption

Figure 2 presents the temporal variations in $B_{abs}(BC)_{370}$, $B_{abs}(BrC_{pri})_{370}$, and $B_{abs}(BrC_{sec})_{370}$, together with temperature, relative humidity (RH), and wind speed at Purang, Yadong, and Zayü. Among the three absorbing components, BC consistently exhibited the highest absorption at 370 nm. Mean $B_{abs}(BC)_{370}$ values were 5.6 ± 4.0 Mm^{-1} at Purang, 47.7 ± 17.0 Mm^{-1} at Yadong, and 36.4 ± 16.9 Mm^{-1} at Zayü, with daily values ranging from 0.1–22.2, 14.0–95.4, and 12.3–94.3 Mm^{-1} , respectively.



245 The highest mean $B_{\text{abs}}(\text{BrC}_{\text{pri}})_{370}$ was also observed at Yadong ($21.9 \pm 6.6 \text{ Mm}^{-1}$),
 246 which was approximately 13.3 and 20.1 times higher than those at Purang (1.6 ± 1.1
 247 Mm^{-1}) and Zayü ($1.1 \pm 0.7 \text{ Mm}^{-1}$), respectively. Daily $B_{\text{abs}}(\text{BrC}_{\text{sec}})_{370}$ ranged from 0.1
 248 to 27.2 Mm^{-1} at Purang, 0.1 to 32.8 Mm^{-1} at Yadong, and 0.4 to 58.0 Mm^{-1} at Zayü.
 249 Although mean $B_{\text{abs}}(\text{BrC}_{\text{sec}})_{370}$ in Yadong ($10.0 \pm 7.1 \text{ Mm}^{-1}$) was lower than $B_{\text{abs}}(\text{BC})_{370}$
 250 and $B_{\text{abs}}(\text{BrC}_{\text{pri}})_{370}$, it remained the highest among the three sites. Mean $B_{\text{abs}}(\text{BrC}_{\text{sec}})_{370}$
 251 values were $5.7 \pm 9.4 \text{ Mm}^{-1}$ and $2.8 \pm 3.6 \text{ Mm}^{-1}$ at Zayü and Purang, respectively, both
 252 exceeding the corresponding $B_{\text{abs}}(\text{BrC}_{\text{pri}})_{370}$ values, suggesting a stronger contribution
 253 of BrC_{sec} to light absorption at these sites.
 254 Overall, $B_{\text{abs}}(\text{BC})_{370}$, $B_{\text{abs}}(\text{BrC}_{\text{pri}})_{370}$, and $B_{\text{abs}}(\text{BrC}_{\text{sec}})_{370}$ all reached their highest
 255 absolute levels in the central Himalayas, indicating a stronger influence of primary
 256 emissions in this region. In contrast, BrC_{sec} contributed more to light absorption than
 257 BrC_{pri} in the eastern and western Himalayas.

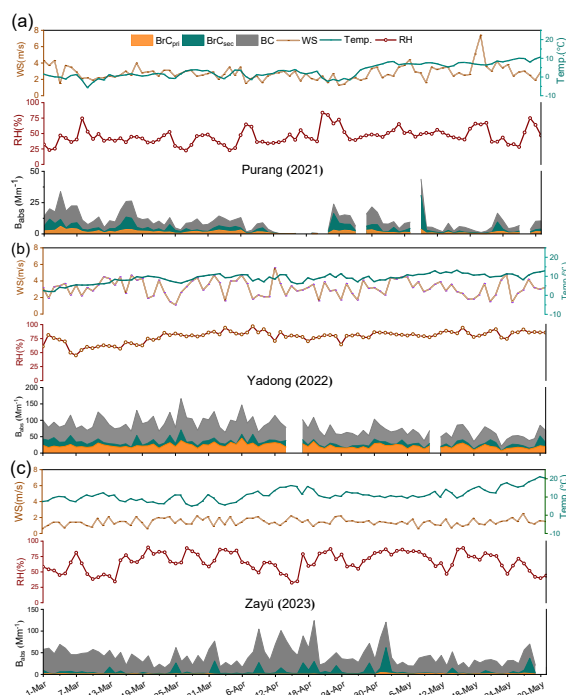


Figure 2. Light absorption of the BC, BrC_{pri} , and BrC_{sec} at 370 nm and associated meteorological parameters (wind speed, relative humidity, and temperature) at Purang (a), Yadong (b), and Zayü (c), respectively.



3.3 Spatial Variations in BC, BrC_{pri}, and BrC_{sec} Contributions

Figure 3 (I) presents the mean $B_{\text{abs}}(\text{BC})$, $B_{\text{abs}}(\text{BrC}_{\text{pri}})$, and $B_{\text{abs}}(\text{BrC}_{\text{sec}})$, together with their contributions to total light absorption across wavelengths from 370 to 660 nm. $B_{\text{abs}}(\text{BC})$, $B_{\text{abs}}(\text{BrC}_{\text{pri}})$, and $B_{\text{abs}}(\text{BrC}_{\text{sec}})$ all decreased with increasing wavelength. The highest $B_{\text{abs},370}$ was observed at Yadong ($79.6 \pm 25.5 \text{ Mm}^{-1}$), approximately 1.8 and 8 times higher than those at Zayü and Purang, respectively.

BC was the dominant light-absorbing component across the measured wavelength range at all three sites, consistent with previous observations (Ramachandran et al., 2020). Its contribution increased from the western to the eastern Himalayas. At 370 nm, BC accounted for 56%, 60%, and 84% of total absorption in Purang, Yadong, and Zayü, respectively. The highest BC contribution was observed in Zayü at 660 nm (95%).

The contribution of BrC_{pri} to total absorption was highest at Yadong and exceeded that of BrC_{sec} throughout the measured wavelength range. At Yadong, BrC_{pri} accounted for 28%, 18%, 12%, 10%, and 8% of total absorption at 370, 470, 520, 590, and 660 nm, respectively. In contrast, the lowest BrC_{pri} contribution was observed at Zayü (4%).

The contribution of BrC_{sec} to total absorption decreased from the western to the eastern Himalayas. The highest BrC_{sec} contributions were observed at Purang, reaching 26%, 13%, 9%, 6%, and 3% at 370 nm, 470 nm, 520 nm, 590 nm, and 660 nm, respectively. As shown in Figure 3 (II), the mean daily $B_{\text{abs}}(\text{BrC}_{\text{sec}})/B_{\text{abs}}(\text{BrC})$ ratios at 370 nm were 0.56, 0.29, and 0.50 for Purang, Yadong, and Zayü, respectively. These results indicate that BrC_{sec} contributed a larger fraction of total BrC absorption in the western and eastern Himalayas, whereas BrC_{pri} made a relatively greater contribution in the central Himalayas.

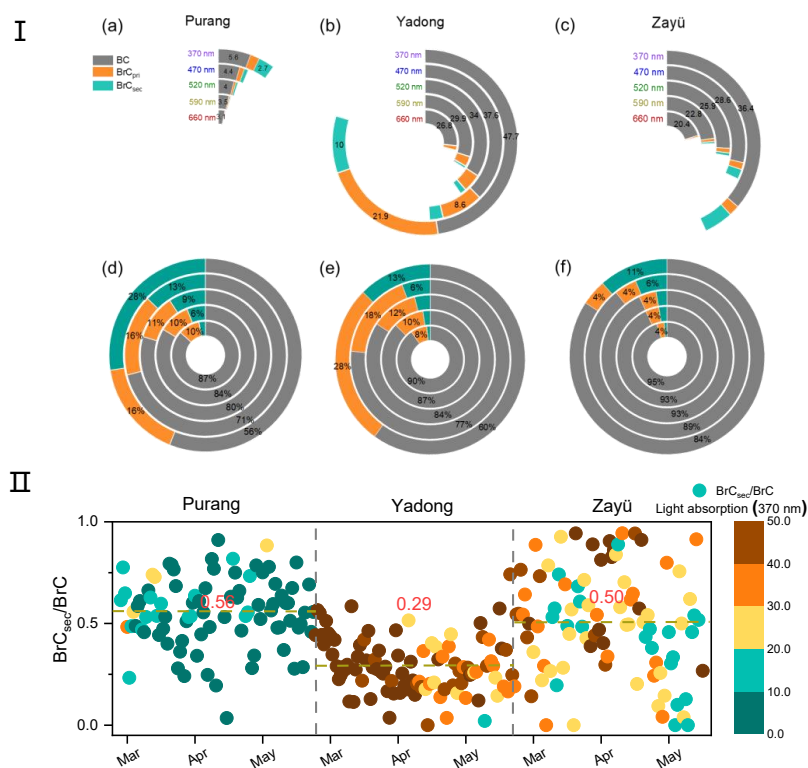


Figure 3. Relative contributions of BC, BrC_{pri}, and BrC_{sec} to B_{abs} (a-c), and wavelength-dependent absolute light absorption of BC, BrC_{pri}, and BrC_{sec} for Purang, Yadong, and Zayü (d-f), respectively (I). Ratios of BrC_{sec}/BrC at 370 nm for Purang, Yadong, and Zayü, respectively; the color of the points represents total light absorption at 370 nm (II).

3.4 Spectral Dependence of Aerosol Absorption and Implications for BrC Sources

The absorption Ångström exponent (AAE) is widely used to characterize the wavelength dependence of aerosol light absorption and to infer the relative contributions of different light-absorbing aerosol components. As shown in Table 1, the mean AAE values (370–950 nm) were 1.77, 1.71, and 1.22 at Purang, Yadong, and Zayü, respectively, exceeding those reported for Lhasa (1.04) and Lulang (1.18) during the postmonsoon season (Zhu et al., 2017). The relatively higher AAE values at Purang and Yadong suggest a greater influence of BB-related BrC, whereas the lower AAE at Zayü is more consistent with absorption dominated by BC from fossil-fuel combustion (Kirchstetter et al., 2004).

At all three sites, AAE values for BrC_{sec} exceeded those for BrC_{pri}. This likely reflects



the stronger wavelength dependence of absorption by BrC_{sec} chromophores, which preferentially absorb radiation at shorter wavelengths. The elevated AAE values of BrC (4.3) and BrC_{pri} (4.0) at Yadong may be associated with the enhanced contribution of BrC_{pri} to total aerosol absorption. The AAE values of BrC_{sec} followed the order Yadong (5.3) > Purang (5.2) > Zayü (4.5). These results further suggest that BrC_{sec} also represents a non-negligible contributor to carbonaceous aerosol light absorption in the Himalayas.

311

3.5 Orographic Control of Aerosol Transport across the Himalayas

Figure 4 presents 24-h backward and forward air-mass trajectories at Purang, Yadong, and Zayü during the pre-monsoon period. At Purang and Zayü, >94.6% of backward trajectories originated from low-altitude regions, and >98.9% of forward trajectories entered the TP, indicating efficient cross-Himalayan transport through canyon topography. In contrast, Yadong showed mixed transport pathways, with the north–south valley dominating (61.9–85.9%), while plateau and transverse valley contributions were smaller. Most forward trajectories (69.6–98.9%) entered the plateau, while a fraction recirculated within adjacent valleys (see Table S2 for details). These results indicate that aerosol transport in the central Himalayas is strongly modulated by complex terrain and valley circulations, which enhance cross-Himalayan exchange (Yu et al., 2024; Zhang et al., 2020a).

$B_{\text{abs}}(\text{BC})$, $B_{\text{abs}}(\text{BrC}_{\text{pri}})$, and $B_{\text{abs}}(\text{BrC}_{\text{sec}})$ exhibited pronounced diurnal cycles with two daytime peaks and a nighttime minimum (Figure 4 I). Morning peaks likely reflect nocturnal accumulation in elevated valley layers followed by daytime upslope transport (Sellegrì et al., 2010). At Yadong, $B_{\text{abs}}(\text{BC})$ and $B_{\text{abs}}(\text{BrC}_{\text{pri}})$ peaked at 18 h, consistent with primary emissions. $B_{\text{abs}}(\text{BrC}_{\text{sec}})$ peaked around noon at all sites, indicating enhanced daytime secondary formation. In Zayü, BrC_{sec} contributed 74.5–83.9% of BrC absorption during daytime, suggesting a strong influence of secondary formation and regional transport. Satellite observations further show widespread pre-monsoon aerosol loading over South Asia, with westerly flow and local valley circulations jointly enhancing aerosol accumulation along the southern Himalayas (Figure S1, Text S3).

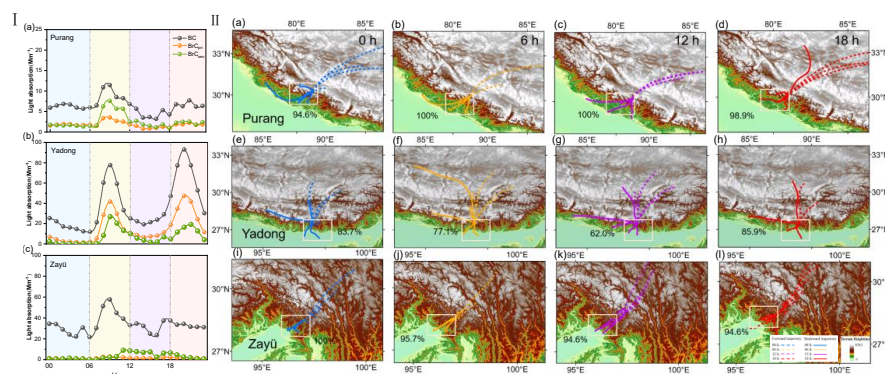


Figure 4. Diurnal patterns of 6-h integrated BC, BrC_{pri}, and BrC_{sec} at 370 nm (I (a-c)). 24-h air-mass trajectory cluster analysis for Purang (a), Yadong (b), and Zayü (c), respectively (II (a-l)).

Fire counts (<https://firms.modaps.eosdis.nasa.gov/>) indicate low local emissions over the TP during the pre-monsoon period (Figure 5 a-c), while active combustion sources are mainly distributed in South and Southeast Asia. The PSCF was calculated using $B_{abs}(BC)_{370}$, $B_{abs}(BrC_{pri})_{370}$, and 72-h back trajectories at 00, 06, 12, and 18 local standard time (Text S4). Cluster analysis of backward trajectories identified dominant transport pathways for Purang (C1–C4), Yadong (C1–C5), and Zayü (C1–C4) (Table S3).

At Purang, air masses from India (C1, C2) dominated the transport pathways (48 and 38%), with C2 showing the highest $B_{abs}(BC)_{370}$ (7.6 Mm^{-1}) and $B_{abs}(BrC_{pri})_{370}$ (2.2 Mm^{-1}), indicating polluted air mass influence. At Yadong, the dominant cluster (C1, 65%) also originated from India, with $B_{abs}(BC)_{370}$ and $B_{abs}(BrC_{pri})_{370}$ reaching 47.4 and 21.6 Mm^{-1} , respectively. Air masses from the southwestern TP (C2, 16%) were also observed. At Zayü, air masses were mainly transported along the Chayu River valley from Myanmar, Bangladesh, and India. PSCF analysis showed spatial patterns consistent with regional fire activity, highlighting the potential influence of BB and fossil-fuel combustion sources in South and Southeast Asia, while suggesting a weaker contribution from local emissions. Overall, aerosol distributions over the southern Himalayas appear to be shaped by the combined effects of regional transport and topography-driven circulation. (Zhang et al., 2020a).

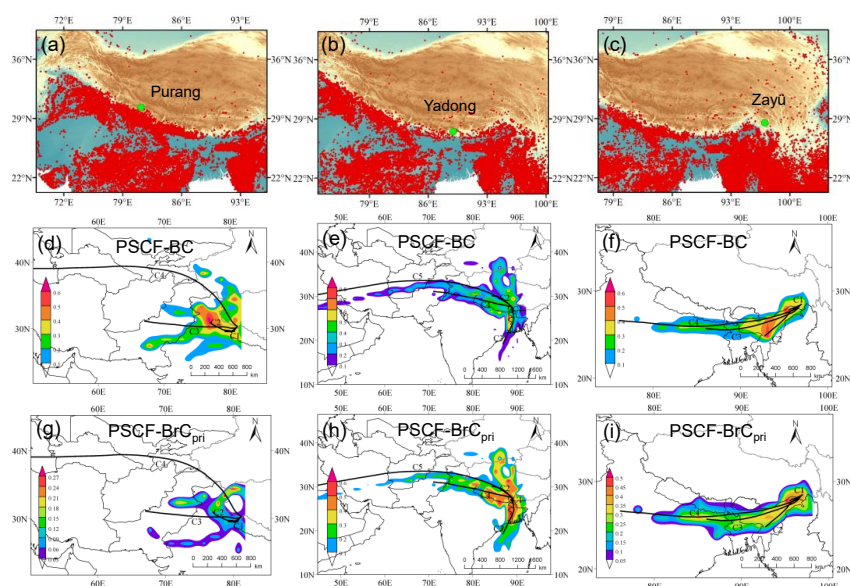


Figure 5. Seasonal distributions of fire hot spots from the Terra and Aqua satellites during the sampling period (a-c). The 72-h backward air-mass trajectory clusters and potential source regions of $B_{abs}(BC)$ and $B_{abs}(BrC_{pri})$ in Purang, Yadong, and Zayü, respectively (d-g).

3.6 Contrasting Aerosol Vertical Structures across the Southern Himalayas

Figure 6 presents CALIPSO profiles during representative aerosol pollution episodes at the three sites, which corresponded to consistently elevated B_{abs} (Table S4). Marked differences were observed in vertical aerosol profiles across the western, central, and eastern Himalayas during the pre-monsoon season. In the western Himalayas, the aerosol layer followed the terrain with a thickness of 2–3 km. Extinction (2 km^{-1}) and backscatter ($4 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$) showed a slight decrease, while volume depolarization ratio (VDR) and color ratio (CR) decreased from 0.4 to 0.1 and from 1.0 to 0.5, respectively, suggesting the presence of mixed dust and polluted aerosol layers associated with anthropogenic influence and long-range transport. This is consistent with enhanced BrC_{sec} absorption at Purang.

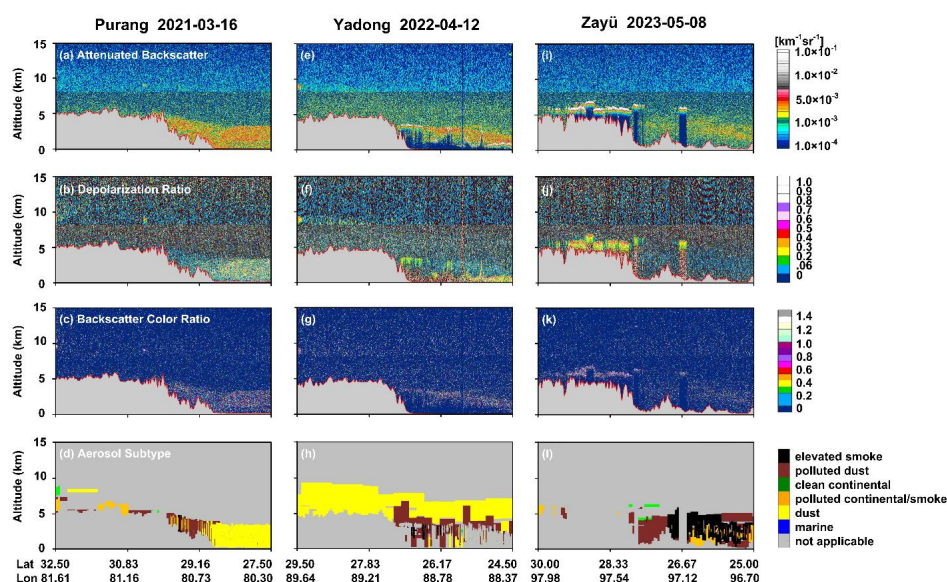
In the central Himalayas, a ~4 km thick dust layer persisted above 5 km altitude, with extinction ($\sim 0.04 \text{ km}^{-1}$) and backscatter ($\sim 7 \times 10^{-4} \text{ km}^{-1} \text{ sr}^{-1}$) decreasing with height. In the lower troposphere (0–4 km) over northern India and Bhutan, higher extinction (1.6 km^{-1}) and backscatter ($3 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$) were observed, together with low VDR (~ 0.1)



377 and moderate CR (~ 0.5), indicating polluted dust mixed with smoke. This likely
 378 contributed to the highest B_{abs} observed at Yadong.

379 In the eastern Himalayas, extinction (0.075 km^{-1}) and backscatter ($1 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$)
 380 were lower along air-mass pathways originating from Myanmar and northern India. The
 381 combination of elevated CR (~ 0.5) and low VDR (~ 0.1) suggests diluted polluted dust,
 382 while elevated BC absorption indicates long-range transport of BB aerosols influencing
 383 Zayü.

384 Overall, the southern Himalayas are influenced by multiple aerosol transport regimes,
 385 including dust-dominated, pollution-modified dust, and BB-influenced aerosols in the
 386 western, central, and eastern regions, respectively. Compared with Arctic and Antarctic
 387 regions, which are generally characterized by relatively pristine marine and mineral
 388 dust-dominated aerosol (Yang et al., 2021), the southern Himalayas exhibit more
 389 complex aerosol mixtures influenced by mineral dust, BB emissions, and anthropogenic
 390 pollution.



391
 392 Figure 6. CALIPSO overpass aerosol vertical profile, including attenuated backscatter (a,e,i),
 393 depolarization ratio (b,f,j), backscatter color ratio (c,g,k), and aerosol subtype (d,h,l) over the
 394 Himalayan on representative aerosol pollution episodes: 16 March 2021 (Purang), 12 April 2022
 395 (Yadong), and 8 May 2023 (Zayü), respectively.

396



3.7 Spatial Variability in the Radiative Forcing of BrC

Radiative forcing contributions of BrC_{pri} and BrC_{sec} relative to BC were evaluated at three sites during the pre-monsoon period (Table 1). In the UV range (300–400 nm), relative radiative forcing contributions of BrC_{pri}/BC were 34.3% (Purang), 54.1% (Yadong), and 5.3% (Zayü), respectively. These values were higher than those reported for the northeastern TP (Qinghai Lake, 16.7%), central TP (Beiluhe, 2.4%), and southwestern TP (Ngari, 13.2%) (Zhu et al., 2021).

The highest radiative forcing contributions of BrC_{sec}/BC were observed at Purang (55.7%), followed by Yadong (25.7%) and Zayü (15.5%), indicating an important contribution of BrC_{sec} to aerosol radiative absorption, particularly in the western Himalayas. Over the full solar spectrum (300–950 nm), BrC_{pri}/BC contributions decreased to 19.0%, 21.3%, and 4.3% at Purang, Yadong, and Zayü, respectively. Nevertheless, the relative radiative forcings of BrC_{pri}/BC (300–950 nm) exceeded those of BrC_{sec}/BC in Purang (16.2%) and Yadong (7.9%). Overall, these results suggest a stronger radiative influence of BrC_{sec} in the western Himalayas, whereas BrC_{pri} contributes relatively more to aerosol absorption in the central Himalayas.

Table 1. Aerosol light absorption coefficients and Ångström absorption exponents (AAE) of BC, BrC_{pri}, and BrC_{sec}, and the relative direct solar absorption of BrC_{pri} and BrC_{sec} compared with BC at three sites.

	Parameters	Purang	Yadong	Zayü
Seasonal Light absorption coefficient (Mm ⁻¹)	B _{abs} (BC) ₃₇₀	5.6 ± 4.0	47.7 ± 16.9	36.4 ± 17.0
	B _{abs} (BrC) ₃₇₀	4.4 ± 4.4	31.9 ± 11.0	6.8 ± 9.6
	B _{abs} (BrC _{pri}) ₃₇₀	1.6 ± 1.1	21.9 ± 6.6	1.1 ± 0.7
	B _{abs} (BrC _{sec}) ₃₇₀	2.8 ± 3.6	10.0 ± 7.1	5.7 ± 9.4
Absorption Ångström Exponent	AAE _{370/950}	1.77	1.71	1.22
	AAE _{BrC} (330/660)	3.96	4.40	3.18
	AAE _{BrC_{pri}} (330/660)	2.70	4.07	1.33
	AAE _{BrC_{sec}} (330/660)	5.22	5.30	4.50
BrC _{pri} /BC(%)	f _{300-400 nm}	34.3 ± 5.9%	54.1 ± 12.1%	5.3 ± 2.6%



	$f_{300-700\text{nm}}$	$21.6 \pm 3.3\%$	$25.6 \pm 4.3\%$	$4.4 \pm 1.2\%$
	$f_{300-950\text{nm}}$	$19.0 \pm 2.9\%$	$21.3 \pm 3.5\%$	$4.3 \pm 0.9\%$
$\text{BrC}_{\text{sec}}/\text{BC}(\%)$	$f_{300-400\text{ nm}}$	$55.7 \pm 12.7\%$	$25.7 \pm 5.9\%$	$15.5 \pm 5.8\%$
	$f_{300-700\text{nm}}$	$20.0 \pm 5.0\%$	$9.7 \pm 1.9\%$	$6.2 \pm 2.6\%$
	$f_{300-950\text{nm}}$	$16.2 \pm 4.1\%$	$7.9 \pm 1.5\%$	$5.1 \pm 2.1\%$

417

418 4. Conclusion

419 Light absorption by carbonaceous aerosols, including BC, BrC_{pri} , and BrC_{sec} ,
 420 together with BB tracers (levoglucosan and K^+), was investigated at three high-altitude
 421 Himalayan sites during the pre-monsoon period. The results showed that the central site
 422 exhibited the highest B_{abs} , with mean values of $53.1 \pm 18.5 \text{ Mm}^{-1}$ at 370 nm and $14.2 \pm$
 423 5.5 Mm^{-1} at 880 nm, and also showed elevated concentrations of BB tracers. The
 424 observed B_{abs} and levoglucosan concentrations were higher than those reported for the
 425 northeastern, central, and southwestern TP (Zhu et al., 2021; Zhu et al., 2022b), with
 426 the elevated levoglucosan concentrations suggesting a stronger influence of BB
 427 emissions along the southern slopes of the Himalayas. BC was the dominant light-
 428 absorbing component across the investigated wavelength range, consistent with
 429 observations from other high-altitude regions (Ramachandran et al., 2020; Zhu et al.,
 430 2022a). The BC contribution to total light absorption increased from the western to the
 431 eastern Himalayas, reaching $\sim 95\%$ at 660 nm at Zayü, which was higher than values
 432 reported for most other regions (Zhu et al., 2021). Across the investigated wavelengths,
 433 BrC_{pri} and BrC_{sec} contributed 8–28% and 3–26% of total light absorption, respectively,
 434 with the highest BrC_{pri} and BrC_{sec} contributions observed at the central and western
 435 sites, respectively. Additionally, the central Himalayan site exhibited a stronger BrC_{pri}
 436 contribution to solar radiation absorption, in contrast to other sites in the TP and
 437 Himalayan regions where BrC_{sec} was reported to make a greater contribution to
 438 radiative forcing (Zhu et al., 2021). Together with the elevated BB tracer concentrations,



439 the enhanced BrC_{pri} contribution suggests a stronger influence of primary combustion
 440 emissions, particularly BB emissions, in the central Himalayas.

441 Previous studies have shown that pollutant transport over the southern Himalayas is
 442 strongly influenced by mountain-valley circulation systems (Yu et al., 2024; Zhang et
 443 al., 2020a). Our results indicate that transboundary transport from South and Southeast
 444 Asia is strongly modulated by the complex Himalayan topography and terrain-induced
 445 atmospheric circulation, with particularly pronounced influences in the central
 446 Himalayan region. Importantly, consideration of the vertical distribution of aerosols
 447 provides additional insight into their regional transport and source characteristics.
 448 CALIPSO observations further reveal distinct vertical aerosol profiles, with mineral
 449 dust, pollution-modified dust, and BB-related aerosols predominating over the western,
 450 central, and eastern Himalayas, respectively. Together, these results highlight
 451 pronounced spatial heterogeneity in carbonaceous aerosol characteristics, transport
 452 pathways, and associated radiative impacts over the southern Himalayas. These
 453 findings further improve our understanding of the vertical distribution, sources, and
 454 atmospheric transformation of carbonaceous aerosols over the Himalayas.

455 However, it should be noted that quantitative estimations of BrC absorption based
 456 on spectral apportionment commonly rely on an assumed AAE_{BC}, which may introduce
 457 uncertainty into the separation of BC and BrC absorption because the spectral
 458 dependence of BC absorption can vary with its microphysical properties, including
 459 particle size, morphology, and coating characteristics (Luo et al., 2024; Luo et al., 2021;
 460 Zhang et al., 2020b). Nevertheless, this uncertainty is expected to primarily affect the
 461 quantitative partitioning of BC and BrC absorption, while the major spatial distribution
 462 and transport patterns of carbonaceous aerosols identified in this study are likely to
 463 remain robust.

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465

466



467 **Data availability**

468 The HYSPLIT model, developed by the NOAA Air Resources Laboratory, is accessible
469 via its official website at <https://www.ready.noaa.gov/HYSPLIT.php>. The
470 meteorological data used in the HYSPLIT model were from the GDAS
471 (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/>), and the CALIPSO data obtained from
472 the NASA Langley Research Center Atmospheric Science Data Center
473 (<https://asdc.larc.nasa.gov/>), the MODIS AOD time-averaged map was generated using
474 the GIOVANNI online data system (<https://giovanni.gsfc.nasa.gov/giovanni/>), and the
475 fire data were from the Fire Information for Resource Management System (FIRMS,
476 <https://firms.modaps.eosdis.nasa.gov/>).

477

478 **Author contributions**

479 Ju-Lian Shi: Conceptualization, Methodology, Formal analysis, Writing-original draft.
480 Chong-Shu Zhu: Conceptualization, Resources, Writing-review & editing.
481 Yao Qu: Investigation, Conceptualization, Data Curation, Validation.
482 Xiao Wang: Conceptualization, Data Curation, Formal analysis.
483 Nan Wang: Investigation, Data Curation.
484 Lu-Yao Wang: Investigation, Data Curation.
485 Wen-Ting Dai: Formal analysis, Conceptualization.
486 Hong Huang: Formal analysis, Conceptualization.
487 Jun-Ji Cao: Formal analysis, Conceptualization.

488

489 **Competing interests**

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

491

492 **Acknowledgments**

493 This research was funded by the National Natural Science Foundation of China
494 (42277098, 41771518), the Second Tibetan Plateau Scientific Expedition and Research



495 Program (STEP), Grant 2019QZKK0602, the Natural Science Basic Research Program
 496 of Shaanxi (Program No.2025JC-YBQN-430), and the Young Talent Fund of Xi'an
 497 Association for Science and Technology (0959202513117). The authors thank NASA
 498 for providing CALIPSO data and the MODIS AOD time-averaged map. The authors
 499 thank the NOAA Air Resources Laboratory for the HYSPLIT model and the NCEP for
 500 the GDAS meteorological data. The authors thank FIRMS for the fire counts data. We
 501 also appreciate the anonymous reviewers for their valuable comments.

502

503 **Financial support**

504 This research was funded by the National Natural Science Foundation of China
 505 (42277098, 41771518), the Second Tibetan Plateau Scientific Expedition and Research
 506 Program (STEP), Grant 2019QZKK0602, the Natural Science Basic Research Program
 507 of Shaanxi (Program No.2025JC-YBQN-430), and the Young Talent Fund of Xi'an
 508 Association for Science and Technology (0959202513117).

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