

Measurement report: Contrasting elevation-dependent changes in light absorption of by black and brown carbon: lessons from *in-situ* measurements from highly polluted Sichuan Basin to pristine Tibetan Plateau

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Abstract. The scientific knowledge on light absorption of by aerosols is extremely limited at the eastern slope of the Tibetan Plateau (ESTP). We conducted the first aerosol field experiment at six sites (Chengdu, Sanbacun, Wenchuan, Lixian, Maerkang, Hongyuan) along the ESTP ranging in elevation extending elevation from 500 m to 3500 m. The fraction of light absorption by brown carbon (BrC) to total carbon increases from 20%~~The light absorption of brown carbon (BrC) accounting for that of total carbon increases from 20%~~ to 50% with altitude, and the mass absorption efficiency (MAE) of BrC over the TP is 2–3 times higher than that inside the ~~SiChuan-Sichuan~~ Basin (SCB), especially in winter. In contrast, the MAE of elemental carbon (EC) in winter decreases with altitude~~Contrary to BrC aerosols, winter EC (elemental carbon) mass absorption efficiency declines with altitude~~. The contrasting variation of EC and BrC MAE with altitude is mainly attributed to source difference between the TP and SCB. Emissions from the~~The~~ more urban sources (motor vehicles, industries, etc.) inside the SCB fail to be transported to the TP due to the stable air in winter inside~~to winter stable air inside~~ the basin, which ~~also is~~ also favorable for aerosol ageing to enhance absorption efficiency. The

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radiative forcing of BrC relative to EC varies from 0.10 to 0.42 as altitude increases with the higher OC/EC ratio over the TP than SCB. ~~Thus, the reason of the enhanced relative BrC to EC radiative forcing from polluted SCB to pristine TP is that the BrC concentration decreases more slowly than the EC concentration with altitude and thus the enhanced radiative forcing of BrC relative to EC from polluted SCB to pristine TP is because the concentration of OC decreases more slowly with altitude than does EC. This study contributes to the understanding of the difference in light absorption by EC and BrC with altitude, from polluted lower-altitude basins to the pristine TP, and provides a data set for regional climate model validation. This study will deepen the understanding of EC or BrC light absorption difference between the highly polluted basins and clean TP and provide a basic data set for optimization of regional climate modeling.~~

1 Introduction

Some *in-situ* observations, available satellite data, and model simulations indicate that a greater surface warming trend over time occurs at higher altitudes in the mountainous regions worldwide over the world (Gao et al., 2018; Guo et al., 2019; Mountain Research Initiative EDW Working Group, 2015; Palazzi et al., 2017; Pepin et al., 2019; Rangwala and Miller, 2012; You et al., 2020). Rangwala and Miller (2012) reviewed elevation-dependent warming (EDW) and its possible causes over four high mountain regions, i.e., the Swiss Alps, the Colorado Rocky Mountains, the Tibetan Plateau (TP), and the Tropical Andes. Their examinations found that the available observations indicate that some mountain regions show much greater warming rates at seasonal scales than others. The mechanisms that can produce enhanced warming rates at higher altitudes may be related to the differential sensitivities of surface warming to changes in the climate drivers, at different elevations, such as snow-ice cover, clouds, atmospheric water vapor, aerosols, land use, and vegetation, at different elevations (Rangwala and Miller, 2012; You et al., 2020).

The Tibetan Plateau (TP, hereafter), known as the “third pole”, is an ideal place to examine EDW and its mechanism (Guo et al., 2021). The warming rates (rising temperature per 10 years) over the TP were found to be the most notable in winter and autumn (Liu and Chen, 2000), especially for the central and eastern Plateaus (Duan and Wu, 2006), which may be partly associated with human activities, such as more anthropogenic emissions in the sub-regions (Lu et al., 2010). The effect of

carbonaceous aerosols on regional and even global climate is more uncertain ~~because of~~ their shorter life than ~~the~~ long-lived aerosols, such as carbon dioxide and methane (Chung et al., 2012; Ramanathan and Carmichael, 2008). ~~The~~ absorbing aerosols (black carbon and dust) from local emissions or long-range transport heat the atmosphere in two ways (Tian et al., 2018). They absorb radiation and decrease the surface albedo when deposited on snow and ice (Kang et al., 2019; Lau et al., 2010; Xu et al., 2009). Ramanathan and Carmichael (2008) suggested that black carbon (BC) in the Himalayas, arising from anthropogenic activities ~~in~~ the Indo-Gangetic Plain (IGP), could account for half of the local warming during the past several decades. In addition to the well-known BC, the recent work by Wu et al. (2018) suggested that the light absorption efficiency (LAE) of brown carbon (BrC, a certain type of organic aerosols) in winter is 2–3 times higher than that in summer for the central ~~Tibetan Plateau~~. However, ~~the~~ scientific knowledge ~~of~~ the optical properties of carbonaceous aerosols (elemental carbon (EC), BrC) ~~is extremely limited~~ over ~~the~~ eastern TP is extremely limited, and *in-situ* aerosol measurements at varying altitudes from ~~the~~ the heavily polluted Basins to the relatively clean TP ~~are~~ crucial ~~important~~ for better understanding their light absorption.

~~The~~ previous *in-situ* measurements have mainly ~~primarily~~ focused on the southern and northern slopes (Cong et al., 2015; Huang et al., 2007; Kang et al., 2020), ~~whereas~~ the fewer observations have been ~~were~~ conducted ~~on~~ the eastern slope of the TP (ESTP). ~~The~~ SiChuan Basin (SCB) ~~—~~, a highly polluted region in China caused by ~~due to~~ more rapid economic development ~~—~~, is located on the eastern side of the TP (Zhao et al., 2018). The BrC LAE was strong inside the Basin (Peng et al., 2020a), especially ~~in~~ for ~~the~~ rural areas, because of ~~due to~~ increased ~~more~~ biomass and coal burning impacts (Zhao et al., 2021). Our previous studies ~~works~~ indicated that aerosols from the SCB are transported upslope along the ESTP and reach the eastern part of the TP by gradient *in-situ* observations at the ESTP (Yin et al., 2020). The recent study by S. Y. Zhao et al. (2020) suggested the strong light-absorbing BrC from biomass and coal burning inside the Basin can be transported to the main part of the TP by the enhanced “‘heat pump”’ in response to rapid warming over the TP. The aerosols over the TP from local emissions and long-range transport from the surrounding highly polluted areas ~~affected~~ its weather, climate, and water cycle (C. F. Zhao et al., 2020). ~~The~~ clouds and radiation are particularly sensitive to aerosols over pristine regions (Garrett and Zhao, 2006; Zhang et al., 2021). However, it is unclear ~~fuzzy~~ whether ~~that~~ the light absorption and radiative forcing of

carbonaceous aerosols change from ~~the~~ highly polluted SCB to ~~the~~ cleaner TP.

In this ~~studywork~~, we investigated the changes in ~~the~~ light absorption of carbonaceous aerosols (EC, BrC) and calculated ~~the~~ relative radiative forcing of BrC to EC aerosols from ~~the~~ SCB to TP in the four seasons. The sources and origins ~~were~~ also ~~were~~ determined ~~by using some~~ statistical methods and ~~the hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) back-trajectory~~ model. ~~Our goals are~~ ~~We aimed~~ to understand ~~the difference in~~ EC or BrC light absorption ~~difference~~ between the highly polluted Basins and clean TP, ~~and~~ to reveal the ~~corresponding mechanisms~~ ~~causes of the difference~~, and to ~~provide-generate~~ a basic data set for ~~the~~ optimis~~z~~ation of regional climate modell~~ing~~.

10 2 Data and methods

2.1 Observation sites and aerosol sampling

Compared ~~with to that of~~ the coarser fraction of ~~particulate matter (PM)~~, ~~the size of~~ strong light-absorbing carbonaceous particles ~~wasare mainly/primarily located in~~ ~~the~~ submicron range. Therefore, ~~samples PM₁~~ ~~(of~~ particulate matter with ~~an~~ aerodynamic diameter smaller than 1 μm ~~(PM₁) samples-~~ were collected at six sites (Chengdu, Sanbacun, Wenchuan, Lixian, Maerkang, and Hongyuan) from ~~the~~ western SCB to ~~the~~ eastern part of ~~the~~ TP ~~at elevations varying with varying elevation~~ from 500 m to 3500 m (Figure 1, Table 1). Each sampling site ~~was~~ selected to represent ~~the~~ background level at ~~the~~ local scale as completely as possible, without local emission impacts. ~~A total of~~ ~~The~~ 1024 PM₁ samples ~~in total~~ were collected from ~~21~~ December ~~21~~, 2018 to ~~18~~ December ~~18~~, 2019 on a ~~day/night/day/night~~ pattern ~~using an~~by aerosol sampler (LY-2034, Laoying Instrument Co., Ltd., China) at ~~at~~the flow rate of 100 L min⁻¹. The samples were stored frozen in pre-baked glass jars until further analysis (Kawamura et al., 2010). ~~M~~~~The~~ meteorological variables (temperature, relative humidity, wind speed, and direction) were downloaded ~~from by~~ ~~the~~ China Meteorological Data Service Center (<http://data.cma.cn/>). PM₁ samples were collected near ~~the~~ meteorological observation sites; ~~and thus~~, the meteorological variables ~~could~~ean represent the situation ~~in at~~ the study region. ~~The~~ MODIS active fire data (<https://earthdata.nasa.gov/active-fire-data>) ~~also~~ were used in this study.

2.2 Chemical analysis

A quarter of each filter was used to analy~~ze~~ water-soluble inorganic ions (Na⁺, NH₄⁺, K⁺, Ca²⁺, Mg²⁺,

F⁻, Cl⁻, SO₄²⁻, and NO₃⁻), and the ions were extracted and filtered using ultrapure water and a 0.45 μm pore syringe filter. The concentrations of the cations and anions were measured using ion chromatography (DX-600 & ICS-2500, Dionex, USA). The carbonaceous aerosols, that is, organic carbon (OC) and Elemental carbon (EC), were analysed using a seven-wavelength carbon analyser (Model-2015, DRI, USA). The carbon analyser measured the OC and EC concentrations using the thermal/optical reflectance (TOR) method (Chow et al., 2007). Briefly, the OC/EC was determined by progressively heating the sub-filter. The OC fractions were determined by heating at 120 °C (OC1), 250 °C (OC2), 450 °C (OC3), and 550 °C (OC4) in a pure He atmosphere. Subsequently, EC fractions were measured at 550 °C (EC1), 700 °C (EC2), and 800 °C (EC3) in an oxidising atmosphere of 2% O₂ and 98% He. The carbon involved is oxidised to CO₂ and then reduced to CH₄ for detection by a flame ionisation detector. The pyrolysed organic carbon (OPC) was monitored when the reflected laser signal returned to its initial value after the introduction of O₂ into the analysis atmosphere. The OC was defined as the sum of OC1, OC2, OC3, OC4, and OPC, whereas the EC was defined as EC1 + EC2 + EC3 – OPC. The EC and BrC were derived from the light absorption coefficient (b_{abs}) depending on the transmittance attenuation. For the seven-wavelength carbon analyser, the filter transmittance (FR_λ, fraction of light transmitted through the filter) uncertainties ranged from 5% to 18%, with the best precision observed at 450 nm and 808 nm (Chen et al., 2015). This uncertainty is attributed to the quality of the laser and the sensitivity of the photodiode detector for different wavelengths.

The coefficient of variation (CV), in conjunction with correlation coefficients (r), can be used to characterise the intra-location variability of chemical species (Zhao et al., 2021). The CV was calculated using the following equation:

$$CV_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^p \left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}, \quad (1)$$

where x_{ij} and x_{ik} are the average concentrations of a chemical component i at sites j and k , respectively; and p is the number of samples. The CV values of zero and approaching one indicate mean no difference and absolute heterogeneity between the two sites for the specific chemical component, respectively. A CV lower than 0.2 is usually considered to represent a relatively similarity of spatial pattern (Wang et al., 2018).

2.3 Calculation of light absorption parameters

The BrC light absorption increases sharply as ~~the wavelength decreases~~~~decreased wavelength~~, and thus, it can be separated from ~~the~~ EC (Peng et al., 2020a). The light absorption induced by carbonaceous aerosols (~~the~~ sum of EC and BrC) on a quartz filter was estimated ~~using~~~~by~~ an algorithm of transmittance attenuation (ATN):

$$ATN_{\lambda} = \ln \left(\frac{FT_{\lambda,a}}{FT_{\lambda,b}} \right), \quad (2)$$

where, $FT_{\lambda,a}$ and $FT_{\lambda,b}$ ~~in the right hand~~ represent ~~the~~ filter transmittance after and before thermal analysis for the specific wavelength (λ), ~~respectively~~. Referring to the work by Chen et al. (2015), the relationship ~~between~~~~of~~ ATN ~~and~~ ~~with~~ the absorption optical depth (τ_a) can be given as follows:

$$\tau_{a,\lambda} = a_{\lambda} \times ATN_{\lambda}^2 + c_{\lambda} \times ATN_{\lambda} \quad (3)$$

This study uses ~~the~~ the two coefficients (a_{λ} and c_{λ}) reported by Chen et al. (2015). The light ~~absorption-~~~~absorption~~ coefficients (b_{abs}) ~~were can be~~ calculated ~~using the following with the~~ equation:

$$b_{abs,\lambda} = \tau_{a,\lambda} \times \left(\frac{A}{V} \right), \quad (4)$$

where, A and V are ~~the~~ filter area and ~~the~~ sampling volume, respectively. The total b_{abs} can be separated into EC and BrC ~~using~~~~by~~ a simplified two-component model (Chen et al., 2015):

$$b_{abs,\lambda} = b_{abs,\lambda,EC} + b_{abs,\lambda,BrC} = K_1 \times \lambda^{-AAE_{EC}} + K_2 \times \lambda^{-AAE_{BrC}}, \quad (5)$$

where, K_1 and K_2 are fitting coefficients. AAE_{EC} and AAE_{BrC} represent ~~the~~ EC and BrC absorption Ångström exponents (AAE), respectively. They do not change ~~as with~~ the wavelength. AAE_{EC} was assumed ~~to be~~ 1 (Bond, 2001), and the other three parameters in Eq. (5) were obtained for AAE_{BrC} values between 2 and 8 with ~~an~~~~the~~ increment of 0.1, by least-square linear regression. ~~T-~~~~and~~~~the~~ AAE_{BrC} that led to the overall best fit in terms of R^2 was selected as the effective BrC AAE. The mass absorption efficiency (MAE) was obtained ~~from~~~~by~~ the ratio of light absorption coefficients ($b_{abs,\lambda,EC}$ or $b_{abs,\lambda,BrC}$) to the corresponding EC or OC mass concentrations (Olson et al., 2015). The estimated MAE_{BrC} was much lower than the true value by replacing BrC with OC, ~~because~~~~due to~~ BrC accounts ~~ing~~ for only a small fraction of OC. The main shortcoming of the separation of ~~the~~ total aerosol absorption into EC and BrC (Eq. 5) ~~does not consider~~~~is lack of considering the~~ mineral dust

impacts. According to ~~the~~ recent study ~~by~~ Zhang et al. (2021), mineral dust may be ~~an~~ ~~important~~ ~~atmospheric aerosol species~~ ~~species of the atmospheric aerosols~~ over the Tibetan Plateau. However, the study region is located ~~on~~ the eastern slope of ~~the~~ TP ~~during our campaign~~, which is more easily affected by anthropogenic sources from ~~the~~ heavily polluted Sichuan Basin than natural sources such as mineral dust (Yin et al., 2020) as compared to the northern areas close to ~~the~~ Taklimakan and Gobi Deserts. ~~The~~ One main aim of this study ~~was~~ to reveal the gradient distributions of aerosol optical properties from the pollution ~~of the Sichuan Basin~~ to ~~the~~ eastern TP; ~~and thus~~, the impact of ~~this~~ shortcoming may be negligible when studying the spatial heterogeneity of aerosol optical properties at ~~a~~ relatively small spatial scale. ~~In a~~ ~~addition~~ ~~ally~~, ~~the~~ AAE of EC ~~was~~ assumed to ~~be~~ 1, and the ageing of EC ~~was not considered~~ ~~did not take~~ when separating the total aerosol absorption into EC and BrC (Eq. 5) in our study.

The ~~light~~ absorbed ~~light~~ by ~~the~~ carbonaceous component can be estimated as follows (Huang et al., 2018):

$$15 \quad \frac{I_0 - I}{I_0}(\lambda, EC) = 1 - e^{-\left(MAE_{\lambda_0, EC} \times \left[\frac{\lambda_0}{\lambda} \right]^{AAE_{EC}} \times C_{EC} \times PBLH \right)} \quad (6)$$

$$\frac{I_0 - I}{I_0}(\lambda, BrC) = 1 - e^{-\left(MAE_{\lambda_0, BrC} \times \left[\frac{\lambda_0}{\lambda} \right]^{AAE_{BrC}} \times C_{OC} \times PBLH \right)}, \quad (7)$$

where, 405 nm is ~~the~~ ~~determined as~~ reference wavelength λ_0 , and C_{EC} and C_{OC} represent ~~the~~ EC and OC concentrations, respectively. The planetary boundary layer (PBL) height (~~PBLH~~) was obtained from the HYSPLIT model, and ~~we assumed~~ no vertical gradients ~~were assumed~~ within the PBL. ~~This~~ assumption might overestimate the radiative forcing of aerosols, while ~~it that~~ has ~~a~~ small effect on the radiative forcing of BrC relative to EC (f), which can be estimated ~~using~~ ~~by~~ the ~~fol~~ ~~lowing~~ equation (Zhao et al., 2019):

$$f = \frac{\int I_0(\lambda) \left[\frac{I_0 - I}{I_0}(\lambda, BrC) \right] d\lambda}{\int I_0(\lambda) \left[\frac{I_0 - I}{I_0}(\lambda, EC) \right] d\lambda}, \quad (8)$$

where $I_0(\lambda)$ is ~~the~~ wavelength-dependent solar emission flux, which is ~~the~~ clear sky ~~a~~ ~~air~~ ~~mass~~ 1 ~~g~~ ~~global~~ ~~h~~ ~~horizontal~~ solar irradiance (Levinson et al., 2010). ~~The~~ light absorption by BrC at 405 nm and 445 nm is much stronger than that at ~~the~~ longer wavelengths inside the SCB (Zhao et al., 2021).

The 405 nm ~~wavelength was~~ the lower limit of detection ~~of by~~ the ~~DRI-2015~~ instrument ~~of DRI-2015~~. Therefore, ~~the~~ fraction (f) ~~was~~ obtained by numerical integration of the above formula ~~in at~~ the wavelength ranges of 405–980 nm and 405–445 nm for each sample, ~~respectively~~. ~~The~~ nighttime samples were excluded when calculating the radiative forcing of ~~the~~ BrC relative to ~~the~~ EC.

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The exponential function was selected to fit the relationships between BrC MAE and altitude (AT). The equation is ~~given~~ as follows:

$$MAE_{\lambda, BrC} = a_{\lambda} \cdot e^{b \times AT}, \quad (9)$$

where, a_{λ} and b are the fitted coefficients. The EC MAE can be parameterized with ~~the~~ altitude by replacing the subscript of BrC with EC in Eq. (9).

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2.4 HYSPLIT ~~backward trajectory~~ model

~~The hybrid Single Particle Lagrangian Integrated Trajectory~~ (HYSPLIT) model developed by the National Oceanic and Atmospheric Administration's (NOAA) is a complete system for computing simple air parcel trajectories (Draxler et al., 2009). ~~It HYSPLIT is~~ ~~continues to be~~ one of the most extensively used atmospheric transport and dispersion models. A common application is ~~a back-trajectory trajectory~~ analysis to determine the origin of air masses and establish source-receptor relationships. In this study, ~~the~~ HYSPLIT model was used to determine ~~the~~ potential source regions of air pollutants ~~during~~ the four seasons at the six sites. The 96-h backward trajectories arriving at 500 m above ground level (~~AGL~~) and initializing ~~at~~ each hour of ~~the~~ day were calculated with ~~the~~ $0.25^{\circ} \times 0.25^{\circ}$ Global Data Assimilation System (~~GDAS~~) data from ~~the~~ National Centers for Environmental Prediction (~~NCEP~~). The gridded back-trajectory frequencies were calculated ~~using the~~ ~~with~~ Openair package ~~in of~~ Rplot.

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2.5 PMF receptor model

~~The~~ EPA PMF receptor model (version ~~5.0~~) is a mathematical approach for quantifying the contribution of sources to samples based on their composition or fingerprints ~~of the sources~~. A special ~~list~~ed data set can be viewed as a data matrix X of $i \times j$ dimensions, in which i number of samples and j chemical species were measured, with uncertainties u . The goal of ~~the~~ PMF model ~~was~~

to ~~solve-determine~~ the chemical mass balance between ~~the~~ measured species concentrations and source profiles, as shown in ~~the below~~-Eq. (10), with ~~the~~ number of factors p , ~~the~~ species profile f of each source, and ~~the~~ amount of mass g contributed by each factor to each sample:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij}, \quad (10)$$

- 5 where e_{ij} is the residual ~~of each sample or species for each sample/species~~. In this study, the uncertainties of the chemical species concentrations were estimated ~~by using the~~ Eq. (11):

$$Unc = \sqrt{(0.1 \times concentration)^2 + (0.5 \times MDL)^2}, \quad (11)$$

where MDL is ~~the~~ species-specific method detection limit. The water-soluble ions and carbonaceous aerosols in the four seasons at the six sites were used as input variables to run ~~the~~ PMF model. The

- 10 MDL of the species can refer to Cui et al. (2019).

3 Results and discussion

3.1 Light absorption of EC and BrC

- Table 2 summarizes ~~the~~ seasonally mean OC and EC concentrations, light absorption coefficients, ~~and~~ efficiency~~s~~ (b_{abs} , MAE) of EC and BrC at 405 nm, and the meteorological variables at the six sites
- 15 during the campaign. The average winter EC concentration ranges from 2.2 $\mu\text{g m}^{-3}$ at Maerkang to 7.9 $\mu\text{g m}^{-3}$ at Sanbacun, which is ~~a bout~~ 2–6 times higher than ~~that~~ ~~these~~ in the other seasons in response to more primary emissions in winter with similar wind speeds (Table 2). The much higher OC/EC ratios at the Plateau sites than ~~those that~~ at the Basin sites suggests that more secondary OC is formed by chemical reactions over the TP, which ~~has been corroborated by~~ ~~can be supported by the works of~~ Wu et
- 20 al. (2018). ~~H~~ ~~The higher~~ OC/EC ratios with ~~increasing the~~ altitude can also result from stronger EC emissions at lower altitudes. Combined with ~~those obtained from the~~ previous studies, the winter OC concentration ~~was~~ found to vary from 15.0 to 20.1 $\mu\text{g m}^{-3}$, whereas ~~the~~ EC ~~is ranged~~ between 4.3 and 4.7 $\mu\text{g m}^{-3}$ at urban areas inside the SCB, which ~~was~~ ~~substantially significantly~~ lower than that at ~~the Indo-Gangetic Plain~~ (Table S1). However, OC and EC concentrations ~~in at the~~ eastern TP ~~were~~ much
- 25 more abundant than ~~those at in at the~~ western and southern TP sites ~~because due to the~~ denser population and ~~widespread industrial activities~~ (Table S1). ~~Briefly Thus~~, carbonaceous aerosol pollution is much more severe inside the Basin than ~~that~~ over the TP, indicating that ~~the~~ large amounts

of air pollutants ~~are~~ trapped inside the deep Basin ~~because of due to~~ calm and stable air.

Figure 2 compares ~~the~~ spectral total and separated EC and BrC b_{abs} in spring and winter at ~~the~~ six sites along the ESTP. The measured (green hollow points) and calculated b_{abs} (yellow dashed ~~ed~~ lines) for total carbon (~~(TC₂₇ - sum of EC and BrC)~~ ~~were~~ comparable, and the difference ~~was~~ within 5%. For Sanbacun, a rural site inside the Basin, the b_{abs} is much higher than the other sites, especially for ~~the~~ shorter wavelengths, ~~because of due to~~ more BrC emissions from coal and biomass burning for cooking and heating ~~in~~ rural areas inside the SCB (Zhao et al., 2021). The light absorption of EC aerosols decreased ~~s~~ with altitude, primarily because of ~~the~~ ~~decreas~~ed EC concentration (see Table 2). ~~These~~ phenomenon may be partly ~~due to caused by the~~ stable air inside the deep Basin (Feng et al., 2020); ~~but however, it that~~ would also apply to BrC ~~as in so~~ far as EC and BrC share sources, and vertical mixing is primarily due to fair weather convection rather than deep convective storms (Zhang et al., 2017). However, ~~the~~ light absorption by BrC does not monotonically change ~~with~~ altitude ~~because due of to the~~ more complicated sources and origins of BrC. The 405 nm b_{abs} of BrC accounting for that of TC increased ~~s~~ from 20% at Chengdu to ~ 50% at Hongyuan, ~~whereas the~~ the proportion significantly reduced ~~s~~ with increased ~~ing~~ed wavelength (Figure S1), suggesting that light absorption of BrC aerosols is much stronger at high altitudes than ~~that~~ at lowlands.

Compared ~~to with~~ b_{abs} , MAE can better reflect ~~the LAE~~ light absorption efficiency of aerosols. The average winter MAE_{EC} is $6.0 \pm 1.0 \text{ m}^2 \text{ g}^{-1}$ among all sites, which is within the range of $3.9\text{--}11.9 \text{ m}^2 \text{ g}^{-1}$ over the TP and ~~the the~~ surrounding basins (Tables 2 and S1). Except our result in the rural site, the mean winter MAE_{BrC} of $0.7\text{--}0.8 \text{ m}^2 \text{ g}^{-1}$ inside the SCB is ~~approximately bout~~ half of that at ~~the Indo-Gangetic Plain (IGP)~~ probably ~~because of due to~~ the differences in BrC emissions, PM size distribution and chemical composition between ~~the~~ SCB and IGP (Choudhary et al., 2018). Figures 3 and S2 show box plots of spectral MAE_{BrC} and MAE_{EC} in the four seasons from the Basin to Plateau sites, extending elevation from 0.5 to 3.5 km. ~~In contrast to Different from~~ EC, MAE_{BrC} at 405 nm over the TP ~~was~~ 2–3 times higher than that inside the SCB with strong elevation-dependent ~~light absorption~~ light absorbing, and the only clear dependence ~~was~~ in winter. Wu et al. (2018) found that winter MAE_{BrC} ~~was~~ $4.5 \text{ m}^2 \text{ g}^{-1}$ for a pristine environment over the TP (Nam Co, 4730 m asl), which is significantly higher than that at Hongyuan (3500 asl) ~~in for~~ our study. The average winter OC/EC ratio of 14.1 at Nam Co ~~was~~

significantly largely higher than ~~that~~ at our sampling sites. Therefore, the clearly increased winter MAE_{BrC} with altitude may be related to BrC composition seasonally, ~~whereas~~ while winter MAE_{EC} decreases with altitude, possibly ~~because due of~~ the difference in source composition and ageing aerosols inside the deep Basin (Liu et al., 2020). This mechanism ~~is will be~~ discussed in the following sections.

Figure 4 shows MAE_{BrC} and MAE_{EC} variations as altitude in spring and winter during the campaign. The relationships between average MAE and altitude of the measurement sites were fitted by exponential function, and coefficients of determination (R^2) were given in the figure. R^2 reflects the strength of the relationships between two parameters. The contrasting MAE variation as altitude between BrC and EC in winter (R^2 of 0.89 for MAE_{BrC} and 0.86 for MAE_{EC}) is more significant than ~~that one~~ in spring (R^2 of 0.45 for MAE_{BrC} and 0.06 for MAE_{EC}). The better relationships in winter may be because more urban and aged aerosols are trapped inside the deep Basin in response to strong winter temperature inversion (Feng et al., 2020). The relation of MAE_{BrC} or MAE_{EC} at 405 nm with altitude can be parameterized with exponential function (Eq. 9). The spring and winter MAE_{BrC} can be parameterized with altitude (AT) as follows:

$$MAE_{405,BrC,spr} = 1.33 \cdot e^{0.18 \cdot AT} \quad (12)$$

$$MAE_{405,BrC,win} = 0.82 \cdot e^{0.33 \cdot AT} \quad (13)$$

Similarly, the winter MAE_{EC} can be parameterized by altitude (AT) as follows:

$$MAE_{405,EC,win} = 11.35 \cdot e^{-0.18 \cdot AT} \quad (14)$$

3.2 Sources impacting ~~on the~~ light absorption of EC and BrC

The OC/EC ratio can be used to ~~approximate~~ roughly identify sources of carbonaceous aerosols, and the ratio of aerosols from fossil ~~fuel~~ combustion is generally lower than that ~~from~~ biomass burning (Bond et al., 2004). Figure S3 shows the relationship between OC and EC concentrations inside the SCB and ~~that~~ over the TP during the campaign, and the OC/EC ratio was obtained by fitting the relationships with univariate linear regression. The significantly simultaneous change between OC and EC ($R^2 = 0.80$ for SCB, and $R^2 = 0.75$ for TP) indicated that the sources may be similar. The OC/EC ratios of 2.14 for western SCB and 2.06 for eastern TP are significantly lower than ~~those at~~ at Nam Co

(13.8–14.1, Wu et al., 2018) representing a pristine environment over central TP (Cong et al., 2009), whereas ~~the~~ the ratios are much higher than those ~~at~~ at Lhasa ~~—~~, the largest city of ~~the~~ ~~ver~~ TP (1.46, Li et al., 2016). The OC/EC ratios ~~for~~ ~~in~~ our study ~~are~~ ~~is~~ slightly lower than those ~~at~~ ~~at~~ ~~for~~ urban areas in eastern China and Helsinki in Finland (Han et al., 2014; Viidanoja et al., 2002), indicating that
5 carbonaceous aerosols ~~in~~ western SCB and eastern TP may be significantly affected by primary sources.

~~In addition to~~ ~~Besides~~ primary sources, secondary formation largely contributes to OC aerosols ~~;~~ ~~and~~ thus, secondary organic carbon (SOC) was calculated ~~using the~~ ~~with~~ EC-tracer method (Turpin and
10 Lim, 2001). To better understand ~~the~~ light absorption of primary ~~organic carbon (POC)~~ and ~~secondary~~ ~~SOC~~, Figures S4 and 5 show sample-to-sample and average MAE_{BrC} variations as SOC and POC concentrations for each site in spring and winter during the campaign, respectively. The ~~LAE~~ ~~light~~ ~~absorption efficiency~~ of BrC significantly declined ~~s~~ as the ~~increased~~ OC composition ~~increased~~ with ~~the~~ better relationships for POC at each site (Figure S4). The average winter MAE_{BrC} decreased by
15 ~~approximately~~ ~~about~~ 70% as POC increased ~~s~~ from 3.0 $\mu\text{g m}^{-3}$ at Hongyuan to ~~more~~ ~~higher~~ than 20 $\mu\text{g m}^{-3}$ at Chengdu (Figure 5). SOC accounting for OC significantly increased ~~s~~ from ~~the~~ western SCB to ~~the~~ eastern TP, and it ~~was~~ ~~is~~ higher than 50% at Maerkang and Hongyuan ~~because of~~ ~~due to~~ relatively fewer primary sources over the TP. The large winter MAE_{BrC} increment ~~in~~ ~~as~~ ~~the~~ ~~as~~ the SOC/POC ratio indicates that ~~the~~ more SOC and ~~the~~ fewer POC ~~are~~ ~~is~~ a favourable ~~condition~~ for BrC light absorption
20 enhancement (Figure 5). Therefore, the strong elevation-dependent MAE_{BrC} in winter (Figure 4) may be induced by SOC/POC ratio variations from ~~the~~ western SCB to ~~the~~ eastern TP.

The EC ~~LAE~~ ~~light absorption efficiency~~ largely ~~decreased as the~~ ~~reduces as~~ EC concentrations ~~s~~ increased ~~d~~ at each site (Figure S5). However, the average winter MAE_{EC} inside the highly polluted SCB
25 ~~is~~ ~~was~~ much higher than that over the clean TP, ~~whereas~~ ~~it~~ for similar EC concentrations among the Plateau sites, ~~the~~ MAE_{EC} at Wenchuan ~~is~~ ~~was~~ ~~approximately~~ ~~about~~ ~~two~~ ~~2~~ times higher than that at Hongyuan, with a strong dependence on elevation. Therefore, winter aerosol ageing ~~ing~~ inside the deep Basin and large source differences may induce light absorption reduction from ~~the~~ western SCB to ~~the~~ eastern TP. The increase ~~in~~ ~~of~~ MAE_{EC} as the ratios of water-~~s~~ soluble ions (K^+ , Cl^- , SO_4^{2-} , and NO_3^-) to
30 EC concentrations ~~at~~ ~~on~~ different levels suggests that EC light absorption ~~was affected~~ ~~is~~ ~~certainly~~

impacted by many anthropogenic sources at the six sites (Figure S6). A specific inorganic component can be considered as an indicator of the specific emission sources. K^+ and Cl^- ions are usually used to characterise biomass burning (BB) and coal combustion (CC), respectively (Tao et al., 2016). NO_3^- and SO_4^{2-} reflect motor vehicle and industry source impacts, respectively.

5 Therefore, to further identify the key sources impacting MAE_{EC} , we checked the spring and winter mean MAE_{EC} variations as the concentrations of K^+ , Cl^- , NO_3^- and SO_4^{2-} ions at the six sites (Figure 6). Compared with the spring value, the winter MAE_{EC} was lower owing to high EC concentrations and was more sensitive to the chemical species from anthropogenic emissions. Furthermore, the NO_3^- difference among the sites (Figure 6a) was much larger than that of K^+ , Cl^- and SO_4^{2-} because of the combustion of fossil fuels many fossil fuel combustion at the Chengyu City Clusters inside the Basin. The spatial heterogeneity in the $(NO_3^- + SO_4^{2-}) / (K^+ + Cl^- + NO_3^- + SO_4^{2-})$ ratio in winter was more significant than that in spring, and winter MAE_{EC} evidently increased as the ratio from the TP to the Basin sites. Therefore, the emissions from fossil fuel combustion may be a key source influencing winter MAE_{EC} .

15

The above paragraphs separately analysed the LAE of BrC and EC and their variations as chemical species, while the change in radiative forcing of BrC relative to EC (f) from Chengdu to Hongyuan is shown in Figure 7a to reveal the mechanism. The parameter (f) reflects the light absorption strength of BrC at the shorter wavelengths as compared to that of EC aerosols at all the whole wavelengths. The much higher f values indicated that the radiative forcing of BrC aerosols is much stronger for the similar EC radiative forcing; and thus, this parameter can be used to better understand the radiative forcing of secondary aerosols relative to primary aerosols at a specific location. The altitude (AT) increased by 3 km, and while the median f increased from approximately 0.10 inside the Basin to 0.42 over the eastern TP. The relationship between f and altitude can be parameterized as follows the below equation:

25

$$f = 0.077 \cdot e^{0.480 \cdot AT} \quad (15)$$

Some a few studies have found that the direct radiative forcing of BrC/(BrC+EC) increases with altitude simply because due to the fact that the concentration of BrC decreases more slowly with altitude than that of EC (Liu et al., 2014, 2015; Zeng et al., 2020; Zhang et al., 2017).

Therefore, we also checked the median OC/EC ratio variations from the Basin to ~~the~~ Plateau sites during the campaign (Figure 7b). The OC/EC ratio ~~change~~^s within the range of 2–4, and the 75th percentiles of the ratio ~~increase~~^s more significantly than the median values from the Basin to Plateau sites. Therefore, the increased *f* from ~~the~~ western SCB to ~~the~~ eastern TP may be closely related to more
5 secondary formation and fewer primary emissions over the TP than ~~over the~~ SCB (also see Figure 5c).

As previously mentioned, ~~the~~ MAE of carbonaceous aerosols largely depends on emission sources. ~~The~~ PMF receptor model is widely used to apportion the sources influencing air pollutants at a specific site based on the fingerprints of the sources; for example, K^+ and Cl^- are usually used as tracers for
10 ~~BB~~ biomass burning (BB) and ~~CC~~ coal combustion (CC), respectively (Tao et al., 2016). ~~The~~ PMF analysis was conducted in this study for each season. ~~The~~ motor vehicles, biomass and coal burning, dust, sea salt, and secondary formations ~~we~~ are found to be the main sources at the six sites. Figure 8 shows mass concentrations of species for each source at each site apportioned ~~using the~~ PMF model in winter during the campaign. The PMF results for the other seasons are ~~shown~~ illustrated in Figures
15 S7–S9. The winter NO_3^- concentrations for secondary nitrate ~~decrease~~^d from $3.44 \mu g m^{-3}$ ~~in~~ at Sanbacun to $0.07 \mu g m^{-3}$ ~~in~~ at Maerkang, which is more heterogeneous than that in summer and fall. As a main source inside the SCB, the winter secondary nitrate is in response to the intensive mixing between ~~the~~ motor vehicle emissions and other primary pollutants trapped inside the Basin by strong capping inversion (Feng et al., 2020). Additionally, high humidity inside the SCB facilitates ~~the~~ secondary nitrate
20 formation, and the average nitrogen oxidation ratio in Sichuan (average RH = 80%) is 3.1 times of that in winter ~~in~~ Beijing (average RH = 27%) (Wang et al., 2021). EC aerosols from ~~the~~ intensive human activities inside the SCB are easily aged by coating the secondary-~~formed~~ nitrate in winter, which further ~~causes the~~ enhancement of ~~the~~ Basin EC light absorption. The latest ~~studies~~ study by ~~of~~ Zhang et al. (2022) found that ~~the~~ light absorption and radiative forcing of ~~BC~~ black carbon coated ~~with~~ by
25 inorganic salts are much stronger than ~~of~~ that inside organic materials. The chemical species (K^+ , Cl^-) from ~~BB~~ biomass burning and ~~CC~~ coal combustion ~~decline~~^d from the Basin to Plateau sites; ~~however,~~ ~~but~~ the declining ranges in ~~the~~ warm seasons (summer and fall) ~~were~~ ~~are~~ more significant than those in the cold seasons (spring and winter) ~~because of the~~ ~~used~~ ~~to~~ ~~usage~~ of more fuel for heating over the TP. Therefore, the primary BrC from ~~BB~~ biomass burning and ~~CC~~ coal combustion for winter heating
30 over the TP may partly contribute to ~~the~~ strong TP BrC light absorption.

3.3 Impacts of regional and long-range transport on the light absorption of aerosols

The fresh aerosol particles are gradually aged by mixing with other pollutants during the long-range transport, and thereby enhance their light absorption and radiative forcing. The similarities of the major chemical species between the two sites should represent regional air pollution, whereas the differences should reflect local source impacts. The comparisons between Basin (Chengdu, Sanbacun) and Plateau sites (Wenchuan, Lixian, Maerkang, Hongyuan) related to the average mass concentrations of water-soluble ions and carbonaceous species in the four seasons are shown in Figures 9 and S10–S12. The numerical ranges between the two axes of each subplot were set to be equal to more clearly observe the spatial heterogeneity of the chemical species in the region. The combination of the CV and correlation coefficients can be used to better understand intra-location variability (Wilson et al., 2005). The CV was between 0 and 1 (see Eq. 1), and the smaller value represents the more uniform particle concentrations. The moderate differences and relatively high CV values (0.22–0.75) were observed for the chemical species from anthropogenic sources (NH_4^+ , K^+ , SO_4^{2-} , NO_3^- , F^- , Cl^- , OC, and EC) in the four seasons. The differences indicated that there were limited similarities between the Basin and Plateau sites, and the discrepancies were in major anthropogenic sources. The spatial heterogeneity of K^+ and NO_3^- is more obvious than that of the other species in the four seasons, which is mainly related to more biomass burning and vehicle emissions inside the SCB (Zhao et al., 2021). The weak inter-regional transport between the western SCB and eastern TP suggested that the light absorption of carbonaceous aerosols over the TP is rarely influenced by pollutants from the SCB. Furthermore, the CV values for K^+ , NO_3^- , and EC in winter were the lowest among the seasons because of increased biomass burning and coal combustion for winter heating over the Tibetan Plateau. Unlike CV, a high correlation coefficient for a specific chemical component does not necessarily indicate uniformity, which may suggest source similarity between sites. The correlation largely depended on the season (Figures 9 and S10–S12). The strong correlations for NH_4^+ , K^+ , SO_4^{2-} , NO_3^- , OC, and EC in the spring and winter implied that Basin and Plateau sites shared similar sources for the species, whereas weak correlations for NO_3^- , OC, and EC in summer and fall indicated impacts of dissimilar sources between the Sichuan SCB and Tibetan Plateau.

Compared ~~to with~~ the species from anthropogenic sources, the lowest CV values for Na⁺, Mg²⁺, and Ca²⁺ among the species indicated that they ~~we are~~ more comparable between ~~the~~ Basin and Plateau sites. Furthermore, changes in Na⁺ values ~~we are~~ more synchronous than ~~those of~~ Mg²⁺ and Ca²⁺ in ~~the~~ summer and fall. Na⁺ concentrations ~~were is~~ found to be high in salt-rich dust from saline soils (Quick and Chadwick, 2011). Dust events frequently occurred in spring and winter over ~~the Tibetan Plateau~~ and northwest China, where saline and alkaline land and dried salt-lakes ~~are~~ located (Jiang et al., 2021; Zhang et al., 2009; Zhang et al., 2021), ~~and thus~~, the weak correlations for Na⁺, Mg²⁺, and Ca²⁺ values in spring and winter may suggest local and regional dust plume impacts. Therefore, the lack of considering mineral dust impacts in separation of BrC light absorption from total aerosol absorption (Eq. 5) might cause some errors. The errors should be much smaller ~~as than those~~ ~~of compared to~~ the studies ~~on at the~~ north or northeast TP close to ~~the~~ Taklimakan and Gobi Deserts.

MODIS active fire data suggests that ~~BB biomass burning~~ is ~~mainly primarily~~ ~~le conducted~~ in South Asia around our study regions, which ~~was~~ more ~~abundant-frequent~~ in cold than warm seasons during our campaign (Figure S13). The PM mass concentrations in conjunction with wind data can be used to identify the local PM origins, ~~and~~ Figure 10 shows ~~that~~ K⁺ pollution ~~increased~~ ~~rose~~ in the four seasons at the six sites. The back-trajectory calculation can ~~provide give~~ PM origins from long-range transport, ~~and~~ Figures 11 and S14–S16 illustrate the gridded back-trajectory frequencies in the four seasons. K⁺ stratification in warm seasons ~~is was~~ more ~~obvious-evident~~ than that in cold seasons, ~~which~~ ~~implying infers~~ that there ~~were are~~ ~~substantial more~~ ~~BB biomass burning~~ plumes over ~~the Tibetan Plateau~~ in spring and winter. The change in wind direction ~~is not obvious~~ from ~~the Sichuan-SC~~ Basin to ~~the TP~~ ~~Tibetan Plateau~~ during warm seasons ~~is not evident~~. However, the predominant wind direction is northwest–southeast in cold seasons for the Basin sites, ~~whereas while~~ ~~it that~~ ~~mainly primarily~~ focusses on ~~the~~ southwest for the Plateau sites (Figure 10). The highest frequency of ~~the~~ back trajectory ~~was~~ also ~~is~~ in ~~the~~ southwest of the sampling sites in winter (Figure 11). Therefore, the ~~BB biomass burning~~ emissions originat~~ing ed~~ from South Asia are transported to ~~the~~ eastern ~~Tibetan Plateau~~ by highly frequent southwesterly winds, ~~and~~ thus induc~~ing e~~ high K⁺ concentrations in spring and winter. ~~The~~ BrC aerosols from ~~the~~ intensive ~~BB biomass burning~~ in South Asia are gradually aged by internal or external mixing with ~~the~~ other anthropogenic emissions during ~~the~~ long-range transport. The light absorption of ~~the~~ aged BrC aerosols over the TP is enhanced by coating ~~with e~~ inorganic components (Zhang et al.,

2022), which may partly contribute to ~~the~~ stronger BrC light absorption at the Plateau sites than ~~that at~~ the Basin sites. Unlike ~~the~~ eastern TP, the carbonaceous aerosols in ~~the~~ western SCB are regionally transported from ~~the~~ central and eastern SCB, ~~as which~~ can be seen from ~~the increase in~~ pollution ~~rose-~~ and back trajectories. The aerosols accumulate and stagnate at the front areas of the mountains ~~because~~
 5 ~~of due to the~~ terrain block~~s~~; ~~and thus,~~ ~~the~~ light absorption ~~of by~~ EC aerosols emitted from motor vehicles is enhanced by the intensive mixing among the air pollutants.

4 Summary and conclusions

Tibetan Plateau (~~TP~~) is surrounded by the three highly polluted regions, i.e., ~~Indo-Gangetic Plain (IGP)~~, Taklimakan and Gobi Deserts (TGDs) and ~~SiChuan Basin (SCB)~~. However, ~~the~~ previous studies ~~have~~
 10 ~~mainly primarily~~ focussed on the south (IGP) and north slopes (TGDs); ~~and thus~~ ~~†~~The first *in-situ* aerosol measurements were conducted ~~at on the eastern slope of Tibetan Plateau (ESTP)~~ to study the elevation-dependent light absorption ~~of by~~ carbonaceous aerosols from the highly polluted SCB to the pristine TP. The source and origin impacts on light absorption ~~of by~~ aerosols ~~also~~ were ~~also~~ discussed by combining with PMF and HYSPLIT results.

The EC and BrC light absorptions ~~were was~~ separated ~~using by at the~~ simple two-component model. The BrC light absorption coefficient at 405 nm, accounting for that of ~~total carbon (TC;~~ (sum of EC and BrC), ~~was is~~ found to increase from ~ 20% inside the SCB to ~ 50% over the TP. The BrC ~~mass-~~ ~~absorption efficiency (MAE)~~ over ~~the~~ eastern TP ~~was~~ 2–3 times higher than that inside the SCB, with
 20 strong elevation-dependent absorption. The most significant elevation-dependent winter MAE_{BrC} ~~was is~~ closely related to the high ratio of ~~SOC to POC~~ ~~secondary to primary organic carbon (OC)~~, ~~that is i.e.~~, more OC from secondary formation than ~~those from~~ primary emissions at high altitudes. ~~In contrast~~ ~~to~~ ~~Different from~~ BrC, winter MAE_{EC} declined ~~sd~~ from the highly polluted SCB to ~~the~~ clean TP, ~~which is~~ ~~because of due to~~ source differences between the two regions. ~~Substantial More~~ urban ~~sources-~~
 25 ~~emissions~~ (vehicles, industries, etc.) ~~are were~~ trapped inside the deep SCB ~~owing due~~ to poor dispersion and frequent temperature inversion ~~during~~ cold seasons. ~~H~~The high primary emissions and weak dispersion conditions ~~were are~~ favourable for mixing and aerosol ageing ~~ing~~ to enhance ~~the~~ light absorption inside the Basin. The median radiative forcing of BrC relative to EC increased ~~sd~~ from ~~approximately about~~ 0.10 inside the Basin to 0.42 over ~~the~~ eastern TP, which ~~was is~~ associated with ~~the~~

OC/EC ratio. Therefore, the enhanced radiative forcing of BrC relative to EC ~~occurred~~ because OC
~~the concentration of OC decreases~~ more slowly with altitude ~~than the EC concentration~~.

5 The first aerosol field experiment was conducted ~~in at the~~ specific study region; ~~however, but~~ only six
sampling sites ~~were used~~ from the deep SCB to ~~the~~ eastern TP ~~were used~~ in this study. ~~M~~The more
measurement sites ~~should~~ be established to better understand the chemical composition and light
properties of aerosols ~~in at the~~ unique region. The light absorption coefficients and efficiencies of BrC
~~could not failed to~~ be separated from ~~those~~ of TC in summer and fall at Maerkang and Hongyuan
~~because of due to~~ instrument failure, which limited ~~the revelation or reveal of the~~ elevation-dependent
10 light absorption. Furthermore, replacing BrC, OC mass concentration was used to estimate MAE_{BrC} ,
which may cause ~~significant large~~ uncertainty; ~~and thus~~ these are expected to be corrected in ~~the~~
future studies.

Data availability. Raw data sets (Zhao et al., 2022, DOI: 10.5281/zenodo.6474199) used in this manuscript were
15 available at https://zenodo.org/record/6474199#.YmCn_YtByUk.

Author contributions. Suping Zhao and Ye Yu designed the study. Suping Zhao analyzed the data with help from
Ye Yu, Jinbei Chen and Shichang Kang. Daiying Yin and Longxiang Dong collected and analyzed data during the
campaign. Shaofeng Qi conducted the field experiment.

20

Competing interests. The authors declare that they have no conflict of interest.

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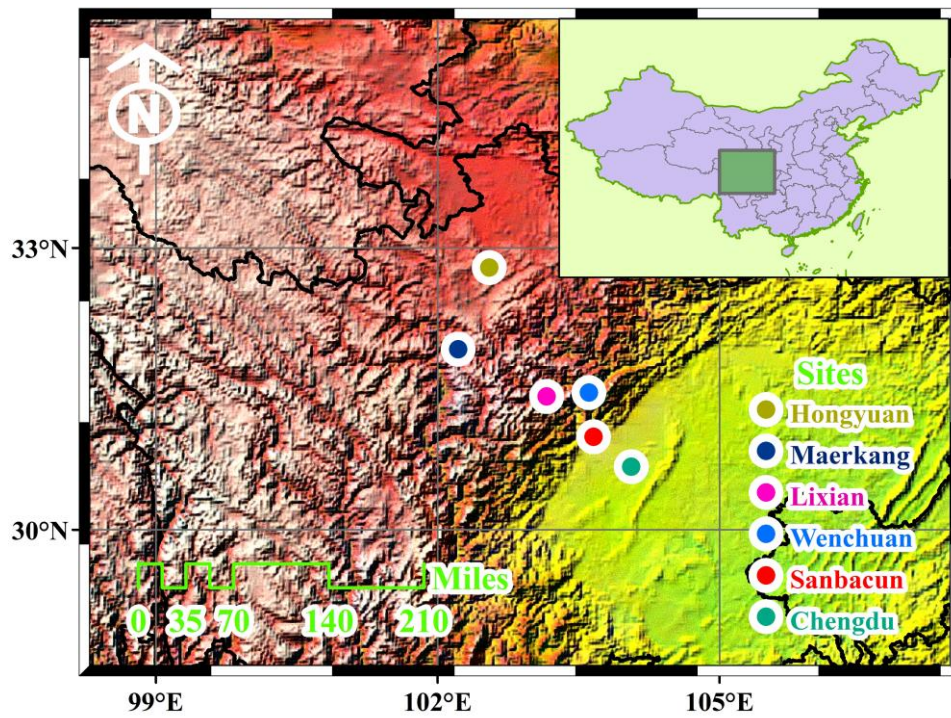


Figure 1: Geographic location of the six *in-situ* measurement sites (Chengdu, Sanbacun, Wenchuan, Lixian, Maerkang, and Hongyuan) along the **eastern slope of Tibetan Plateau**ESTP. The map is a pure reproduction of Google Maps with added a marks for our study locations.

5 Copyright © Google Maps.

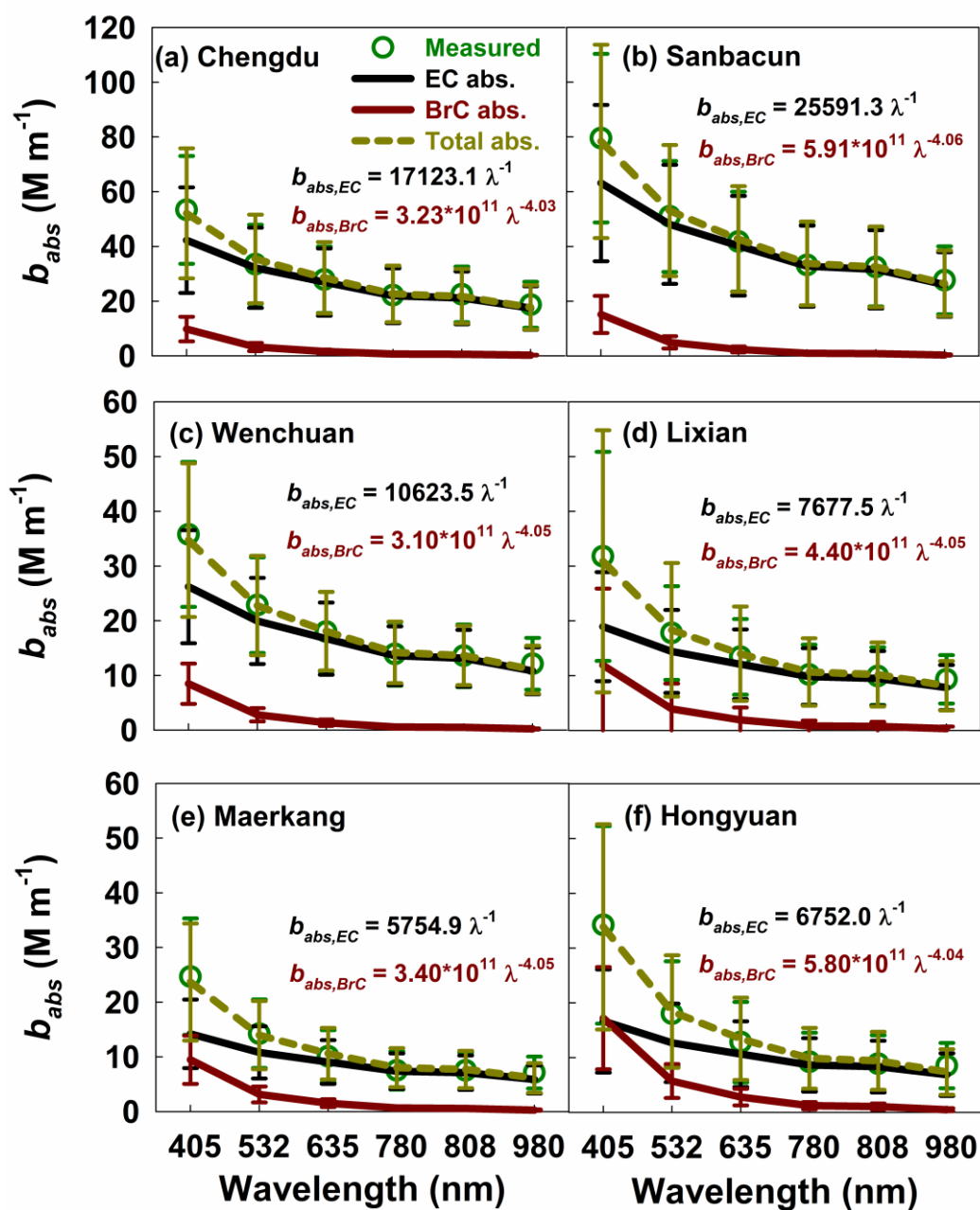


Figure 2: Spectral light absorption coefficients (b_{abs}) by EC and BrC in spring and winter at the six sites along the ESTP. The subplots depict the decomposition of total light absorption by EC and BrC with the model given in Eq. 4. Error bars represent uncertainties derived from replicate

5 analyses and lower quantifiable limits.

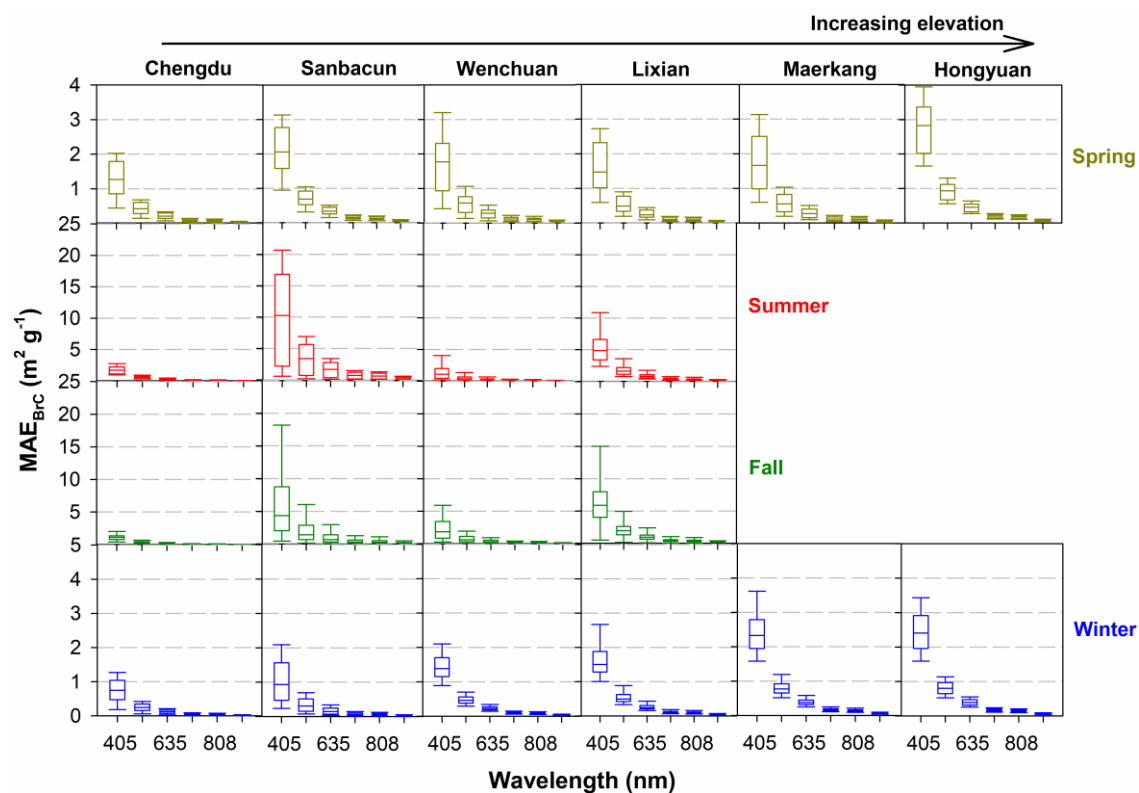


Figure 3: Box plots of spectral **BrC**-mass absorption efficiency of **BrC** (MAE_{BrC}) in each season from Chengdu inside the SCB to Hongyuan over the TP extending ranging in elevation from 500 m to 3500 m. The lines inside the boxes denote the median values, and the two whiskers and the top and bottom of the boxes denote the 5th and 95th and the 75th and 25th percentiles.

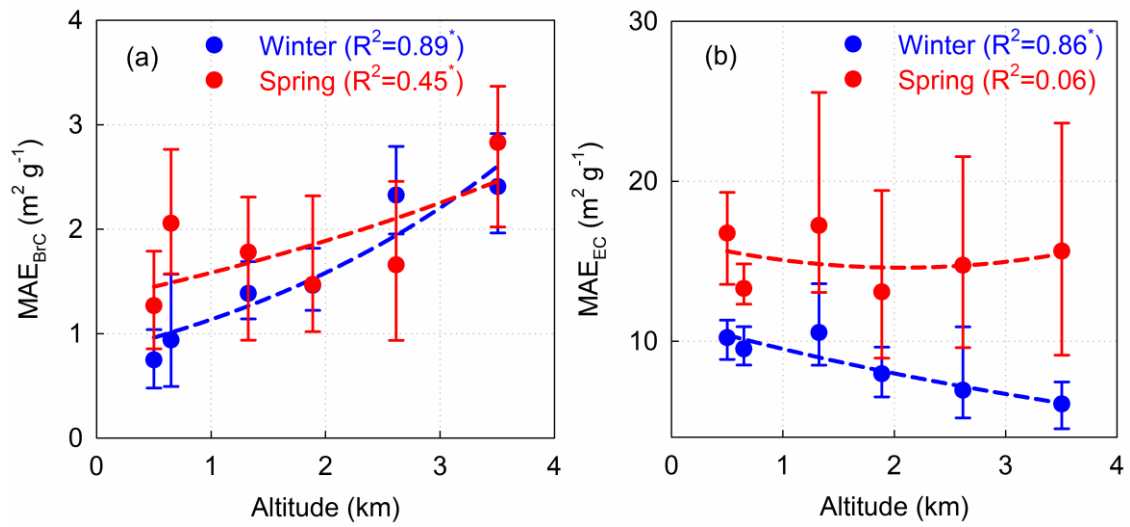


Figure 4: Variations of (a) MAE_{BrC} and (b) MAE_{EC} at 405 nm as altitude in spring and winter during the field campaign. The solid dots denote the median values, and the two whiskers of the dots denote the 25th and 75th percentiles. The relationships between averaged MAE and altitude of the measurement sites were fitted by exponential function, and the coefficients of determination (R^2) also were given in each subplot. The relationships (R^2 with the superscript of an asterisk) passed the significance level of 0.01.

Spring ○ Chengdu ○ Sanbacun ○ Wenchuan ○ Lixian ○ Maerkang ○ Hongyuan
 Winter ● Chengdu ● Sanbacun ● Wenchuan ● Lixian ● Maerkang ● Hongyuan

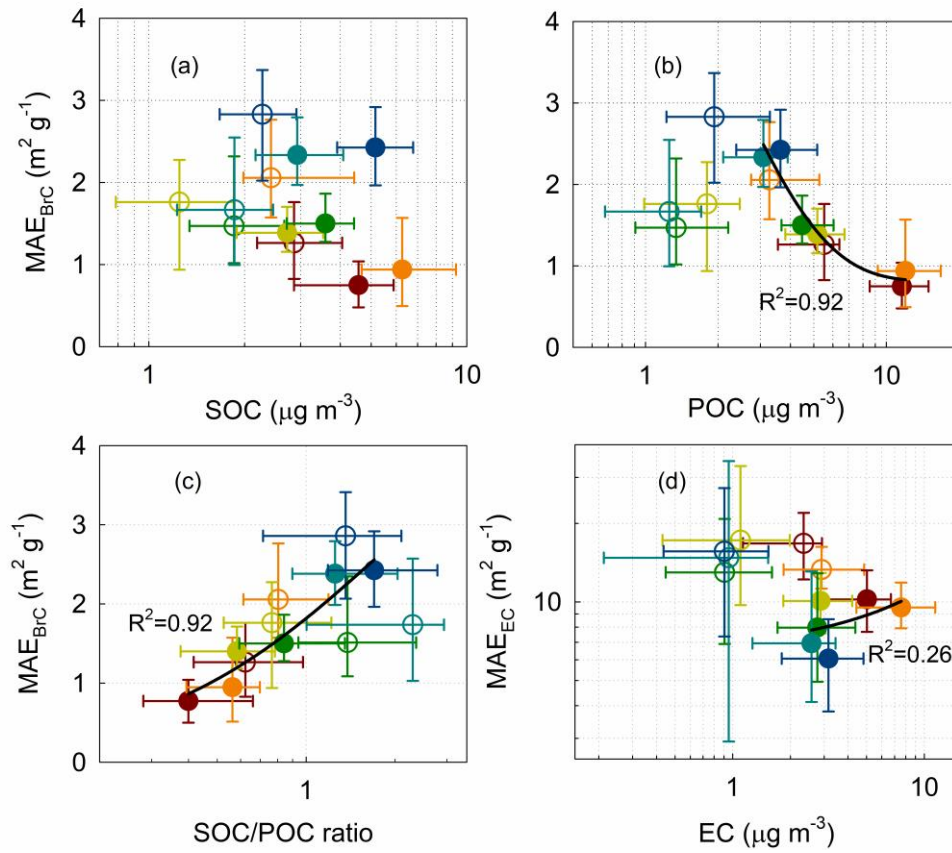


Figure 5: Variations of spring and winter mean-averaged MAE_{BrC} in spring and winter as (a) SOC, (b) POC, (c) SOC/POC ratio and (d) MAE_{EC} as EC concentrations at the six sites. The hollow and solid dots denote the median values in spring and winter, and the four whiskers of the dots denote the 25th and 75th percentiles of the corresponding two variables. The horizontal axis in each subplot is showed on a logarithmic scale to more clearly see the details.

Spring ○ Chengdu ○ Sanbacun ○ Wenchuan ○ Lixian ○ Maerkang ○ Hongyuan
 Winter ● Chengdu ● Sanbacun ● Wenchuan ● Lixian ● Maerkang ● Hongyuan

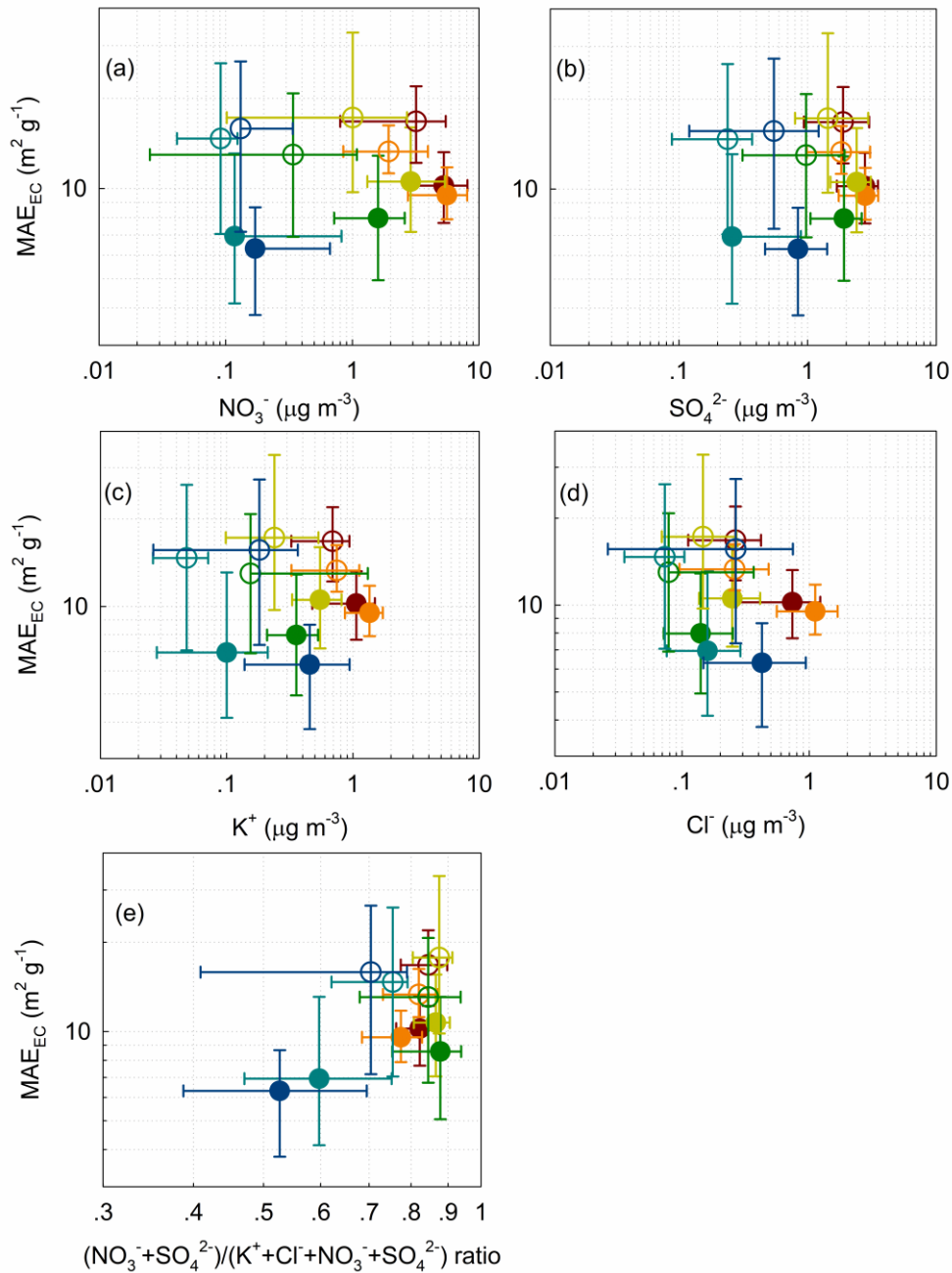


Figure 6: Variations of spring and winter-MAE_{EC} in spring and winter as (a) NO₃⁻, (b) SO₄²⁻, (c) K⁺, and (d) Cl⁻ concentrations, and (e) (NO₃⁻+ SO₄²⁻) / (K⁺+ Cl⁻+ NO₃⁻+ SO₄²⁻) ratio at the six sites. The hollow and solid dots denote the median values in spring and winter, and the four whiskers of the dots denote the 25th and 75th percentiles of the corresponding two variables. The axes in each subplot are showed on a logarithmic scale to more clearly see the details.

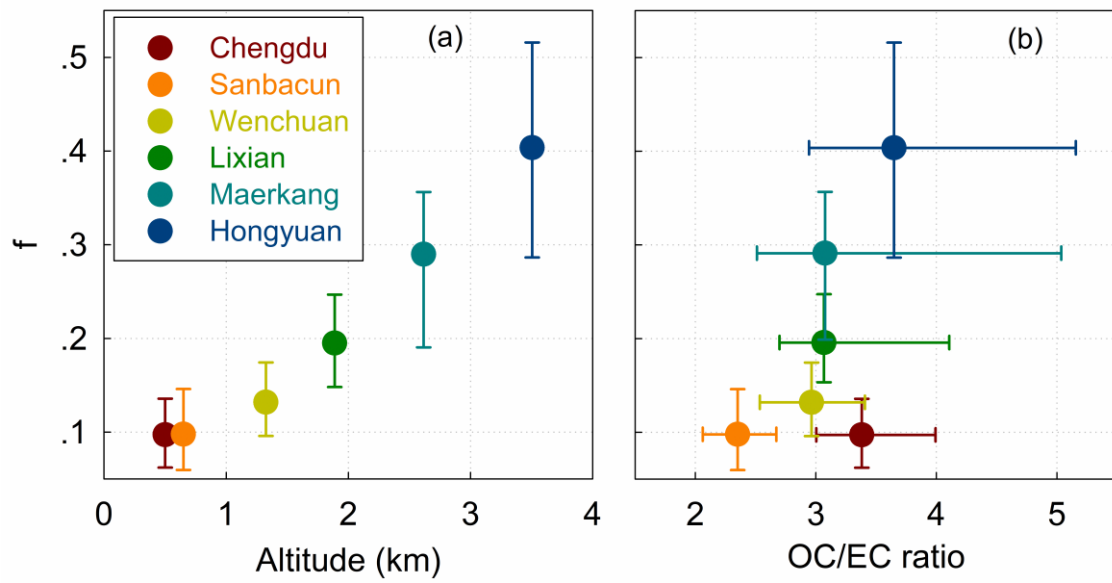


Figure 7: Variation of radiative forcing of BrC relative to EC (f , see Eq. 8) as (a) altitude and (b) OC/EC ratio for each site. The solid dots denote the median values, and the two whiskers of the dots denote the 25th and 75th percentiles of the variables.

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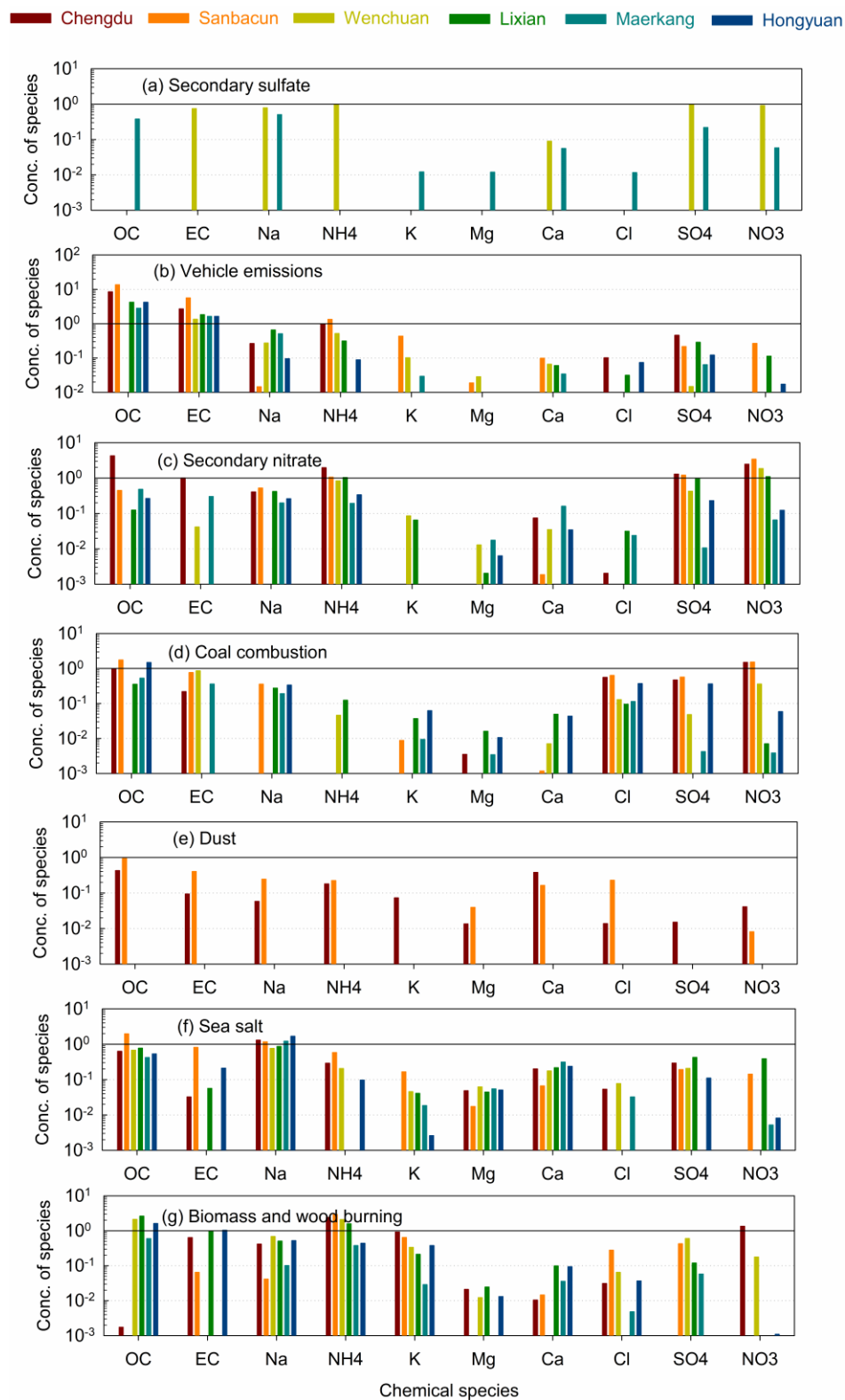


Figure 8: Mass concentrations of species for each source at each site apportioned by PMF model in winter during the campaign. The vertical axes are showed on logarithmic scale to better distinguish the concentrations of chemical species among the measurement sites.

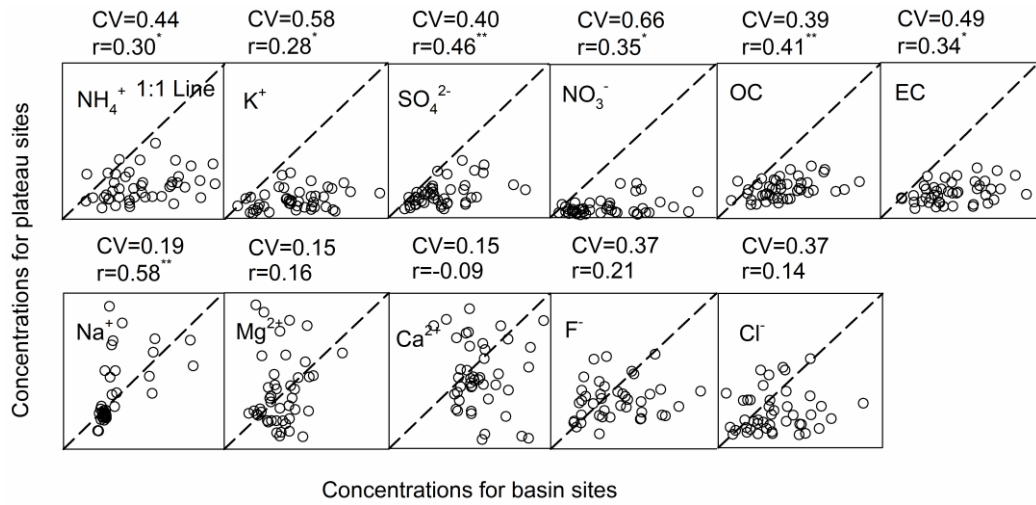


Figure 9: Relationships of **spring** PM_1 chemical components concentrations **in spring** between basin (horizontal axes, including Chengdu and Sanbacun) and plateau sites (vertical axes, including Wenchuan, Lixian, Maerkang and Hongyuan). The correlation coefficients (r) with an asterisk and two asterisk superscripts passed the significance level of 0.05 and 0.01, respectively.

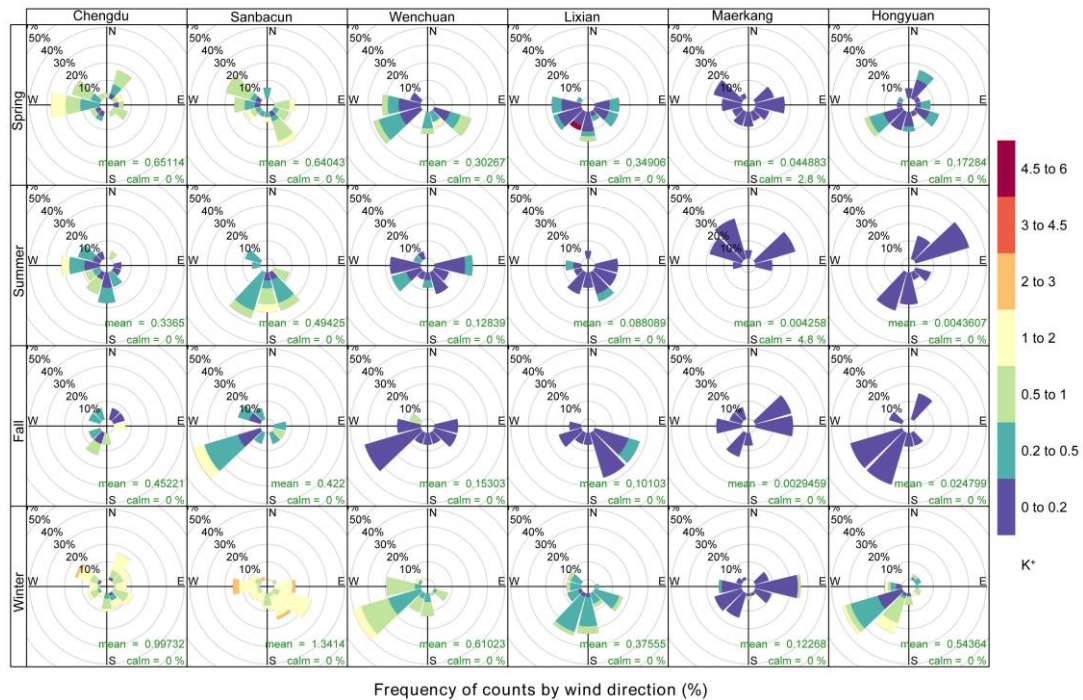


Figure 10: K^+ pollution rose in the four seasons at the six sites along the ESTP. Mean K^+ concentrations and calm frequencies also were given in each subplot.

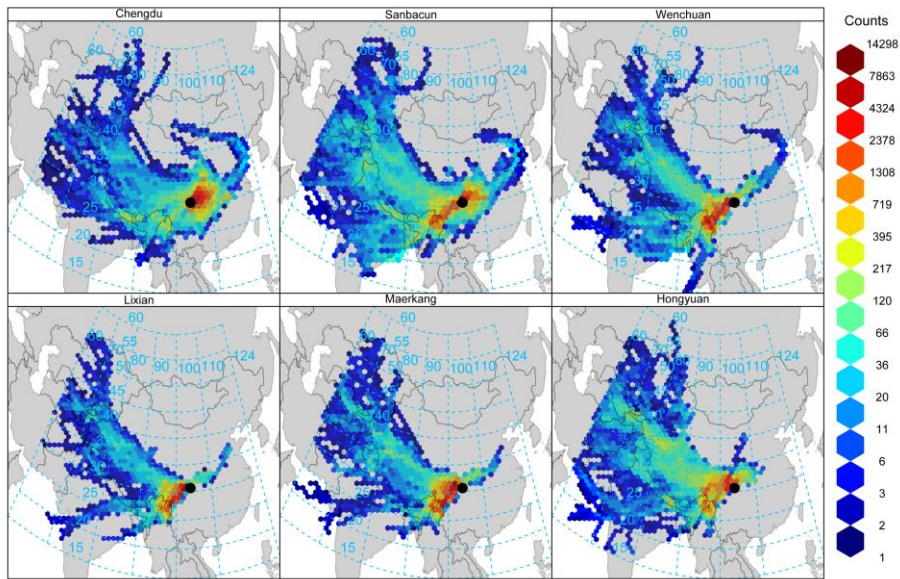


Figure 11: Gridded back trajectory frequencies with hexagonal binning in winter at the six sites from west Sichuan Basin to Tibetan Plateau. The map is a pure reproduction of Google Maps with added the trajectory frequencies. Copyright © Google Maps.

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Table 1 Summary of the measurement sites (name, location and altitude).

Name	Latitude (degree)	Longitude (degree)	Altitude (km)
Chengdu	30.67	104.06	0.50
Sanbacun	30.99	103.66	0.65
Wenchuan	31.46	103.61	1.33
Lixian	31.42	103.16	1.89
Maerkang	31.92	102.22	2.62
Hongyuan	32.79	102.55	3.50

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Table 2 Seasonally averaged values (mean \pm std.) of OC and EC concentrations, light absorption coefficient (b_{abs}), mass absorption efficiency (MAE) and meteorological variables (wind speed (WS), temperature (Tem.), relative humidity (RH)) at the six sites during the measurement campaign. There is no b_{abs} or MAE reported for MEK and HY in summer and fall as the used DRI instrument does not work at the 2 wavelengths of 405 nm and 445 nm when the samples are measured, and thus separation of EC and BrC cannot be conducted by Eq. (5). Chengdu, Sanbacun, Wenchuan, Lixian, Maerkang and Hongyuan are abbreviated as CD, SBC, WC, LX, MEK and HY, respectively.

Season	Sites	OC ($\mu\text{g m}^{-3}$)	EC ($\mu\text{g m}^{-3}$)	b_{abs} (M m^{-1})		MAE ($\text{m}^2 \text{g}^{-1}$)		WS (m s^{-1})	Tem. ($^{\circ}\text{C}$)	RH (%)
				BrC, 405	EC, 405	BrC, 405	EC, 405			
Spring	CD	7.9 \pm 3.7	2.2 \pm 1.2	8.5 \pm 2.8	32.4 \pm 12.7	1.3 \pm 0.6	17.1 \pm 4.8	1.6 \pm 0.7	17.5 \pm 4.3	80.3 \pm 19.9
	SBC	6.7 \pm 3.0	3.3 \pm 1.5	13.7 \pm 5.6	44.2 \pm 16.9	2.1 \pm 0.9	13.7 \pm 2.5	1.4 \pm 0.6	16.9 \pm 4.1	77.6 \pm 15.9
	WC	3.2 \pm 1.6	1.2 \pm 0.8	4.8 \pm 2.3	20.2 \pm 9.0	1.6 \pm 0.9	21.5 \pm 11.6	2.4 \pm 1.0	15.1 \pm 4.4	65.1 \pm 17.4
	LX	3.5 \pm 1.4	1.0 \pm 0.6	5.4 \pm 2.5	11.8 \pm 5.5	1.7 \pm 0.8	13.8 \pm 6.9	1.6 \pm 0.5	13.3 \pm 5.3	61.5 \pm 20.4
	MEK	3.0 \pm 1.7	0.8 \pm 0.6	4.8 \pm 2.7	10.2 \pm 3.6	1.9 \pm 1.2	16.6 \pm 9.4	1.1 \pm 0.6	10.6 \pm 5.5	62.0 \pm 26.5
	HY	4.1 \pm 1.6	0.9 \pm 0.6	11.5 \pm 4.9	12.9 \pm 6.2	2.8 \pm 0.9	17.3 \pm 9.8	2.4 \pm 1.0	2.4 \pm 3.6	70.0 \pm 16.6
Summer	CD	5.4 \pm 1.2	1.9 \pm 0.5	9.0 \pm 2.7	29.2 \pm 6.9	1.8 \pm 0.6	16.4 \pm 4.5	1.3 \pm 0.4	25.2 \pm 2.9	84.6 \pm 18.8
	SBC	2.9 \pm 1.2	1.5 \pm 0.7	21.8 \pm 15.0	32.6 \pm 7.9	10.1 \pm 7.1	29.8 \pm 6.5	1.1 \pm 0.4	24.1 \pm 3.0	82.7 \pm 13.9
	WC	2.2 \pm 0.8	1.0 \pm 0.5	2.2 \pm 1.5	18.9 \pm 5.5	1.4 \pm 1.3	23.5 \pm 9.5	1.7 \pm 0.7	23.1 \pm 3.2	64.5 \pm 16.5
	LX	2.7 \pm 0.9	0.8 \pm 0.5	13.3 \pm 5.0	9.5 \pm 2.7	5.4 \pm 2.5	16.4 \pm 11.3	1.4 \pm 0.5	20.9 \pm 4.0	65.2 \pm 18.0
	MEK	2.7 \pm 1.5	0.7 \pm 0.6	—	—	—	—	1.0 \pm 0.4	16.6 \pm 4.3	73.3 \pm 22.6
	HY	3.0 \pm 1.2	0.8 \pm 0.6	—	—	—	—	1.8 \pm 0.6	10.1 \pm 3.3	77.8 \pm 11.6
Fall	CD	4.7 \pm 1.3	2.3 \pm 1.0	5.3 \pm 2.5	40.6 \pm 16.6	1.1 \pm 0.5	18.3 \pm 4.0	1.1 \pm 0.4	15.6 \pm 4.9	88.4 \pm 10.8
	SBC	5.3 \pm 3.4	3.0 \pm 1.8	22.0 \pm 13.7	50.8 \pm 11.2	6.0 \pm 5.6	24.3 \pm 9.3	0.9 \pm 0.2	14.9 \pm 4.4	89.9 \pm 11.6
	WC	1.6 \pm 0.8	0.8 \pm 0.5	3.0 \pm 2.0	18.2 \pm 7.3	2.3 \pm 1.8	27.3 \pm 13.9	1.7 \pm 0.6	14.1 \pm 5.4	72.7 \pm 10.0
	LX	2.4 \pm 1.0	0.9 \pm 0.5	12.7 \pm 6.6	10.5 \pm 3.4	6.5 \pm 3.8	14.7 \pm 10.1	1.3 \pm 0.3	11.9 \pm 5.6	76.8 \pm 11.3
	MEK	2.3 \pm 1.2	0.9 \pm 0.6	—	—	—	—	0.9 \pm 0.4	8.8 \pm 5.5	78.4 \pm 17.0
	HY	3.4 \pm 2.2	1.3 \pm 1.1	—	—	—	—	1.9 \pm 0.7	0.7 \pm 5.6	73.9 \pm 11.0
Winter	CD	15.0 \pm 5.9	4.7 \pm 2.0	10.5 \pm 4.6	47.6 \pm 20.1	0.8 \pm 0.5	10.4 \pm 2.8	1.2 \pm 0.4	6.6 \pm 2.7	78.9 \pm 16.9
	SBC	18.9 \pm 7.6	7.9 \pm 3.4	17.1 \pm 10.2	74.7 \pm 27.9	1.2 \pm 1.0	9.9 \pm 2.0	1.0 \pm 0.3	5.8 \pm 2.7	79.2 \pm 15.0
	WC	8.2 \pm 3.1	2.8 \pm 1.3	11.2 \pm 3.2	29.7 \pm 9.5	1.5 \pm 0.5	11.6 \pm 4.4	1.9 \pm 0.6	3.6 \pm 2.4	60.2 \pm 9.0
	LX	8.4 \pm 2.7	3.0 \pm 1.3	17.1 \pm 15.4	24.3 \pm 9.1	2.2 \pm 2.6	8.9 \pm 3.9	1.4 \pm 0.4	-0.1 \pm 2.1	62.4 \pm 10.3
	MEK	5.3 \pm 2.3	2.2 \pm 1.1	13.2 \pm 4.0	16.6 \pm 6.3	2.5 \pm 0.9	8.6 \pm 4.4	1.1 \pm 0.3	-0.2 \pm 3.7	36.1 \pm 11.0
	HY	8.4 \pm 3.8	3.0 \pm 1.6	21.5 \pm 11.3	18.9 \pm 10.2	2.5 \pm 0.7	6.7 \pm 4.9	2.1 \pm 1.5	-6.5 \pm 6.8	42.8 \pm 21.8