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Key Points:

- Secondary brown carbon (BrC) absorption coefficients were simultaneously measured at mountain and surface stations
- Despite limited precursors and weaker physicochemical drivers, the mountain station showed higher secondary BrC absorption
- Secondary BrC viscosity increased upon exposure to lower temperature and humidity, extending its lifetime and enhancing absorption

Supporting Information:

Supporting Information may be found in the online version of this article.

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Influence of Secondary Brown Carbon Aerosol Lifetime on Radiative Properties

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Abstract Brown carbon (BrC), a light-absorbing organic aerosol, exhibits higher absorption coefficients at short wavelengths. Partial BrC can be formed via secondary process. In this study, we isolated the secondary BrC ($\sigma_{\text{abs,sec BrC}}$) between urban surface and mountain stations (which receive surface pollutants during convective mixing) to investigate how atmospheric lifetime variations influence its absorption properties. Although the surface provided more favorable conditions for precursor concentrations and physicochemical reactions, the surface station observed lower $\sigma_{\text{abs,sec BrC}}$ during the most developed planetary boundary layer period. This was driven by low temperatures and relative humidity (RH) during uplift, which extended the atmospheric lifetime of high-viscosity secondary BrC, thereby enhancing $\sigma_{\text{abs,sec BrC}}$ observed at the mountain station. Variations in near-surface emission intensity dominated peak $\sigma_{\text{abs,sec BrC}}$ values observed at the mountain station during 14:00–15:00. Moreover, decreasing RH promoted higher viscosity, which enhanced $\sigma_{\text{abs,sec BrC}}$.

Plain Language Summary Brown carbon (BrC) significantly impacts atmospheric radiation, especially in the UV region of the electromagnetic spectrum. Higher absorption coefficients are observed during winter due to anthropogenic emissions and meteorological variations. This study demonstrates that secondary BrC viscosity increased after air mass exposure to low temperature and relative humidity, extending its atmospheric lifetime and resulting in higher absorption even under conditions with limited precursors and weaker physicochemical drivers.

1. Introduction

Brown carbon (BrC), a type of light-absorbing organic aerosol (OA), is generated from various sources including biomass burning, biofuel combustion (Ramanathan et al., 2007; Washenfelder et al., 2015), photooxidation of anthropogenic and biogenic volatile organic compounds (VOCs), and aqueous-phase chemistry in cloud droplets (Ervens et al., 2011; Graber & Rudich, 2006). BrC exhibits strong absorption at short wavelengths, particularly in the UV region, significantly affecting atmospheric radiation (Arola et al., 2011; Chen & Bond, 2010; Kirchstetter et al., 2004).

Recent studies have found that BrC optical properties are highly dependent on initial composition and secondary substance formation (Cappa et al., 2020; McClure et al., 2020). BrC absorptivity can be enhanced through two pathways: (a) formation of nitro-aromatics when aromatic compounds react with nitrogen oxides (NO_x)

(Nakayama et al., 2013); and (b) generation of organic amines when aromatics react with ammonia (Updyke et al., 2012). These processes increase BrC absorption either by nitrating existing chromophores or forming new secondary organic aerosol (SOA) chromophores via gas-phase oxidation. BrC absorption reduction is primarily driven by chromophore photobleaching, which occurs within hours to days (Qiu et al., 2024). This process is influenced by hydroxyl radicals, particle volatility, and relative humidity (RH). Several studies have demonstrated relatively rapid BrC absorptivity degradation through photobleaching (Forrister et al., 2015; Zhao et al., 2015).

The chemical lifetime of reactive compounds in atmospheric particles can increase from seconds to days as diffusion rates in semisolid phases decrease by multiple orders of magnitude under low temperature or low RH conditions (Shiraiwa et al., 2013). Consequently, long-range transport enables BrC aerosols to occur in various environments, including urban areas (Healy et al., 2015; X. Liu et al., 2019; Y. Wang, Hu, et al., 2019), plateau regions (Y. Wang et al., 2018; Y. Wang, Hu, et al., 2019), and even Arctic areas (Barrett & Sheesley, 2017; Stohl et al., 2006). The growth and evaporation rates of organic particles depend on particle viscosity (P. Liu et al., 2016; Maclean et al., 2017; Shiraiwa et al., 2017). Liquid organic particles with low viscosity respond rapidly to changes in gas-phase composition and equilibrate with ambient RH and temperature variations (Krieger et al., 2012; Shiraiwa et al., 2013). In contrast, highly viscous organic particles respond slowly to gas-phase compositional changes, particularly under dry or cold conditions (Koop et al., 2011; Mikhailov et al., 2009; Shiraiwa et al., 2013). Field measurements suggest that biogenic SOA particles are typically liquid or semi-solid under prevalent planetary boundary layer (PBL) conditions, largely due to the plasticizing effect of water. The phase state of anthropogenic SOA is more ambiguous, with evidence suggesting considerably higher particle viscosities (Maclean et al., 2017; Shiraiwa et al., 2017).

This study investigates the vertical transport of BrC absorbance between a surface site (50 m elevation, representative of anthropogenic emissions) and a mountain site (1,344 m elevation) in winter Beijing, where the mountain site receives surface emissions during convective mixing. By combining observations with the minimum R squared (MRS) method (C. Wu and Yu, 2016), we quantify primary and secondary BrC separately. Our study captures the enhancement in secondary BrC absorption coefficient ($\sigma_{\text{abs,sec BrC}}$) at the mountain station resulting from increased secondary BrC lifetime.

2. Materials and Methods

2.1. Site and Instrumentation

This study utilized continuous observations at two stations with different elevations. The surface station, located at the Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP; 39°58'28"N, 116°22'16"E; 50 m a.s.l.), is a typical urban station primarily influenced by anthropogenic emissions. The mountain station (115.78°E, 40.52°N; 1,344 m a.s.l.), situated northwest of Beijing, receives surface anthropogenic emissions during convective mixing (Figure S1 in Supporting Information S1). NO_x and O₃ were measured using gas analyzers (model 42i, and 49i, Thermo Scientific) at both sites.

The microphysical properties of black carbon (BC) particles (Figure S2 in Supporting Information S1) were measured by a single particle soot photometer (SP2, DMT Inc.) at both sites. The incandescent and scattering signals were calibrated by using Aquadag standard (Acheson Inc., USA) and monodispersed polystyrene latex spheres (PSL), respectively. The BC core diameter (D_c) was derived assuming a BC density of 1.8 g cm⁻³, combined with measured BC mass. Scattering signals for each BC-containing particle were reconstructed using the leading-edge-only method (R. Gao et al., 2007), and core sizes were input into a Mie lookup table to derive coated particle diameters (D_p) (D. Liu et al., 2014; Taylor et al., 2015). The bulk volume ratio (VR) is derived from the ratio of coated BC volume to BC core volume, integrated over all single particles within a given time window. Since the Mie core-shell model overestimates BC absorption properties compared to measurements, we applied a partial core-shell model to reduce this bias. This approach quantifies the dynamic shape factor to describe the sphericity of BC-containing particles (Hu et al., 2021), and establishes the relationship between nonsphericity fraction (F_{ns}) and VR for BC-containing particles (Hu et al., 2022):

$$F_{\text{ns}} = -0.27 \log(\text{VR}) + 0.64 \quad (1)$$

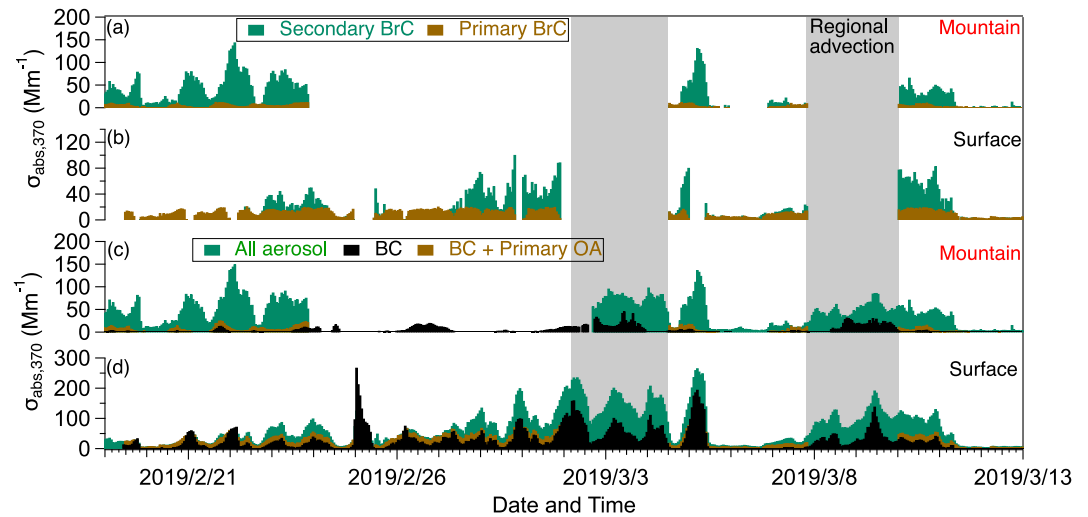


Figure 1. Temporal evolutions of $\sigma_{\text{abs},370}$ for primary (brown) and secondary (green) Brown carbon at mountain (a) and surface (b) sites; and $\sigma_{\text{abs},370}$ for all aerosol (green), black carbon (BC, black), and the sum of BC and primary organic aerosol (OA, brown) at mountain (c) and surface (d) sites. The grey shades indicate the regional advection periods, which was excluded in our analysis.

F_{ns} represent the mass fraction of BC that can be treated as nonspherical particles. Uncertainty in F_{ns} may arise from varying the dynamic shape factor threshold (set at 1.75 in the original study), varying this from 1.6 to 1.9 introduces $\pm 12\%$ uncertainty. Since only nearly spherical BC can be considered as a core-shell structure, the partial core-shell mass absorption coefficient ($\sigma_{\text{partial core-shell}}$, Figure S4 in Supporting Information S1) can be expressed by combining F_{ns} , refractory BC mass ($[\text{rBC}]$), and uncoated ($\text{MAC}_{\text{uncoated}}$) and coated ($\text{MAC}_{\text{coated}}$) mass absorption cross-sections (derived using Mie theory with D_c and D_p , Figure S3 in Supporting Information S1) as:

$$\sigma_{\text{partial core-shell}} = ([\text{rBC}] \times F_{\text{ns}} \times \text{MAC}_{\text{uncoated}} + [\text{rBC}] \times (1 - F_{\text{ns}}) \times \text{MAC}_{\text{coated}}) \quad (2)$$

An Aethalometer AE33 (Magee Scientific Inc., USA) with a $\text{PM}_{2.5}$ impactor (BGI SCC1.829) measured absorption coefficient (σ_{abs}) at 370 nm at both sites (Figure S2a in Supporting Information S1). Aerosol was dried using a Nafion tube. The AE33 employs dual-spot measurements at different flow rates to automatically correct for filter loading effects (Hu et al., 2024). The σ_{abs} measured by the AE33 were compared with photoacoustic soot spectrometer (PASS-3, DMT) measurements (Arnott et al., 2005), yielding multi-scattering correction factors (C-values) of 3.5 and 2.9 at $\lambda = 370$ nm for the surface and mountain sites, respectively.

2.2. Air Mass History

The Numerical Atmospheric-dispersion Modeling Environment (NAME) (Jones et al., 2007), driven by meteorological data from the global configuration of the U.K. Met Office's Unified Model, was employed to identify air masses sources (Figure S2d in Supporting Information S1). Particle fluxes from each NAME model grid were integrated over segregated regions, and contributions of each air mass fractions could be obtained to distinguish convective mixing periods from regional advection (D. Liu et al., 2020).

2.3. Attribution of Primary and Secondary BrC Absorption

The σ_{abs} of BrC ($\sigma_{\text{abs,BrC}}$) was derived by subtracting the σ_{abs} of BC ($\sigma_{\text{abs,BC}}$) from the total σ_{abs} , expressed as:

$$\sigma_{\text{abs,BrC}}(\lambda) = \sigma_{\text{abs,total}}(\lambda) - \sigma_{\text{abs,BC}}(\lambda) \quad (3)$$

Notably, the $\sigma_{\text{abs,BC}}$ was calculated using Equation 2, to avoid the inaccuracy of assuming BC absorption Ångström exponent equals unity. The σ_{abs} of secondary BrC ($\sigma_{\text{abs,Sec BrC}}$, Figure 1) was calculated by subtracting primary source contributions from total σ_{abs} , expressed as:

$$\sigma_{\text{abs,secBrC}}(\lambda) = \sigma_{\text{abs,total}}(\lambda) - \sigma_{\text{abs,pri}}(\lambda) \quad (4)$$

where $\sigma_{\text{abs,pri}}(\lambda)$ is the light absorption from primary sources. We assumed that light absorption of aerosols from combustion sources contains BC (Q. Wang, Ye, et al., 2019). Consequently, the total σ_{abs} of primary source was obtained by scaling the rBC mass loading by a factor:

$$\sigma_{\text{abs,pri}}(\lambda) = \left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}} \cdot [\text{rBC}] \quad (5)$$

where [rBC] is the rBC mass measured by the SP2 (to exclude secondary aerosol influences measured by AE33), and $\left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}}$ is the scaling factor used to derive the absorption of primary combustion sources from [rBC]. This scaling factor was obtained using the MRS method, which has been validated for quantifying σ_{abs} of primary and secondary BrC (Li et al., 2023; Q. Wang, Ye, et al., 2019). The theoretical basis is that secondary BrC and rBC exhibit minimal correlation. The MRS method present different $\left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}}$ results for traffic and biomass burning sources, as well as for different air mass sources, which may lead to biases in derived results. However, as this study focuses solely on the convective mixing process, which is dominated by local anthropogenic emissions, differences in emissions and air mass sources are not considered to have significantly influenced the patterns of derived parameters. Moreover, this method is only valid with sufficient data points. As a result, we may only obtain a single mean value representing the mean $\left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}}$ during the experimental period. Previous studies have utilized the MRS method to investigate the mean $\left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}}$ results by focusing on traffic, coal combustion and biomass burning in urban environment (Y. Gao et al., 2022; Q. Wang, Han, et al., 2019). In this study, the $\left(\frac{\sigma_{\text{abs,total}}}{[\text{rBC}]} \right)_{\text{pri}}$ value at surface and mountain at 370 nm is 26 and 21.2, respectively (Figure S5 in Supporting Information S1), which falls within the previously reported range of 11–50 (Q. Wang, Ye, et al., 2019; Zhang et al., 2020). The primary BrC σ_{abs} can be derived by:

$$\sigma_{\text{abs,priBrC}}(\lambda) = \sigma_{\text{abs,BrC}}(\lambda) - \sigma_{\text{abs,sec BrC}}(\lambda) \quad (6)$$

Notably, the apportionment method may introduce errors in distinguishing absorption coefficients, potentially misattributing some primary sources as secondary and vice versa.

3. Results and Discussion

This study used the same period and site as D. Liu et al. (2020), who used the same aerosol measurement instruments at mountain and surface stations to confirm that non-volatile aerosols like BC were effectively transported to the PBL top, while volatile and semi-volatile aerosols were not. The mountain station observed nearly identical BC mass concentrations as the surface station from 12:00 to 15:00, defined here as the most developed PBL period (green bar in Figures 2b and 2c), when surface-emitted aerosols reached the mountain site through uplift.

3.1. Absorption and Vertical Transport Properties of Primary BrC

Surface-observed primary BrC ($\sigma_{\text{abs,pri BrC}}$) exhibited distinct diurnal variation (Figure 2b). As primary BrC is dominated by emission sources, peak values observed during rush hours. Studies have shown that traffic sources in Beijing, particularly diesel vehicles, emit substantial amounts of organic aerosols containing chromophores such as aromatic (Yao et al., 2015) and heterocyclic organic compounds (Gentner et al., 2017; Schuetzle, 1983). Beyond rush hour effects, planetary boundary layer height (PBLH) variation also critically influences surface primary BrC absorption. The midday increase in PBLH dilutes aerosol concentrations, creating a valley-like distribution for primary BrC.

Mountain station-observed primary BrC did not exhibit distinct rush hour peaks, likely due to the station's relatively clean environment and reduced influence from emission sources (Figure 2b). Consequently, $\sigma_{\text{abs,pri BrC}}$

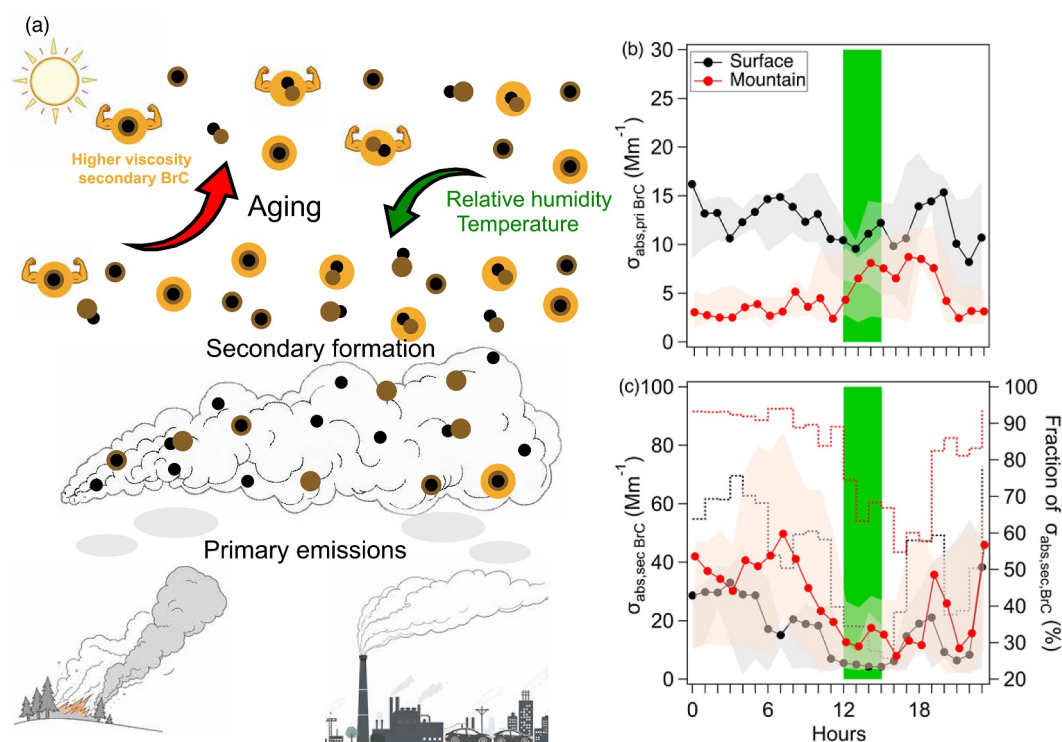


Figure 2. Schematic illustration (a). Diurnal variations of (b) $\sigma_{\text{abs,pri BrC}}$, (c) $\sigma_{\text{abs,sec BrC}}$ at 370 nm (solid lines and markers) and fraction of $\sigma_{\text{abs,sec BrC}}$ (dashed lines) at mountain (red) and surface (black) sites during convective mixing events, with the grey and pink shadow upper and lower boundaries indicating 75th and 25th percentiles, respectively. The green bar represents the time of the day (12:00–15:00) with the most developed planetary boundary layer.

at the mountain station correlated strongly with PBLH development. During the most developed PBL period, surface primary BrC was partially transported to the PBL top, with the fraction of $\sigma_{\text{abs,pri BrC}}$ reaching 37%. However, convective mixing did not transport all primary BrC efficiently, thus, $\sigma_{\text{abs,pri BrC}}$ at the mountain station remained consistently lower than at the surface.

3.2. Absorption Properties of Secondary BrC at Both Surface and Mountain Sites

The diurnal variation of surface $\sigma_{\text{abs,sec BrC,370}}$ showed elevated values at night (Figure 2c) when surface RH ranged between 45% and 50% (Figure S7d in Supporting Information S1), conditions that promote secondary BrC absorption. Kasthuriarachchi et al. (2020) found that the MAC of droplet-formed secondary BrC peaks at RH of 55%–65%. As RH gradually decreased to 27% throughout the day, $\sigma_{\text{abs,sec BrC,370}}$ declined accordingly. In addition to RH, changes in the PBLH also influenced secondary BrC. Surface $\sigma_{\text{abs,sec BrC,370}}$ increased at night due to the accumulation caused by a decrease in the PBLH, but decreased during the day, particularly at noon. Odd oxygen ($\text{O}_x = \text{NO}_2 + \text{O}_3$) is an indicator of air mass aging caused by photochemical reactions (Canonaco et al., 2015), showed a weak negative correlation with $\sigma_{\text{abs,sec BrC}}$ ($R^2 = 0.22$; Figure S6a in Supporting Information S1) during daytime.

The diurnal mountain variation of $\sigma_{\text{abs,sec BrC,370}}$ showed similar patterns to the surface. At night, RH at the mountain station exceeded surface values (Figure S7d in Supporting Information S1) and approached the optimal 55%–65% range for liquid-phase secondary BrC formation. Additionally, a peak transition from primary to secondary BrC at 19:00 suggested active SOA formation. As O_x concentration increased, daytime mountain secondary BrC absorption decreased significantly ($R^2 = 0.81$; Figure S6b in Supporting Information S1), indicating photobleaching caused by photolysis and/or photooxidation as the day progressed.

3.3. Comparative Analysis of Secondary BrC Between Mountain and Surface

Generally, surface pollution concentrations exceed mountain values due to cleaner mountain conditions. Previous research at the same site and period found that surface VOCs concentrations and condensation rates exceeded mountain values (D. Liu et al., 2020). However, this study observed lower $\sigma_{\text{abs,sec BrC},370}$ at the surface than at the mountain station. Since our research was conducted in an urban area, the high NO_x emissions of traffic VOCs may have substantially influenced nitrogen chemistry in photochemical processes (e.g., aromatic hydrocarbons promote NO to NO_2 conversion (Atkinson, 2000)). Although the lowest surface NO_x value was 1.7 times higher than the highest mountain value (Figure S7b in Supporting Information S1), the surface observed $\sigma_{\text{abs,sec BrC},370}$ did not exceed mountain values during the most developed PBL period. Figure 2b shows that surface $\sigma_{\text{abs,pri BrC},370}$ exceeded mountain values, indicating greater availability of surface primary BrC for photochemical conversion to secondary BrC. Aqueous-phase reactions may have promoted secondary BrC formation. Field studies have shown that biomass-burning emissions can form BrC in clouds and fogs (Desyaterik et al., 2013; Gilardoni et al., 2016). However, given the relatively low RH measured during most developed PBL period (Figure S7d in Supporting Information S1) and extremely rare precipitation (Figure S2a in Supporting Information S1), aqueous-phase reactions likely did not dominate secondary BrC formation.

The above findings demonstrate that the surface should have more favorable conditions for secondary BrC generation, yet $\sigma_{\text{abs,sec BrC},370}$ showed the opposite pattern. Q. Liu et al. (2021) also found higher SOA concentrations at the mountain than the surface at the same measurement sites during winter in Beijing. Similarly Y. Wu et al. (2024) utilized PMF and light absorption measurements to reveal higher fractions of secondary BrC at the mountain compared to the surface after aging. Beyond secondary BrC generation, extended atmospheric lifetimes of secondary BrC also contributed to higher $\sigma_{\text{abs,sec BrC},370}$. This conclusion is supported by efficient BC uplift attributed to its sufficient atmospheric lifetime, but does not apply to more volatile aerosols (D. Liu et al., 2020). High viscosity increases the chemical lifetime of reactive compounds in atmospheric particles under dry conditions or low temperatures (Figure 2a) (Koop et al., 2011; Mikhailov et al., 2009; Shiraiwa et al., 2011, 2013).

3.4. Extended Atmospheric Lifetime of High-Viscosity Secondary BrC Aerosols

Lower temperatures extended the viscosity of secondary BrC (Fulcher, 1925), further enhancing $\sigma_{\text{abs,sec BrC},370}$. The surface station exhibited a temperature of $11.8 \pm 0.9^\circ\text{C}$, while the mountain station presented $2.6 \pm 0.3^\circ\text{C}$ during the most developed PBL period, emphasizing more favorable conditions for high-viscosity secondary BrC generation during uplift (Figure S7e in Supporting Information S1). Figure 2c shows that mountain station $\sigma_{\text{abs,sec BrC},370}$ decreased during 12:00–13:00 but increased at 14:00. This diurnal variation mirrored surface observed $\sigma_{\text{abs,pri BrC},370}$, indicating that precursor concentrations from increased surface anthropogenic emissions dominated the observed mountain $\sigma_{\text{abs,sec BrC},370}$ enhancement. Additionally, decreasing RH during 14:00–15:00 promoted higher secondary BrC viscosity, further enhancing $\sigma_{\text{abs,sec BrC},370}$. A recovery of the fraction of $\sigma_{\text{abs,sec BrC}}$ can be observed during this period (Figure 2c). The impact of RH on viscosity primarily stems from water acting as a plasticizer, increasing molecular mobility and reducing viscosity (Russell et al., 2011; Zobrist et al., 2011). The low temperature and RH conditions at the mountain station provided a favorable environment for higher-viscosity and potentially solid-glassy secondary BrC. Amorphous phases can be classified as glasses, rubbers, gels, or ultraviscous liquids (Russell et al., 2011). Research has shown that SOA with viscosity exceeding 10^{12} Pa·s exhibits a solid glassy state (Reid et al., 2018). Virtanen et al. (2010) first confirmed this in ambient atmospheric measurements, where a fraction of biogenic OA rebounded upon striking an impactor stage, providing the first evidence that atmospheric particles could exist as highly viscous semi-solids or amorphous solids. The mountain station provided favorable conditions for glassy secondary BrC formation, especially during 14:00–15:00, due to elevated molecular weight after aging (Di Lorenzo et al., 2017; Q. Liu et al., 2021; Saleh et al., 2014; Y. Wu et al., 2024), peak diurnal O:C ratios and minimum diurnal RH results at both stations (Koop et al., 2011; Shiraiwa et al., 2013, 2017; Zobrist et al., 2008). Notably, enhanced secondary BrC viscosity is inferred mechanistically rather than directly observed in our experiment. Additionally, our findings do not suggest that high-viscosity secondary BrC with longer atmospheric lifetimes occurs only under low temperature and RH conditions. Rather, these environmental conditions enhanced the abundance of such aerosols.

4. Conclusion

This study utilized continuous and simultaneous measurements at both surface and mountain stations to explore the vertical transport of BrC during the convective mixing process. During the most developed PBL period, surface contains with more favorable conditions for precursor concentrations and physicochemical reactions, but observed constant lower secondary BrC absorption coefficient ($\sigma_{\text{abs,sec BrC},370}$). Low RH and temperature during uplift may have promoted secondary BrC viscosity and extended its atmospheric lifetime, resulting in higher $\sigma_{\text{abs,sec BrC},370}$.

This phenomenon may provide an additional explanation for the higher BrC concentrations observed in winter. In addition to variations in anthropogenic emissions, the extended atmospheric lifetime of high-viscosity secondary BrC caused by lower temperature and RH increased the observed $\sigma_{\text{abs,sec BrC},370}$. This phenomenon also emphasizes that when polluted air masses are transported to colder and drier areas, for instance from south regions of Beijing (which have higher pollution concentrations, temperature and RH) to Beijing China during winter, or from urban to suburban areas, the pollution caused by secondary BrC will increase. Moreover, toxic compounds are embedded in high viscosity secondary BrC especially glassy secondary BrC, reducing the attenuation ability of toxicity during transport, further affecting human health at the target site (Shiraiwa et al., 2017).

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Availability Statement

Data is available at Hu et al. (2026).

Acknowledgments

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