

Supplementary Information for

Combined influences of sources and atmospheric bleaching on light absorption of water-soluble brown carbon aerosols

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Supplementary Notes

Supplementary Note 1 Sampling sites in East Asia

- (a) Two megacity sampling sites, Beijing and Shanghai, are the largest cities in the Beijing-Tianjin-Hebei (BTH) region and the Yangtze River Delta (YRD), respectively. The Beijing samples were collected at the Peking University sampling site (N39.59, E116.18) as described by, e.g., *Fang et al.*¹. It is located in the Haidian district in northwestern Beijing and treated as a representative urban atmosphere in Beijing, with no major emission sources in the immediate vicinity except two major roads, 150-m to the east and 200-m to the south. The sampling inlet is about 20-m above ground.
- (b) The Shanghai samples were collected at the campus of Fudan University (N31.30, E121.50) in northeast Shanghai¹. This site is also located in an educational, commercial and residential district, with no obvious emission sources around except one major road about 50-m away in the south. The sampling inlet is about 30-m above ground.
- (c) The sampling site at Wuqing district (N39.38, E117.11) of Tianjin City is approximately 100 km southeast of Beijing^{1,2}. Wuqing is an ideal downwind site with the main prevailing wind directions from northern China, which is regarded as a representative atmosphere of the Beijing-Tianjin-Hebei (BTH) region. The sampling inlet is above 13-m height, with no obvious emission sources around except two major roads, distance to nearby streets: SW 75 m, NE 350 m.
- (d) Haining City (N30.51, E120.69) from Zhejiang province is a site for the representative atmosphere of the Yangtze River Delta (YRD), located about 100 km southwest of Shanghai^{1,2}. The sampling inlet is above 25-m height, with no obvious emission sources around, distance to nearby streets: 300-m.

- (e)** The Korea Climate Observatory at Gosan (KCOG, 72 m above sea level (a.s.l), N33.17, E126.10) is an ideal location to intercept continental outflow from East Asia³. KCOG is located in a rural area on the western tip of Jeju Island, 100 km south of the Korean Peninsula, 500 km east of China, and 200 km west of Kyushu Island. The sampling site is far away from the local residential area of this island. It has been used in many campaigns and programs such as the Gosan Pollution Experiment (GoPoEx)⁴, the Aerosol Characterization Experiments (ACE) Asia⁵, the Advanced Global Atmospheric Gases Experiment (AGAGE, <http://agage.eas.gatech.edu/>), the Global Atmosphere Watch (GAW) program (www.wmo.int/pages/prog/arep/gaw_home_en.html), and the International Atmospheric Brown Cloud (ABC) project (www.rrcap.unep.org/abc/)^{6, 7}. Meteorological data at Gosan, including temperature, relative humidity, wind speed and direction at ground level, were continuously measured at Gosan weather station, maintained by the S. Korea Meteorological Administration (KMA).
- (f)** Zhongshan (22.56N, 113.33E) from Guangdong province (Southern China) is a site for the representative atmosphere of the Pearl River Delta (PRD), approximately 100 km away from the corresponding megacities Guangzhou, Shenzhen, and Hong Kong². The sampling inlet is 30-m in height, with no obvious emission sources around, distance to nearby streets: 300-m.
- (g)** Deyang, a site for the representative atmosphere of Sichuan (SC) province (Western China), corresponding to the megacity Chengdu². The sampling coordinate is (31.09N, 104.38E) and the inlet is above 8-m height, with no obvious emission sources around, distance to nearby streets: 400-m.

Supplementary Note 2 A conceptual model for aerosol lifecycle at primary source emission and a state of oxidative evolution (relation for WS-BrC/OC vs. EC) during aerosol transport

The total carbon aerosol comprises Organic (OC) and Elemental Carbon (EC) pools. The OC pool can be divided into water-soluble (WS-BrC) and water-insoluble (WIOC) fractions. A model is used to help understand aerosols' degradation, source regime, and state of oxidative evolution during atmospheric transport. Assume that WS-BrC decays (lifetime), WIOC decays (lifetime), and EC decays (lifetime) while a certain amount of WIOC is oxidized into WS-BrC during the ageing and transport.

Set k_1 , k_3 , and k_4 as the degradation rate of WS-BrC, WIOC and EC, respectively. The k_2 is the oxidative evolution rate of WIOC to WS-BrC during ageing and transport. Then, we get the formulation for the net loss rates of WS-BrC, WIOC and EC as below:

$$\frac{d[\text{WS-BrC}]}{dt} = -k_1 \cdot [\text{WS-BrC}] + k_2 \cdot [\text{WIOC}] \quad (1)$$

$$\frac{d[\text{WIOC}]}{dt} = -(k_2 + k_3) \cdot [\text{WIOC}] \quad (2)$$

$$\frac{d[\text{EC}]}{dt} = -k_4 \cdot [\text{EC}] \quad (3)$$

Solving Eq. (2) and (3):

$$[\text{WIOC}(t)] = [\text{WIOC}(0)] \cdot e^{-(k_2+k_3)t} \quad (4)$$

$$[\text{EC}(t)] = [\text{EC}(0)] \cdot e^{-k_4 t} \quad (5)$$

Incorporating Eq. (4) in Eq. (1), solving and get

$$[\text{WS-BrC}(t)] = \left([\text{WS-BrC}(0)] - \frac{k_2 \cdot [\text{WIOC}(0)]}{k_1 - (k_2 + k_3)} \right) \cdot e^{-k_1 t} + \frac{k_2 \cdot [\text{WIOC}(0)]}{k_1 - (k_2 + k_3)} \cdot e^{-(k_2 + k_3)t} \quad (6)$$

Setting $k' = (k_2 + k_3) - k_1$ ($k > 0$),

$$\frac{[\text{WS-BrC}]}{[\text{OC}]} = \frac{[\text{WS-BrC}]}{[\text{WS-BrC}] + [\text{WIOC}]} = \frac{\left([\text{WS-BrC}(0)] + \frac{k_2 \cdot [\text{WIOC}(0)]}{k'} \right) - \frac{k_2 \cdot [\text{WIOC}(0)]}{k'} \cdot e^{-k' \cdot t}}{\left([\text{WS-BrC}(0)] + \frac{k_2 \cdot [\text{WIOC}(0)]}{k'} \right) + [\text{WIOC}(0)] \cdot \left(1 - \frac{k_2 \cdot [\text{WIOC}(0)]}{k'} \right) \cdot e^{-k' \cdot t}} \quad (7)$$

Simplifying Eq. (7) and defining constants A and B , we can get a same Eq. (8)^{ref. 8}

$$\frac{[\text{WS-BrC}]}{[\text{OC}]} = \frac{1 + A \cdot e^{-k' \cdot t}}{1 + B \cdot e^{-k' \cdot t}} \quad (8)$$

From the Eq. (5) expression of EC, we can solve for t

$$t = - \frac{\ln \left(\frac{[\text{EC}]}{[\text{EC}(0)]} \right)}{k_4} \quad (9)$$

Incorporating into the Eq. (8) expression for WS-BrC/OC:

$$\frac{[\text{WS-BrC}]}{[\text{OC}]} = \frac{1 + A \cdot \left(\frac{[\text{EC}]}{[\text{EC}(0)]} \right)^{\frac{k'}{k_4}}}{1 + B \cdot \left(\frac{[\text{EC}]}{[\text{EC}(0)]} \right)^{\frac{k'}{k_4}}} \quad (10)$$

Then, we establish the relationship for WS-BrC/OC vs. EC, with constant parameters C , D and k :

$$\frac{[\text{WS-BrC}]}{[\text{OC}]} = \frac{1 + C \cdot [\text{EC}]^k}{1 + D \cdot [\text{EC}]^k} \quad (11)$$

Eq. (11) indicates that WS-BrC/OC ratios increase, while EC concentrations decrease when aerosols transport from source to receptor. The different components have different levels in the source region with different lifetimes. Meanwhile, WIOC is also oxidized into WS-BrC to some extent.

To run a fitting routine according to the established Eq. (11) ($D > C > 0$; $k > 0$), we may derive source and evolutionary information from the fitted parameters. The ratio of C/D denotes the ratio of WS-BrC/OC at the source regime. For instance, when the EC loadings are very high, then we get the following equation:

$$\frac{[\text{WS-BrC}]}{[\text{OC}]} = \frac{1+C \cdot [\text{EC}]^k}{1+D \cdot [\text{EC}]^k} = \frac{\frac{1}{[\text{EC}]^k} + C}{\frac{1}{[\text{EC}]^k} + D} \approx \frac{C}{D} \quad (12)$$

Supplementary Note 3 Source-resolved MAC prediction by Monte Carlo simulation

The Monte Carlo (MC) analysis was conducted using in-house scripts written in Python. The fraction of source distributions was described with normal distributions with mean values from the E. and S. Asia observations and a standard deviation of 6%. The calculations were performed by 100,000 iterations. The MAC and $\Delta^{14}\text{C}$ (here refers to fraction of biomass and fossil fuel burnings, f_{bio} and f_{fossil}) values of WS-BrC from both E. (exclude more-coal Beijing/BTH sites) and S. Asia were used in combination with an MC approach to further constrain the potential $\text{MAC}_{\text{source-predicted}}$ from two source classes: biomass and fossil fuel burnings.

$\text{MAC}_{\text{source-predicted}}$ of radiocarbon-extinct fossil fuel sources ($\text{MAC}_{\text{fossil}}$) versus contemporary biomass/biogenic sources (MAC_{bio}) can be determined using the light absorption mass balance equations:

$$\text{MAC}_{\text{sample}}(\text{E. Asia}) = \text{MAC}_{\text{bio}}(\text{E. Asia}) \cdot f_{\text{bio}}(\text{E. Asia}) + \text{MAC}_{\text{fossil}}(\text{E. Asia}) \cdot f_{\text{fossil}}(\text{E. Asia}), \quad (13)$$

$$\text{MAC}_{\text{sample}}(\text{S. Asia}) = \text{MAC}_{\text{bio}}(\text{S. Asia}) \cdot f_{\text{bio}}(\text{S. Asia}) + \text{MAC}_{\text{fossil}}(\text{S. Asia}) \cdot f_{\text{fossil}}(\text{S. Asia}), \quad (14)$$

$$\text{MAC}_{\text{bio}}(\text{E. Asia}) = \text{MAC}_{\text{bio}}(\text{S. Asia}), \quad (15)$$

$$\text{MAC}_{\text{fossil}}(\text{E. Asia}) = \text{MAC}_{\text{fossil}}(\text{S. Asia}), \quad (16)$$

$$f_{\text{fossil}}(\text{E. Asia}) + f_{\text{bio}}(\text{E. Asia}) = 1, \quad (17)$$

$$f_{\text{fossil}}(\text{S. Asia}) + f_{\text{bio}}(\text{S. Asia}) = 1. \quad (18)$$

First, coal played a much lower role in E. Asia (excluding Beijing/BTH sites) and S. Asia, with a very similar relative contribution of coal to EC. Second, the liquid-fossil sources were predominant in E. Asia (2-3 times higher than coal for tracer EC, except for Beijing/BTH sites) and S. Asia (1-3 times higher than coal for tracer EC). It could be roughly estimated and assumed that MAC_{fossil} was likely similar between E. and S. Asia. Third, the MAC_{WS-BrC} (S. Asia) was expected to be mainly contributed by biomass burning due to an extraordinarily higher f_{bio} ($\sim 80 \pm 6\%$) of WS-BrC. In turn, the MAC_{fossil} (S. Asia) would be a minor contribution to the total MAC_{WS-BrC} (S. Asia). Thus, it could not have a particular influence on the MC simulation if we hypothesize MAC_{fossil} (S. Asia) is equal to that of E. Asia.

The probability density distribution (PDD) for the $MAC_{source-predicted}$ is then calculated using Monte Carlo numerical analysis. The median, interquartile range (IQR) and the numerical spread (e.g., the standard deviation) of $MAC_{source-predicted}$ can be derived from this PDD. Based on the Monte Carlo simulation, the median (the second quartile) and interquartile range (IQR) (the difference between the first and the third quartiles) are used to quantify source-resolved MAC and associated uncertainty, respectively.

Supplementary Note 4 Kinetic model of photochemical bleaching of WS-BrC

Several evaluations and kinetic model frameworks have provided rate equations and parameters for different reaction systems or phases to support the investigation and elucidation of atmospheric chemistry, atmospheric aerosol and gas-particle interactions^{43–45}. The mass transport and chemical reactions at the gas-particle interface and inside the bulk of particles are complex physicochemical processes. Chemical reactions proceed during aerosol ageing both at the surface and in the bulk of solid and liquid particles, influencing the atmospheric aerosol composition and the particle's physicochemical properties (i.e., optical) and isotope kinetic

effects. The organic carbon aerosol divided into water-soluble and water-insoluble components can be changed by photochemical reactions. We assume that both water-soluble Brown Carbon (WS-BrC) and water-insoluble organic carbon (WIOC) are degrading between source-to-receptor sites as well as a fraction of WIOC is oxidized to WS-BrC during long-range transport. Additionally, there are no substantial additional organic carbon emissions during over-ocean transport contributing to the higher levels of WS-BrC during aerosol pollution events. For simplicity, in analogy to well-established formalisms of gas-phase chemistry and kinetic models, tropospheric lifetimes of organic species (including aerosol-phase) due to reaction with oxidants such as OH, NO₃ and O₃. The photochemical degradation can be estimated by combining the rate constant with estimated ambient tropospheric concentrations of WS-BrC, WIOC and oxidants. The followings are the most import photochemical reactions that drive the changes in WS-BrC and WIOC.



This dissociation reaction is described by a formal first-order rate law:

$$\frac{-d[XO]}{dt} = k_a[XO] \quad (19)$$

Upon integration for both sides of the equation, the following forms are more convenient:

$$\ln [XO] = \ln [XO]_0 - k_a t \quad (20)$$

$$[XO] = [XO]_0 e^{(-k_a t)} \quad (21)$$



AO_x denotes oxidized products, M represents third body)

A formal second-order rate law describes this reaction. The reaction rate constant k_{po} is a combination coefficient comprising all effects and processes:

$$\frac{-d[A]}{dt} = \frac{d[AO_x]}{dt} = k_{po}[A][O_x] \quad (22)$$

Considering that $[A]_0 - [A]_t = [O_x]_0 - [O_x]_t$ and integration gives

$$\frac{[A]_t}{[O_x]_t} = \frac{[A]_0}{[O_x]_0} e^{\{([A]_0 - [O_x]_0)k_{po}t\}} \quad (23)$$

To simplify higher-order reactions to be a pseudo-first-order reaction condition, we assume that all the oxidation conditions and levels of oxidants are consistent during air transport ($[O_x]_t \sim [O_x]_0$), then the Eq. (22) is approximately the same as that of first-order rate expression as the following:

$$\frac{-d[A]}{dt} = \frac{-d[AO_x]}{dt} = k_i[A] \quad (24)$$

By integrating, we have the following forms:

$$\ln[A] = \ln[A]_0 - k_i t \quad (25)$$

$$[A] = [A]_0 \cdot e^{(-k_i t)} \quad (26)$$

In analogy to the above formalisms of the first-order kinetic models for WS-BrC and WIOC study, then the rate of degradation of WS-BrC and WIOC can be constants k_1 and k_3 , respectively. In addition, there is a rate of k_2 for conversion/oxidation of WIOC to WS-BrC during aerosol transport. The previously proposed conceptual ageing model⁸ postulated that bleaching is the primary driver of the time evolution of WS-BrC light-absorption and stable-carbon isotope signature of $\delta^{13}C_{WS-BrC}$ during long-range transport. If the light-absorption (b_{abs}) of the light (^{12}C) and heavy (^{13}C) isotopes of WS-BrC degrade with first-order kinetics. Then, the transport-time dependence of the ageing proxy ($[\delta^{13}C]_t$) and WS-BrC light-absorption (MAC_{WS-BrC}) can be expressed as the following:

$$\frac{[WS-Br^{13}C]_0 \cdot e^{\{-(k_{13}-k_{12})t\}}}{[WS-Br^{12}C]_0} = \left(\frac{[\delta^{13}C]_t}{1000} + 1\right) \cdot STD \quad (27)$$

(defining $Q \equiv \left(\frac{[\delta^{13}C]_t}{1000} + 1\right) \cdot STD$)

$$MAC(WS-BrC)_t = \frac{c_1 \cdot Q^i}{1+Q} = h \cdot \left(\frac{[\delta^{13}C]_t}{1000} + 1\right)^i / \left(\frac{[\delta^{13}C]_t}{1000} + q\right) \quad (28)$$

Where constant STD is $(WS-Br^{13}C)/(WS-Br^{12}C)_{standard}$, k_{12} and k_{13} are the first-order rate constants for the isotope ^{12}C and ^{13}C , respectively. The first-order kinetic ageing fitting is parameterized by constants h , q and i , while generally, Q is $\ll 1$. Then, the inter-dependencies of MAC_{WS-BrC} and the ageing proxy $\delta^{13}C_{WS-BrC}$ are established.

Supplementary Note 5 Concentration and isotope measurements of carbonaceous aerosols

The concentrations of elemental carbon (EC) and organic carbon (OC) were measured by a thermal-optical transmission (TOT) analyzer (Sunset Laboratory Inc., Tigard, OR, USA) using the National Institute for Occupational Safety and Health (NIOSH) 5040 method⁴⁶. Total carbon (TC) represents the sum of OC and EC. No EC was detected in the field blanks collected during this study, consistent with our earlier studies^{1,2,4,8,13}.

The analytical protocol for WS-BrC concentration measurement was described previously^{1,4,12–14}. Briefly, WS-BrC was Milli-Q water extracted by ultra-sonication, centrifugation, and filtration of the supernatant using 0.02- μm cutoff aluminium syringe filters (Anotop 10 Plus; Whatman, Kent, UK) and quantified by a high-temperature catalytic oxidation instrument (Shimadzu-TOC-VCPH, Japan) following the non-purgeable organic carbon (NPOC) protocol. The TC and WS-BrC concentration values were blank-corrected by subtracting an average of the field blanks. The mean relative standard deviation of triplicate analysis was $< 5\%$ for EC, TC and WS-BrC. The instruments were frequently calibrated by standards and other reference materials.

Synoptic samples with high carbon loadings, corresponding to high-pollution events, were selected to isolate the different carbon components and their isotope measurements. For EC isolation, a filter area corresponding to at least 60 μg of EC for each sample was first treated to

remove carbonates by hydrochloric acid fumigation. Filter subsamples were then subjected to the NIOSH-5040 protocol for oxidation to CO₂, and the produced CO₂ was cryotrapped during the EC combustion phase (after online removal of water and halogen-containing gases through magnesium perchlorate and silver wool traps, respectively^{1,2,14}). The analytical method for isolation of WS-BrC to determine its carbon isotope composition has also been described previously^{1,4,12–14}. Briefly, a filter area required for the subsequent ¹³C and ¹⁴C measurements was extracted in Milli-Q water. The extracts were freeze-dried and then successively redissolved in 300 µl of 1 M hydrochloric acid in order to decarbonate the WS-BrC samples. The WS-BrC samples were then transferred into precombusted silver capsules and dried in the oven at 60 °C. Finally, the dried WS-BrC samples and EC samples in flame-sealed glass ampules were analyzed for their natural ¹⁴C abundance and ¹³C/¹²C ratio using accelerator mass spectrometry (AMS) at the US-NSF NOSAMS Facility (Woods Hole, MA, USA), as described previously^{1,2,4,12–14}.

Supplementary Note 6 Light-absorption measurement of EC

The optical-attenuation ATN (a unit less parameter) is defined by the following equation:

$$\text{ATN} = -100 \cdot \ln\left(\frac{I}{I_0}\right) \quad (29)$$

where I and I_0 are the intensity of incident light and the transmitted light through the filter substrate and aerosols. The determination of ATN by the TOT analyzer⁴⁶ is similar to the Aethalometer. The TOT analyzer provides absorbance (equivalent to initial transmittance) at 632 nm by measuring the intensities of incident and transmitted light through the filter loaded with aerosols. As demonstrated by the other studies^{47,48}, the equivalence was achieved between ATN retrieved from the TOT analyzer and that measured by the Aethalometer when the measurement wavelength is the same. ATN measured from the TOT analyzer can be used to

determine the absorption coefficient (b_{abs} , Mm^{-1}) of the loaded aerosol, similarly to the approach used in the Aethalometer as the following:

$$b_{\text{abs}} = \text{ATN} \cdot \frac{A}{V} \quad (30)$$

where A is the filter area (mm^2) of particle loading, and V is the volume of air sampled (m^3). The bulk of light attenuation measured by filter-based transmission techniques is primarily due to light absorption. However, several factors must be accounted for to convert ATN coefficients into aerosol absorption coefficients (b_{abs}). A factor of C (e.g., 3.6 ± 0.6 for coated aerosol) was suggested to correct the multiple scattering effects of filter-based absorption measurement⁴⁹. The C value depends on the aerosol's absorbing material and BC mixing state^{49,50}. A similar empirical factor was used in the current study, 3.6 for continental sites (i.e., Beijing, Shanghai, BTH, YRD, PRD and SC), and 4.2 for regional receptor site KCOG. Another artefact involved in the filter-based method is the shadow effect, the factor $S(\text{ATN})$. Such shadow effect decreases the absorption by reducing the optical path length. A wavelength-dependent parameter f was used to estimate $S(\text{ATN})$ as below⁴⁹:

$$S(\text{ATN}) = \left(\frac{1}{f} - 1 \right) \cdot \left(\frac{\ln(\text{ATN}) - \ln 0.1}{\ln 0.5 - \ln 0.1} \right) + 1 \quad (31)$$

where $S(\text{ATN})$ depends on parameter f . We have used $f = 1.1$ for the current wintertime aerosol pollution study, which is the same value as f in the previous study⁴⁷. The calculated values of 0.83 ± 0.04 for $S(\text{ATN})$ during the current campaign were similar to that used in other studies, such as 0.85 ± 0.03 in Indian sites⁴⁷ and 0.80 for an urban site in urban Taipei⁵¹.

Thus, the MAC_{EC} is calculated as the following:

$$\text{MAC}_{\text{EC}} (632 \text{ nm}) = \frac{100}{[\text{EC}_s]} \cdot \frac{\text{ATN}}{C \cdot S(\text{ATN})} \quad (32)$$

where EC_s ($\mu\text{g cm}^{-2}$) is the filter loading of EC, optical-attenuation ATN was calculated based on the $\ln$ of the intensity of incident light to transmitted light. The C and $S(\text{ATN})$ are the two

empirical factors suggested for correcting the measured absorption due to the scattering and shadowing effects, respectively.

Regarding bias introduced by the multiple scattering correction factor, the empirical correction factors were recommended as a range of values. We acknowledge that the different correction values applied in the above calculations may lead to slight overestimates or underestimates for MAC_{EC} . For consistency with the previous studies regarding the filter-based light absorption measurements, the correction factor C for urban fresh aerosols is 3.6, and a relatively higher correction value of 4.2 is applied for aged aerosol intercepted at the regional receptor site.

The linear regression analysis (see Supplementary Fig. 18) generated a slope of $25.5 \text{ m}^2 \text{ g}^{-1}$ (attenuation cross-section) and an intercept of 14; EC_s loadings $>9 \mu\text{g cm}^{-2}$ were excluded from the regression analysis and MAC_{EC} calculations^{47,51}. Similarly, in Asian campaigns, a value of $23.7 \text{ m}^2 \text{ g}^{-1}$ for the slope and 12.0 for the intercept has been reported for urban Taipei⁵¹, and a slope of $22.4 \text{ m}^2 \text{ g}^{-1}$ and an intercept of 6.6 has been provided for Indian sites⁴⁷. As shown in Supplementary Fig. 10, the calculated MAC_{EC} (based on Eq. (32) in Supplementary Note 6) exhibited a larger variability, spanning from 5.3 to $13.3 \text{ m}^2 \text{ g}^{-1}$ during the study periods over E. Asia. Previous studies have suggested that the variability in MAC_{EC} may be due to differences in sources, location, chemical composition and the mixing/coating state of BC in aerosols^{31–33,47–52}. The obtained MAC_{EC} for E. Asia were in the range of the literature's ambient values observed in polluted regions^{31,47–52}.

Supplementary Note 7 Directive solar absorbance of WS-BrC relative to BC

Assuming the light-absorption spectra of aerosols could be separated into water-soluble BrC and BC components (here refers to the mass analogue of EC). Combining the measurements

of the optical properties for both WS-BrC and BC (also well-known spectral properties) will enable us to assess the differences in solar absorbance of these two light-absorbing carbon materials. For this liquid extraction-based method, a previous study estimated that the absorption of WS-BrC in the atmospheric particle phase might be two folds higher than light absorption in extracts²⁶. A correction factor of >1 has been suggested to convert bulk solution absorption to particle light absorption. However, it is unclear how this factor relates to samples collected at different source regions and after long-distance transport⁵³. Thus, in the following discussion, we report both the uncorrected and the corrected (conversion factor of 2) values for the relative solar radiation absorbance of WS-BrC/BC.

In order to compare the amount of solar radiation (integrating over the solar spectrum) absorbed by WS-BrC relative to BC at ground level in E. Asia, a simplistic absorption-based calculation is employed to provide an estimate. The fraction of solar radiation absorbed by WS-BrC relative to BC was calculated using a simplistic model^{13,14,30}. In this model, the solar emission spectrum ($I_0(\lambda)$) is approximated using the clear sky Air Mass 1 Global Horizontal (AM1GH) solar irradiance spectrum at the Earth's surface, including the light-absorptive effects of, e.g., ozone and water vapour⁵⁴.

The fraction (f) solar energy absorbed by WS-BrC relative to EC is calculated as:

$$f = \frac{\int I_0(\lambda) \left\{ 1 - e^{-\left(\text{MAC}_{365} \left(\frac{365}{\lambda} \right)^{\text{AAE}} \cdot C_{\text{WS-BrC}} \cdot h_{\text{ABL}} \right)} \right\} d\lambda}{\int I_0(\lambda) \left\{ 1 - e^{-\left(\text{MAC}_{\text{EC}} \left(\frac{j}{\lambda} \right) \cdot C_{\text{EC}} \cdot h_{\text{ABL}} \right)} \right\} d\lambda} \quad (33)$$

where λ is the wavelength, MAC_{365} is the $\text{MAC}_{\text{WS-BrC}}$ at 365 nm or conversion values with a factor of two. MAC_{EC} is the MAC for EC measured at wavelength j (for this study, 632 nm). The h_{ABL} is the height of the atmospheric aerosol boundary layer (ABL). Here the AAE for BC is set to 1. The h_{ABL} is set to 1000 m (mean ABL ranges 500–1000 m from Mie lidar

measurements at KCOG, see Supplementary Fig. 19) but has very little influence on the computed ratio in the 100–3000 m range.

The simplistic estimate is based on several assumptions: AÅE for WS-BrC measured at 330–400 nm represents the whole spectral range, the BC absorption is a $1/\lambda$ relationship, the measurements at ground level represent the whole ABL and the AM1GH well approximates the solar light. This calculation aims to broadly evaluate the solar spectrum integrated relative absorbance of WS-BrC and EC at a ground-level situation over E. Asia.

Supplementary Note 8 Synoptic weather pattern, AOD and HYSPLIT BTs

Aerosol Optical Depth (AOD) data were obtained from NASA Moderate Resolution Imaging Spectroradiometer (MODIS) level 3 collection 6^{ref. 55}. The monthly mean NCEP/NCAR reanalysis wind vectors (m s^{-1}) and geopotential height at 850 hPa were averaged during the sampling periods, with 6-hourly archived meteorological data provided by the US National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS)^{55,56}. Back trajectories (BTs) at heights 500 m and 100 m (a.g.l.) for KCOG (Gosan, Jeju Island, South Korea) were calculated using the NOAA HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2003)^{40–42,55}.

Supplementary Note 9 Appendix for the abbreviations

AÅE: Absorption Ångström Exponent

AMS: accelerator mass spectrometry

BB: biomass burning

BC: black carbon

BrC: brown carbon

BCOB: Bangladesh Climate Observatory at Bhola

BTs: Back Trajectories

BTH: Beijing-Tianjin-Hebei area

EI: emission inventories

IPCC: Intergovernmental Panel on Climate Change

IQR: interquartile range

KCOG: Korea Climate Observatory at Gosan

KIE: kinetic isotope effects

MAC: mass absorption cross-section

MC: Monte Carlo

MCMC: Markov-Chain Monte Carlo

MCOH: Maldives Climate Observation at Hanimaadhoo

NIOSH: National Institute for Occupational Safety and Health

OA: Organic aerosols

OC: organic carbon

PDD: probability density distributions

PRD: Pearl River Delta

RMSD: root-mean-square deviation

SC: Sichuan

SOA: secondary organic aerosols

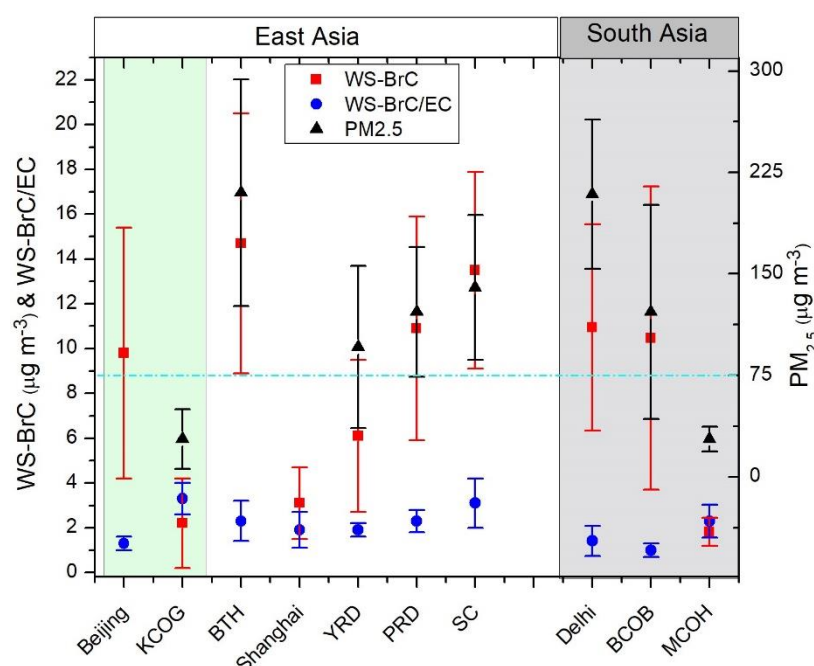
TOT: thermal-optical transmission

VOCs: volatile organic carbons

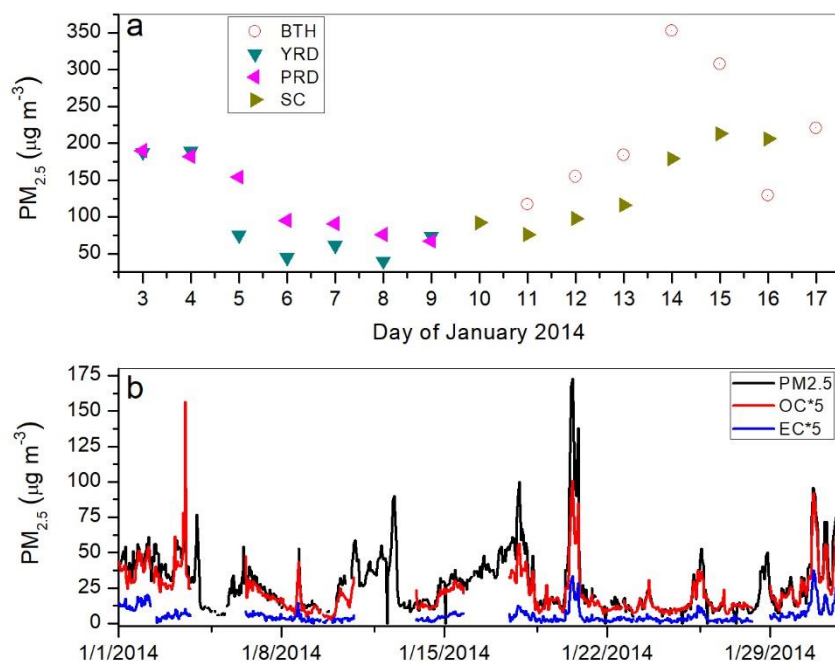
WSOC: water soluble organic carbon

YRD: Yangtze River Delta

Supplementary Figures

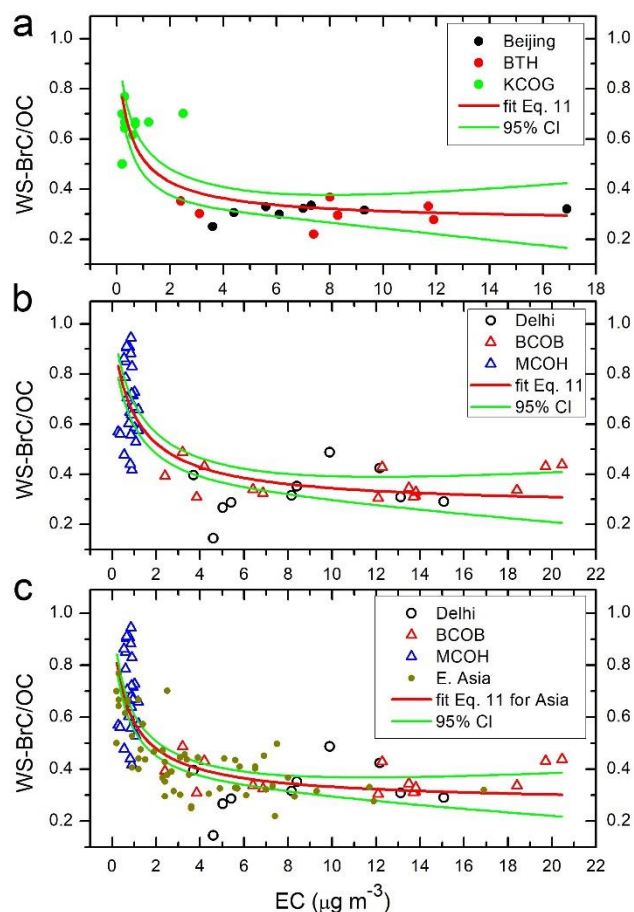


Supplementary Fig. 1 Comparison of PM_{2.5} and WS-BrC levels during winter campaigns between E. and S. Asia. A consistently increasing WS-BrC-to-EC ratio during over-sea transport from the continental source regions to the regional receptor sites suggests an important distinction for the lifecycle of these two sorts of light-absorbing aerosols. For E. Asia, all data were measured from simultaneously collected PM_{2.5} samples in the current Jan. 2014 campaign, covering N. China (Beijing; BTH), E. China (Shanghai; YRD), S. China (PRD), W. China (SC) and Yellow Sea regional receptor site of the Korea Climate Observatory at Gosan (KCOG). Concentration data for winter S. Asia (i.e., urban Delhi: central Indo-Gangetic Plain (IGP); IGP outflow site BCOB: Bangladesh Climate Observatory at Bhola; Indian Ocean regional receptor MCOH (for MCOH, particulate matter concentrations are PM₁₀); Maldives Climate Observatory at Hanimaadhoo) in 2016 from Dasari et al.⁸. Blue dash line denotes China's Ambient Air Quality Standards for maximum allowable daily PM_{2.5} concentration (75 $\mu\text{g m}^{-3}$). Error bars represent the average values with standard deviations for each site. Possible regional source-to-receptor connections are shaded by cyan (E. Asia) and grey (S. Asia). Site acronyms: BTH (Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China). Error bars denote the average values with s.d. for each site.

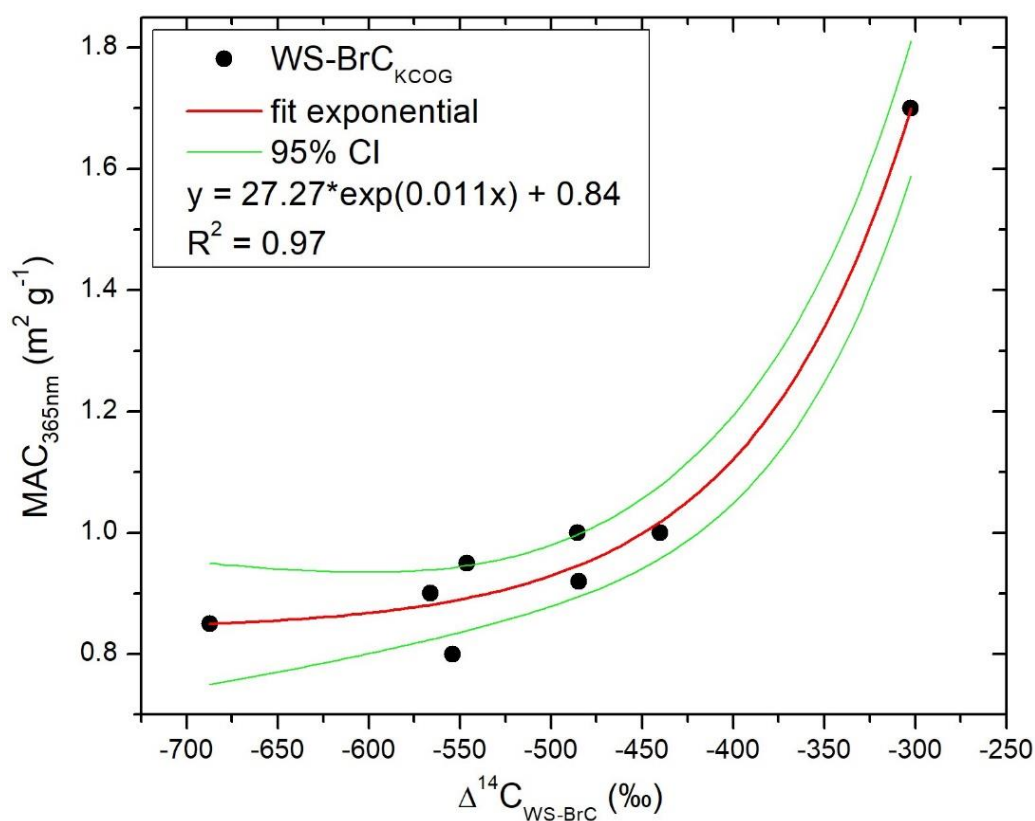


Supplementary Fig. 2 Temporal PM_{2.5} concentrations during E. Asian winter campaign.

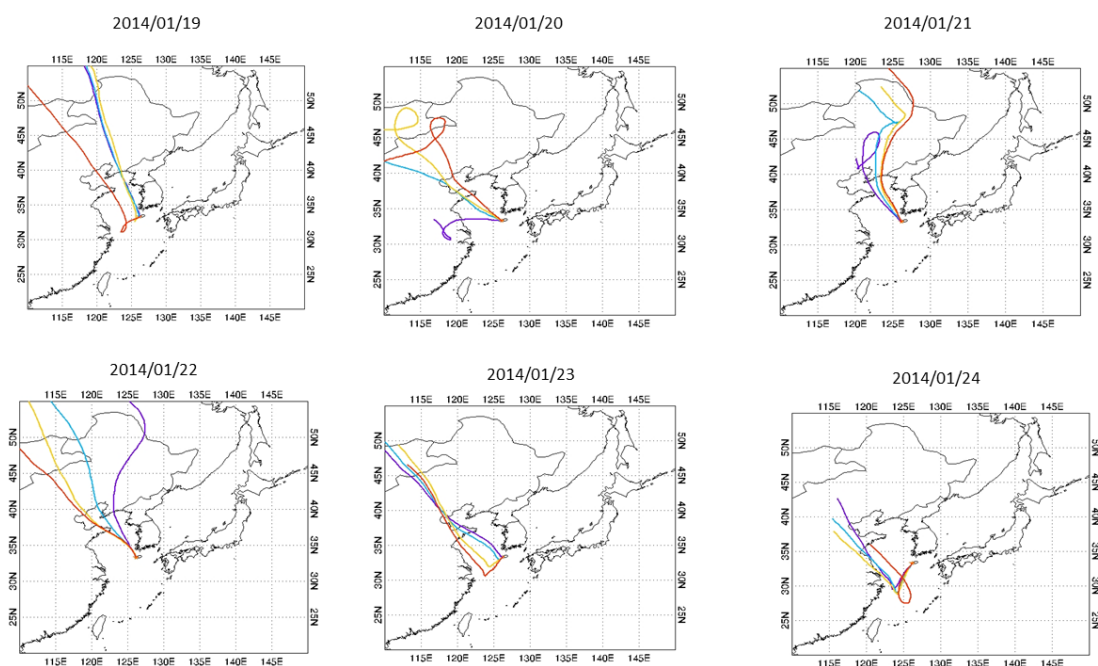
a Daily concentration for collected PM_{2.5} samples in the current January 2014 campaign, BTH (Tianjin in the Beijing-Tianjin-Hebei region, N. China), YRD (Haining in the Yangtze River Delta, nearby megacity Shanghai, E. China), PRD (Zhongshan in the Pearl River Delta, nearby Guangzhou, S. China), SC (Deyang in Sichuan Basin, nearby Chengdu, W. China). **b** Hourly PM_{2.5} concentrations (UTC time) and frequent pollution events at SE Yellow Sea regional receptor site of the Korea Climate Observatory at Gosan (KCOG), some missing OC and EC data were due to semi-online Sunset instrument problem.



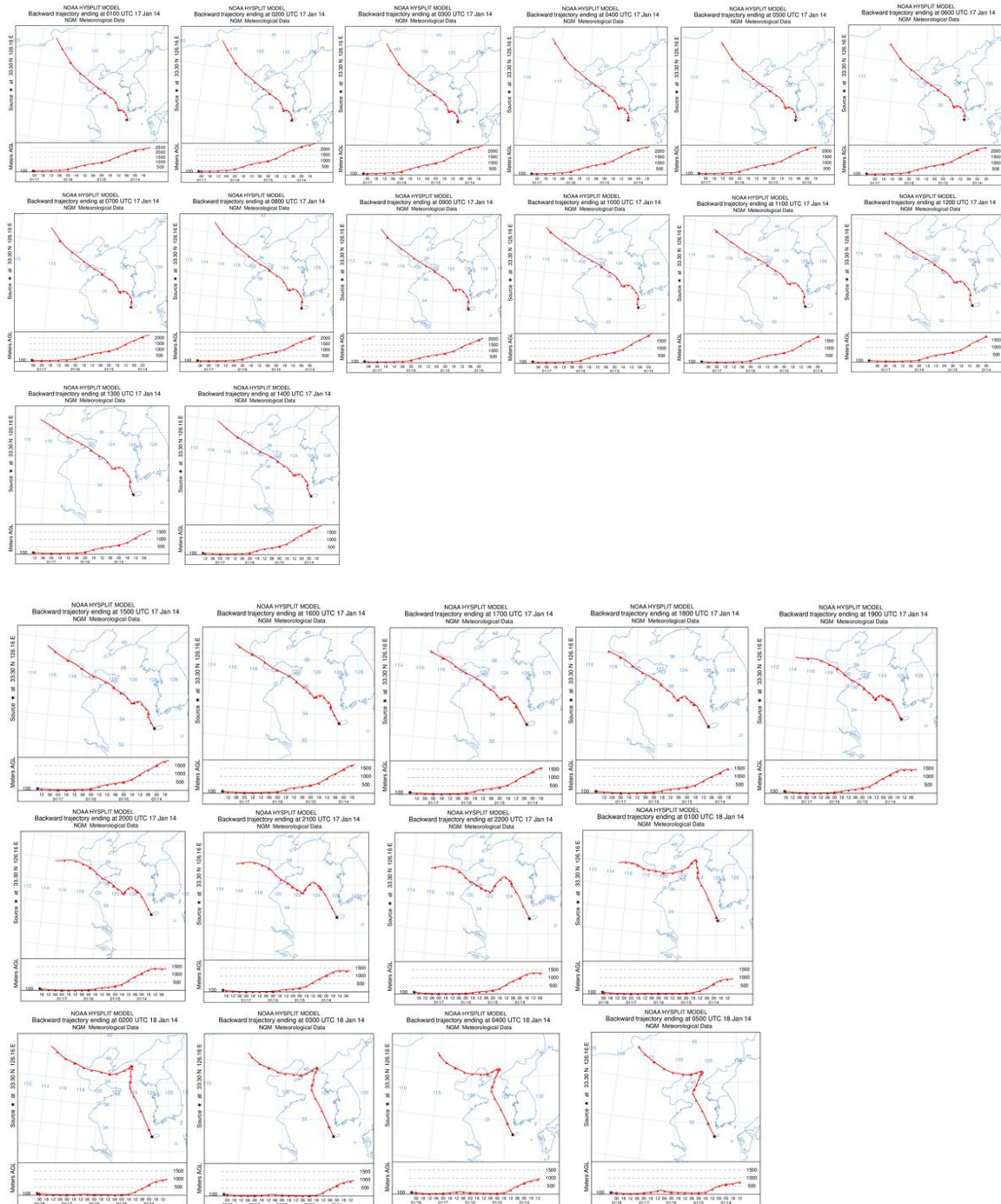
Supplementary Fig. 3 The inverse dependence of WS-BrC-to-OC ratios with EC concentrations in PM_{2.5} from the continental emission source areas to the ocean background receptor sites. A theoretical model function, equation (11) in Supplementary Note 2, is fitted to WS-BrC-to-OC ratios vs. EC loadings for **a** E. Asia (Beijing-BTH-KCOG transit), **b** S. Asia (Delhi-BCOB-MCOH transit) and **c** the combined data from both E. Asia (including Beijing, BTH, Shanghai, YRD, PRD, SC and KCOG) and S. Asia. A model description for the fitting routine is detailed in Supplementary Note 2. The goodness of the best-of-fit RMSD is 0.11 for E. Asian transit (panel **a**: $k = 0.87$; $C/D = 0.26$), 0.15 of RMSD for S. Asian transit (panel **b**: $k = 0.92$; $C/D = 0.28$), and 0.13 of RMSD for E. and S. Asia with an asymptotic value C/D being 0.26 (panel **c**: $k = 0.86$). Raw concentration data for winter S. Asia (i.e., urban Delhi: central Indo-Gangetic Plain (IGP); IGP outflow site BCOB: Bangladesh Climate Observatory at Bhola; Indian Ocean regional receptor MCOH: Maldives Climate Observatory at Hanimaadhoo) in 2016 from Dasari et al.⁸.



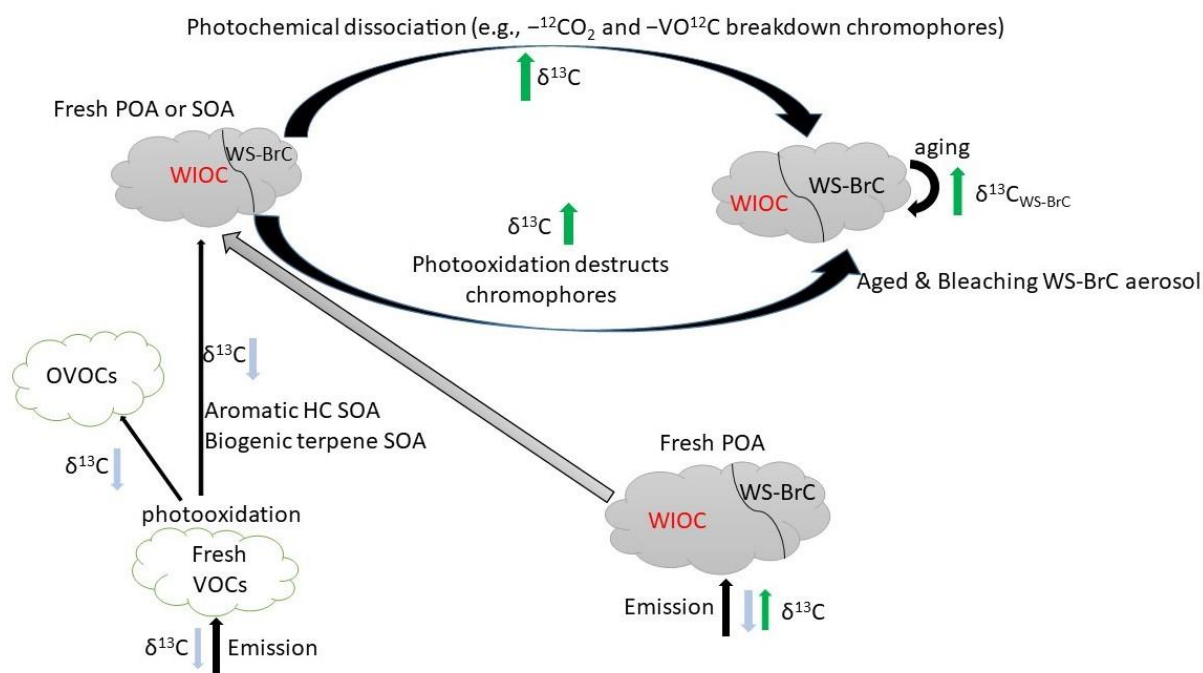
Supplementary Fig. 4 Relationship between $\text{MAC}_{\text{WS-BrC}}$ (365nm) and radiocarbon $\Delta^{14}\text{C}_{\text{WS-BrC}}$ in $\text{PM}_{2.5}$ samples during the aerosol pollution events at the receptor site KCOG. Light-absorption efficiency (per WS-BrC mass) of WS-BrC as a function of radiocarbon signature. The younger (the more modern; higher f_{bio}) the carbon of WS-BrC is, the higher its light-absorption efficiency. The goodness of the best-fit curve showed by RMSD of 0.05. KCOG (Korea Climate Observatory at Gosan).



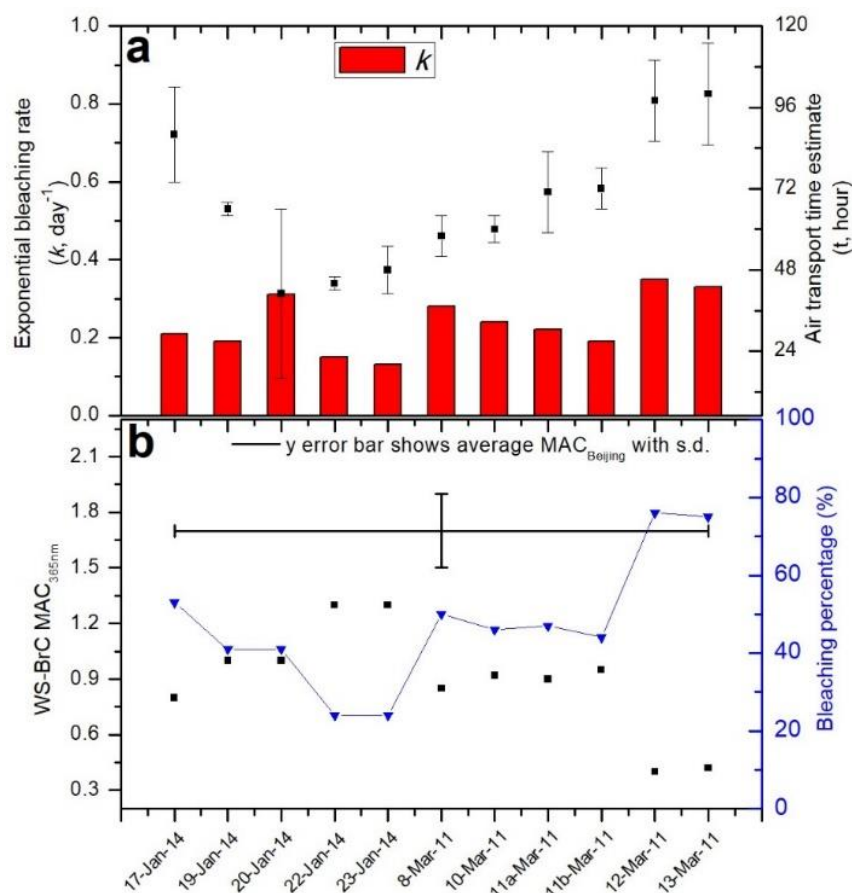
Supplementary Fig. 5 NOAA HYSPLIT^{40–42} five-day air mass back-trajectories (BTs) for KCOG. Starting at 100-m height, every 6 hour a day from 2014/01/19 to 2014/01/24. (Korea Climate Observatory at Gosan).



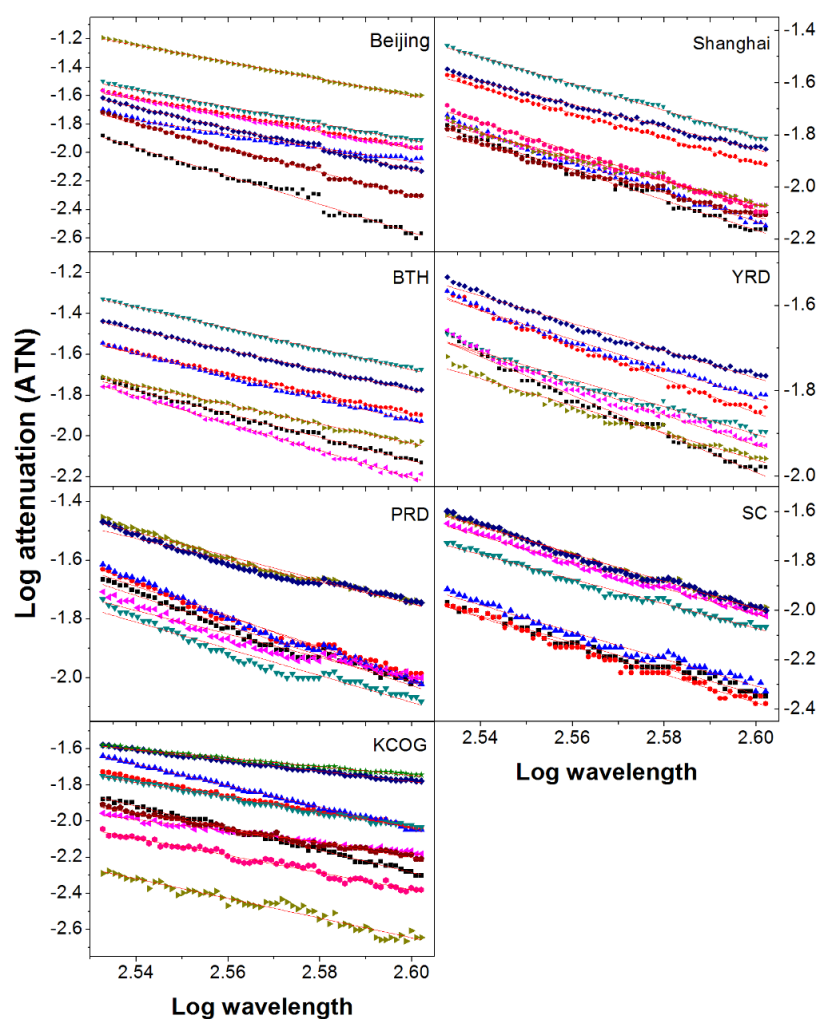
Supplementary Fig. 6 NOAA HYSPLIT⁴⁰⁻⁴² five-day air mass back-trajectories (BTs) for KCOG. Starting at a height of 100 m, and every hour for 2014/01/17 and 2014/01/18.



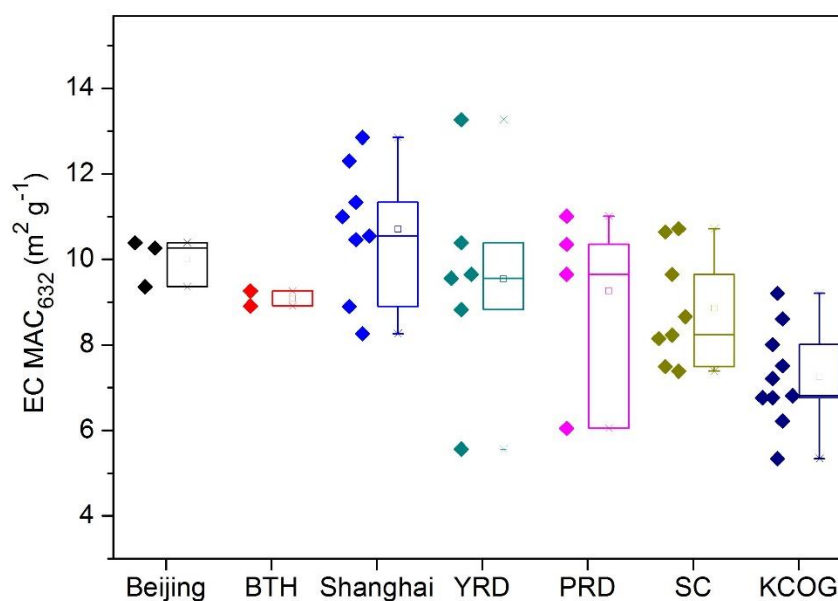
Supplementary Fig. 7 Atmospheric photolysis/oxidation processes destruct chromophores to reduce WS-BrC absorptivity by enriching stable carbon isotope $\delta^{13}\text{C}_{\text{WS-BrC}}$ during anthropogenic aerosol transport. A summary illustrates how different atmospheric processes are expected to affect the $\delta^{13}\text{C}_{\text{WS-BrC}}$ and light absorption of WS-BrC. POA, SOA, VOCs, OVOCs and HC denote primary organic aerosols, secondary organic aerosols, volatile organic compounds, oxygenated VOCs and hydrocarbons, respectively.



Supplementary Fig. 8 First-order bleaching rate k (day⁻¹), air transport times and bleaching percentage for the respective WS-BrC aerosol during the dispersal of East Asian haze. a Red column depicts the calculated exponential bleaching rate for each WS-BrC sample intercepted at the regional receptor KCOG (shown by the left-hand side axis). Squares show the average Beijing-to-KCOG air transport time for each KCOG sample, based on hourly HYSPLIT back trajectories (BTs) calculation (shown by the right axis). **b** Squares and error bars represent the measured WS-BrC MAC_{365nm} at KCOG and Beijing, respectively (shown by the left axis). Blue triangles denote the bleaching percentage for each WS-BrC at KCOG (shown by the right axis). WS-BrC MAC_{365nm} for 2011 KCOG pollution campaign from ref. ⁴. KCOG (Korea Climate Observatory at Gosan).



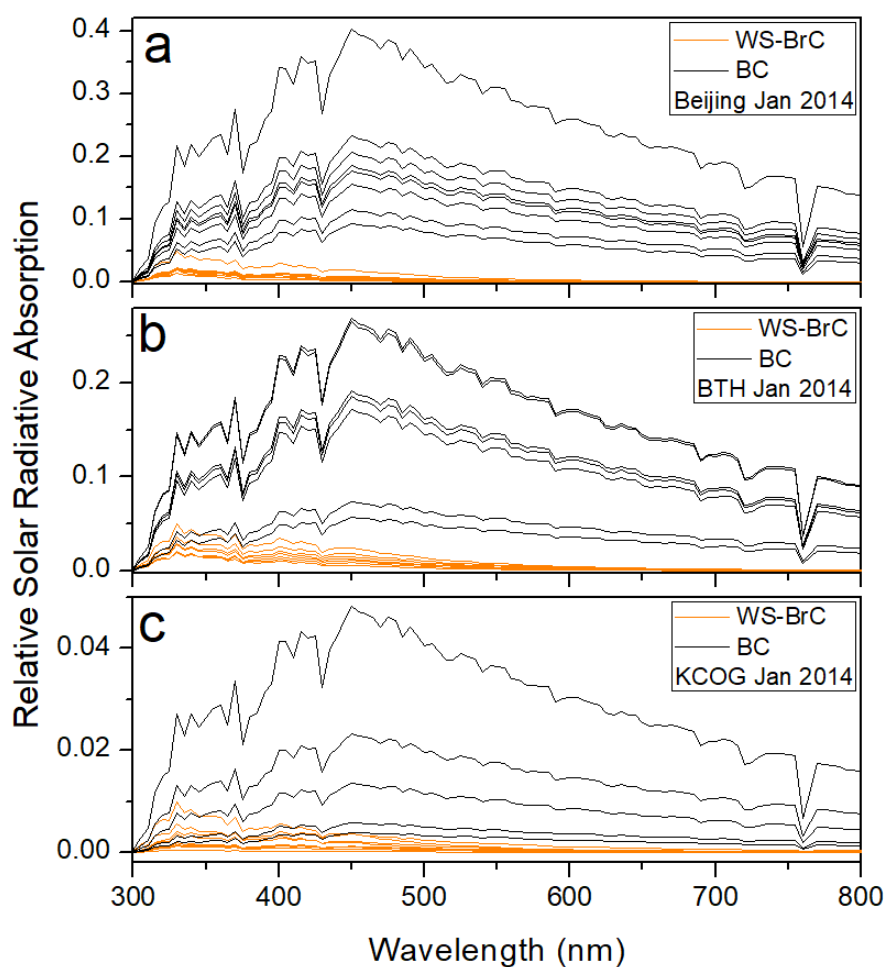
Supplementary Fig. 9 Wavelength-dependent absorption spectra and the linear absorption Ångström coefficient (AAE) fitting for WS-BrC samples during January 2014 E. Asian campaign ($R^2 > 97\%$; $P < 0.05$). ATN denotes the light attenuation of WS-BrC extracts. Each line represents one sample. Site acronyms: BTH (Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China), KCOG (Korea Climate Observatory at Gosan).



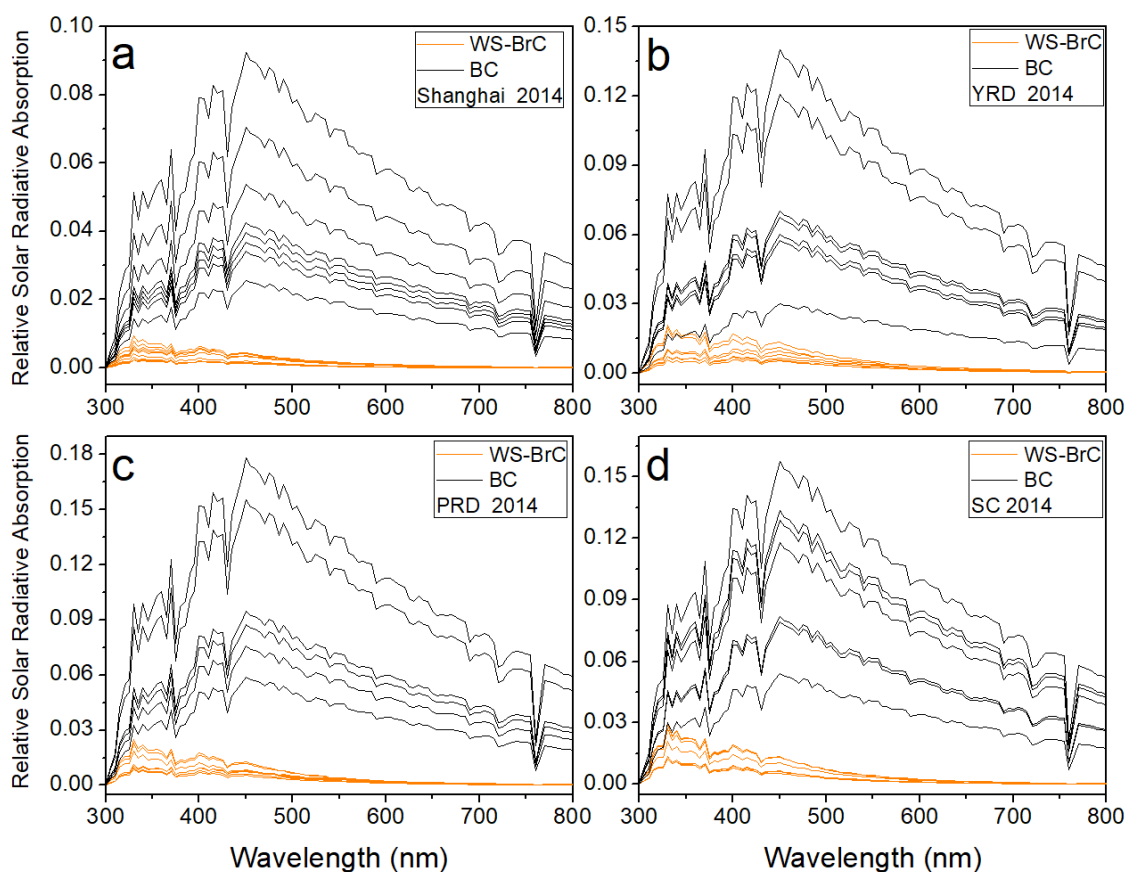
Supplementary Fig. 10 MAC of EC measured during January 2014 E. Asian campaign.

MAC of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $\text{m}^2 \text{g}^{-1}$) using a TOT sunset OCEC analyzer. Site acronyms: BTH (Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China), KCOG (Korea Climate Observatory at Gosan). For the box plot, the centre line shows the median; the centre box defines the mean; the box range is 25% to 75% (upper and lower quartiles); the whiskers represent the maximum and minimum values for each observation.

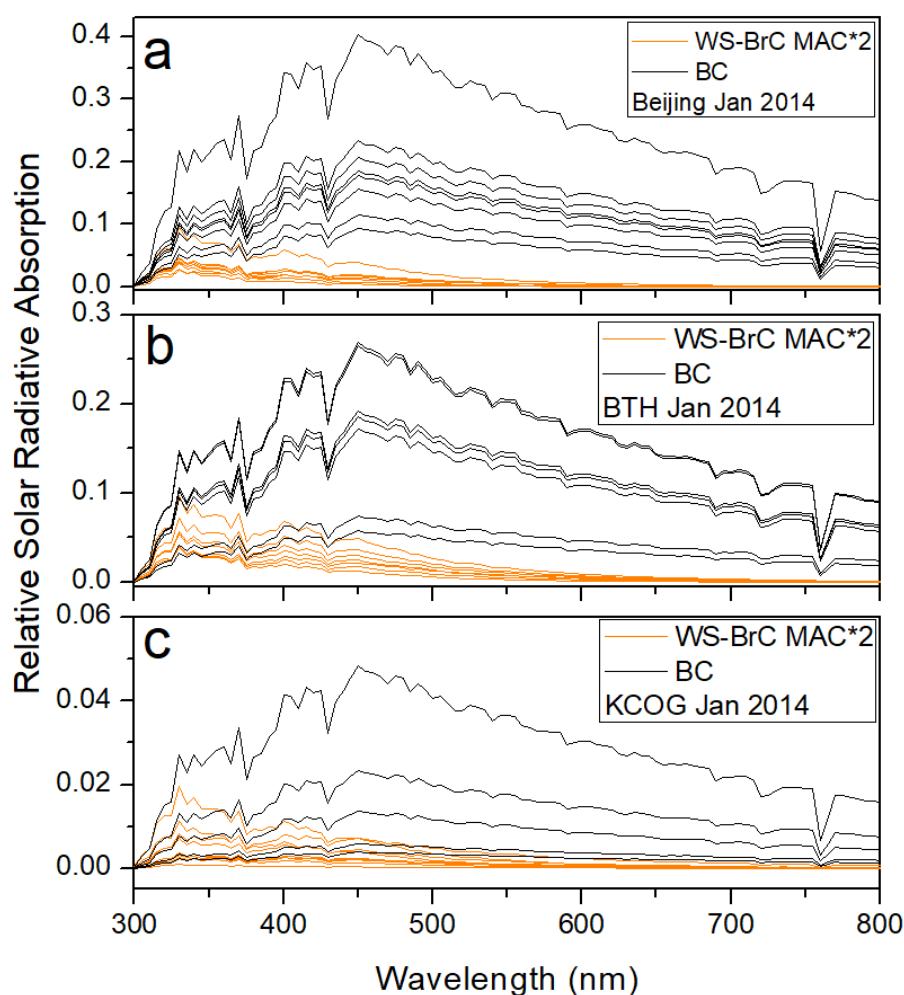
Note: The scattering effects have been corrected by an empirical correction factor (C) of 3.6, except 4.2 for KCOG (see Supplementary Note 6). Filter samples with EC loadings higher than $9 \mu\text{g cm}^{-2}$ were excluded in the EC MAC calculations, as suggested by refs. ^{47,48,52} due to large multi factors effects.



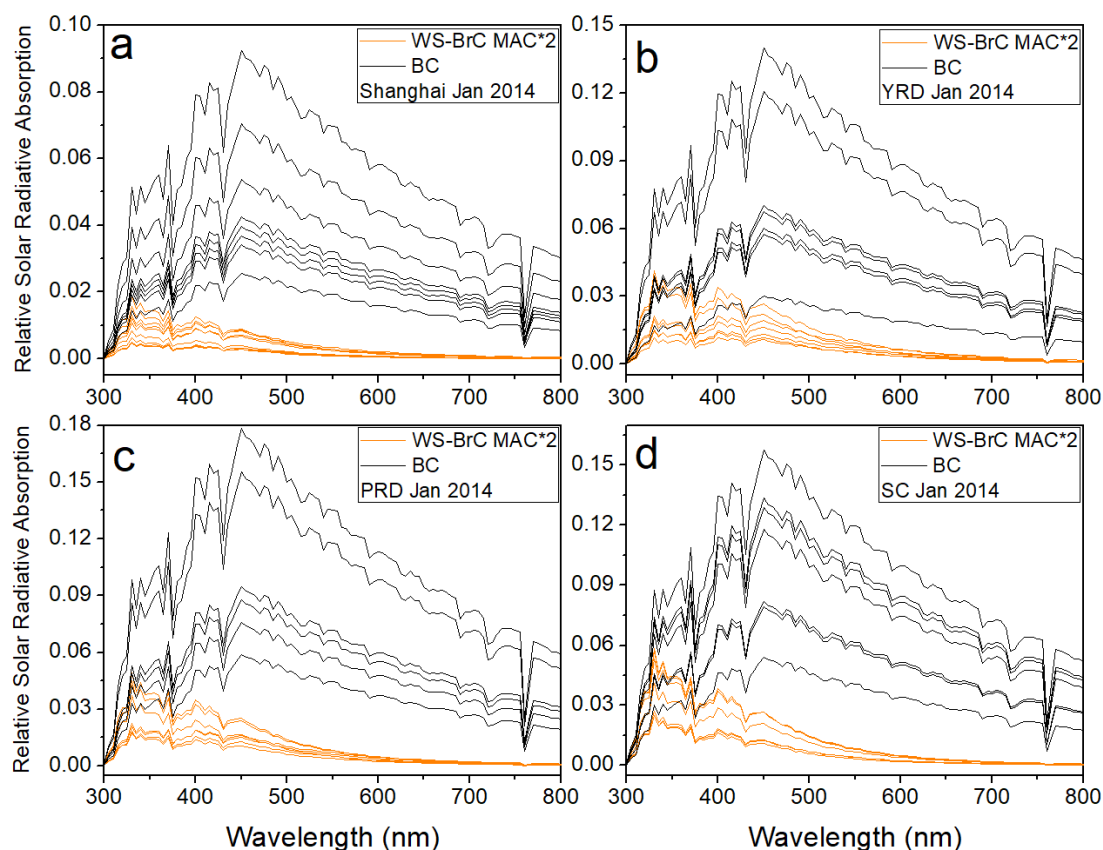
Supplementary Fig. 11 Tropospheric solar absorption of WS-BrC and BC against wavelength (300–800 nm) at ground level in Beijing (a), BTH (b) and KCOG (c). MAC of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $m^2 g^{-1}$). Each orange line represents WS-BrC solar absorption for one sample, while the black line refers to the solar absorption of BC by sample. The plots provide an overall comparison of solar absorption by WS-BrC and BC in the study wavelength. Site acronyms: BTH (Wuqing district of Tianjin in the Beijing-Tianjin-Hebei region, China), KCOG (Korea Climate Observatory at Gosan).



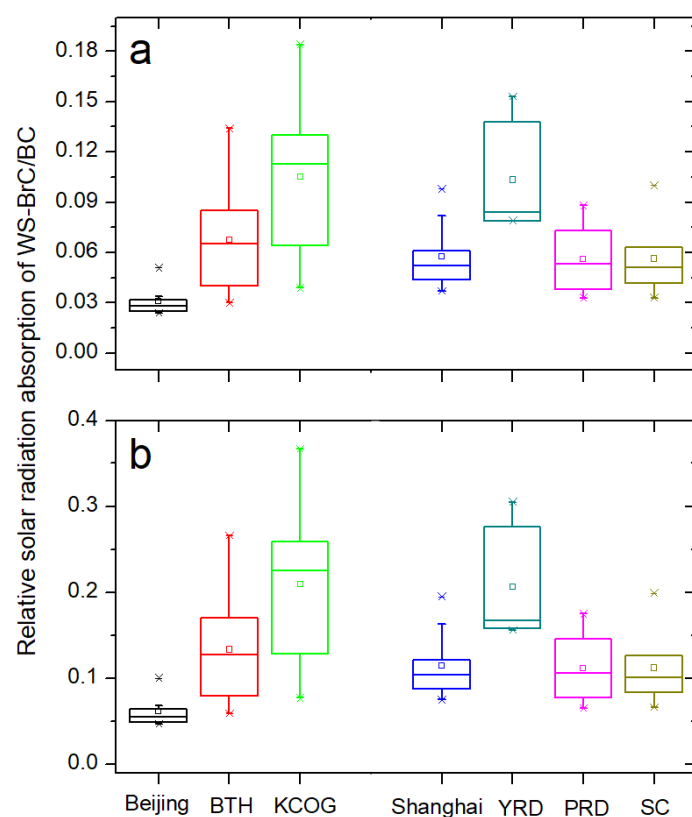
Supplementary Fig. 12 Tropospheric solar absorption of WS-BrC and BC against wavelength (300–800 nm) at ground level in Shanghai (a), YRD (b), PRD (c) and SC (d). MAC of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $\text{m}^2 \text{g}^{-1}$). Each orange line represents WS-BrC solar absorption for one sample, while the black line refers to the solar absorption of BC by sample. The plots provide an overall comparison of solar absorption by WS-BrC and BC in the study wavelength. Site acronyms: YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China).



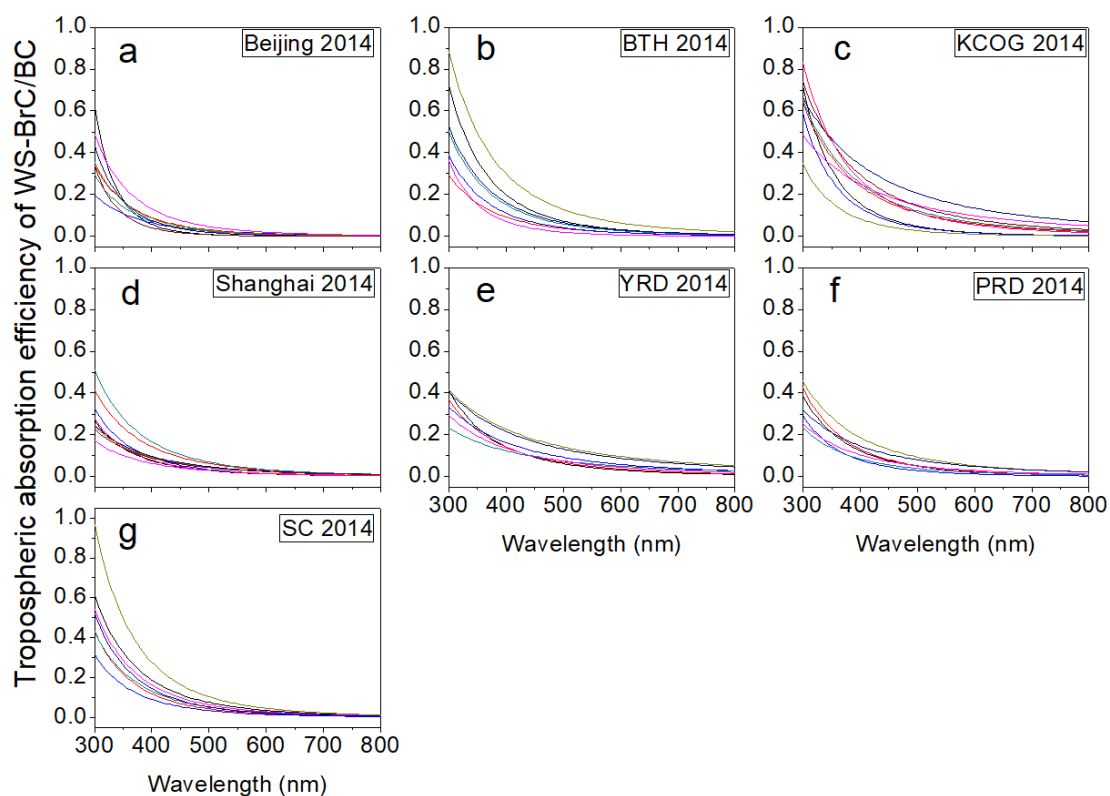
Supplementary Fig. 13 Tropospheric solar absorption of WS-BrC and BC against wavelength (300–800 nm) at ground level in Beijing (a), BTH (b) and KCOG (c). MAC of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $\text{m}^2 \text{g}^{-1}$). The MAC values of WS-BrC used for this computation were corrected by a conversion factor of 2. Each orange line represents WS-BrC solar absorption for one sample, while the black line refers to the solar absorption of BC by sample. The plots provide an overall comparison of solar absorption by WS-BrC and BC in the study wavelength. Site acronyms: BTH (Wuqing district of Tianjin in the Beijing-Tianjin-Hebei region, China), KCOG (Korea Climate Observatory at Gosan).



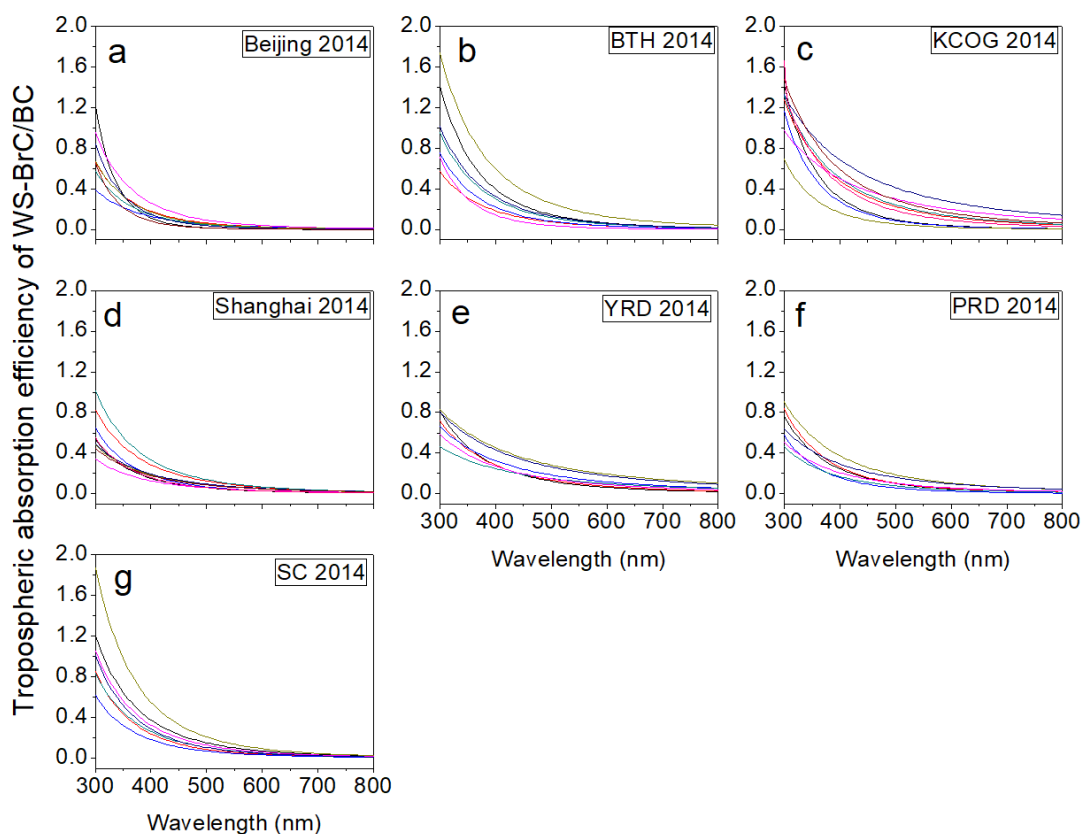
Supplementary Fig. 14 Tropospheric solar absorption of WS-BrC and BC against wavelength (300–800 nm) at ground level in Shanghai (a), YRD (b), PRD (c) and SC (d). MAC of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $\text{m}^2 \text{g}^{-1}$). The MAC values of WS-BrC used for this computation were corrected by a conversion factor of 2. Each orange line represents WS-BrC solar absorption for one sample, while the black line refers to the solar absorption of BC by sample. The plots provide an overall comparison of solar absorption by WS-BrC and BC in the study wavelength. Site acronyms: YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China).



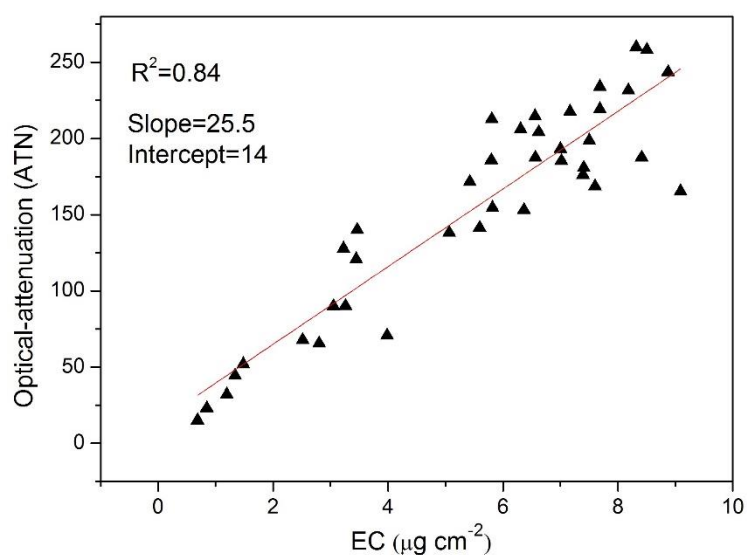
Supplementary Fig. 15 Relative solar radiation absorbance of WS-BrC-to-BC. a Uncorrected WS-BrC MACs used for the fractions (f) calculations. **b** WS-BrC MACs used for the fractions (f) calculations were corrected by a conversion factor of 2. The MACs of EC (or MAC_{BC}) measured at 632 nm (MAC_{632} , $\text{m}^2 \text{g}^{-1}$) from this study were used to calculate the relative absorbance. Site acronyms: BTH (Wuqing district of Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China), KCOG (Korea Climate Observatory at Gosan). For the box plot, the centre line shows the median; the centre box defines the mean; the box range is 25% to 75% (upper and lower quartiles); the whiskers represent the maximum and minimum values for each observation.



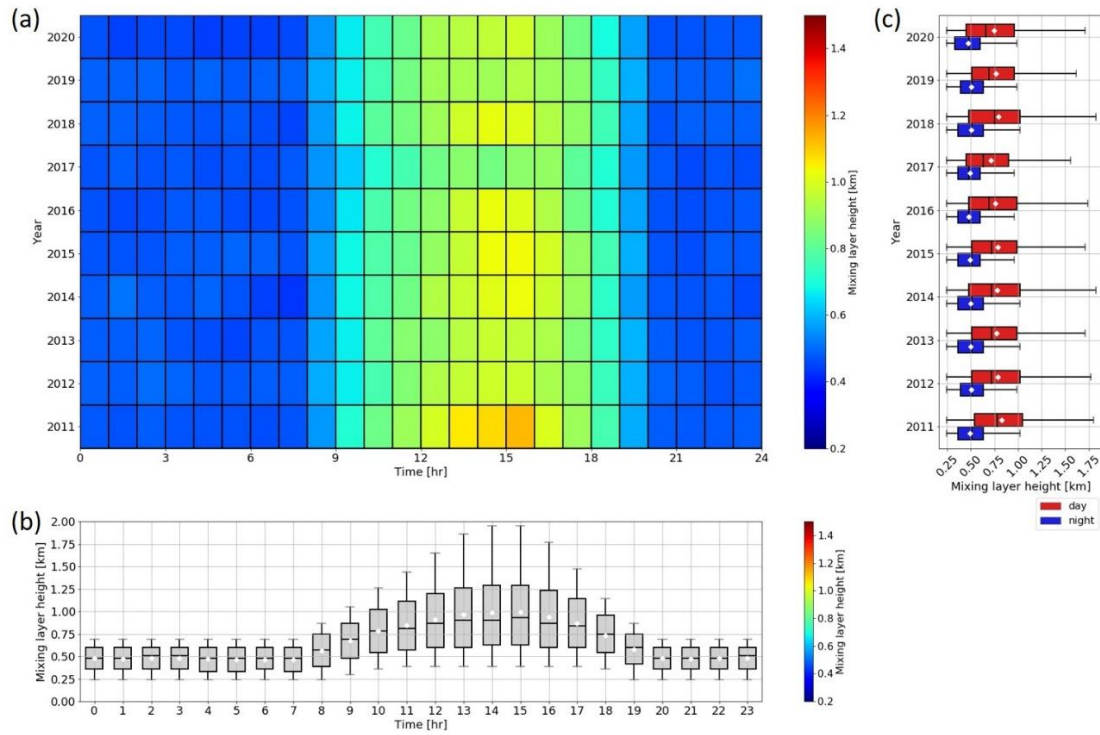
Supplementary Fig. 16 Wavelength dependence (300–800 nm) of tropospheric solar absorption efficiency of WS-BrC/BC at ground level. a Beijing, **b** BTH, **c** KCOG, **d** Shanghai, **e** YRD, **f** PRD and **g** SC. Y axis means the ratios of MAC_{WS-BrC} multiplied by the solar spectrum to the MAC_{BC} multiplied by the solar spectrum. MAC of EC (or MAC_{BC}) used from the measurement at 632 nm (MAC_{632} , $m^2 g^{-1}$). Each line represents one sample. Site acronyms: BTH (Wuqing district of Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China), KCOG (Korea Climate Observatory at Gosan).



Supplementary Fig. 17 Wavelength dependence (300–800 nm) of tropospheric solar absorption efficiency of WS-BrC/BC at ground level. a Beijing, **b** BTH, **c** KCOG, **d** Shanghai, **e** YRD, **f** PRD and **g** SC. Y axis means the ratios of MAC_{WS-BrC} multiplied by the solar spectrum to the MAC_{BC} multiplied by the solar spectrum. MAC of EC (or MAC_{BC}) used from the measurement at 632 nm (MAC_{632} , $m^2 g^{-1}$). The MAC values of WS-BrC used for this computation were corrected by a conversion factor of 2. Each line represents one sample. Site acronyms: BTH (Tianjin in the Beijing-Tianjin-Hebei region, China), YRD (Haining in the Yangtze River Delta, China), PRD (Zhongshan in the Pearl River Delta, China), SC (Deyang in Sichuan province, China), KCOG (Korea Climate Observatory at Gosan).



Supplementary Fig. 18 Linear relationship between optical-attenuation (ATN) and filter-sample EC loading ($\mu\text{g cm}^{-2}$). The linear fitting parameters are shown with slope and intercept. Red line shows the linear regression line and the corresponding correlations (R^2 ; $p < 0.05$). Filter samples with EC loadings $> 9 \mu\text{g cm}^{-2}$ were not included in the regression analysis (suggested by refs. ^{47,48,52} due to large multi factors effects).



Supplementary Fig. 19 a Diurnal variation of Mixing Layer Height (MLH) from year to year; b total diurnal variation of MLH; c the year-to-year MLH during the day (red) and night (blue) from MLH retrieved using Gosan (Jeju Island, S. Korea) Mie lidar measurements.

Supplementary Tables

Supplementary Table 1. Sampling times, PM_{2.5} loadings and OC/EC ratios for samples collected in E. Asia in January 2014 (timing indicated by UTC).

Site-sample ID	Start time (day, time)	End time (day, time)	PM _{2.5} (µg m ⁻³)	OC/EC
Beijing-0109	09, 01:00	10, 00:30	—	4.8
Beijing-0111	11, 02:45	12, 00:45	—	3.2
Beijing-0112	12, 01:00	13, 00:30	—	2.6
Beijing-0113	13, 01:15	14, 00:45	—	3.5
Beijing-0114	14, 01:00	15, 00:30	—	6.3
Beijing-0115	15, 01:00	16, 00:30	—	4.4
Beijing-0117	17, 01:00	18, 00:30	—	5.3
Beijing-0121	21, 01:00	22, 00:30	—	4.4
BTH-0111	11, 11:45	12, 11:35	117	9.1
BTH-0112	12, 11:45	13, 11:35	155	3.1
BTH-0113	13, 11:45	14, 11:35	184	5.2
BTH-0114	14, 11:40	15, 10:47	353	7.9
BTH-0115	15, 11:38	16, 11:28	308	7.8
BTH-0116	16, 11:35	17, 11:25	129	11.1
BTH-0117	17, 11:35	18, 11:25	221	6.9
Shanghai-0109	09, 01:21	10, 01:12	—	3.3
Shanghai-0110	10, 01:23	11, 01:47	—	5.0
Shanghai-0111	11, 01:57	12, 01:04	—	3.9
Shanghai-0112	12, 01:16	13, 01:13	—	6.6
Shanghai-0113	13, 01:25	14, 01:24	—	2.5
Shanghai-0114	14, 01:35	15, 01:20	—	3.5
Shanghai-0115	15, 01:30	16, 01:36	—	4.0
Shanghai-0116	16, 01:45	17, 01:15	—	4.1
Shanghai-0117	17, 01:30	18, 02:30	—	3.9
YRD-0103	03, 10:45	04, 10:30	188	7.0
YRD-0104	04, 10:45	05, 10:35	189	6.2
YRD-0105	05, 10:45	06, 10:35	75	4.5
YRD-0106	06, 10:45	07, 10:35	45	4.1
YRD-0107	07, 10:45	08, 10:35	61	5.6
YRD-0108	08, 10:45	09, 10:35	40	5.5
YRD-0109	09, 10:40	10, 10:29	74	5.5
KCOG-011719	17, 00:43	19, 01:29	10–100 (hourly)	6.1
KCOG-011920	19, 01:50	20, 00:35	10–40	6.7
KCOG-012022	20, 01:35	22, 01:58	10–173	3.9
KCOG-012223	22, 02:05	23, 01:14	10–15	4.2
KCOG-012324	23, 01:20	24, 01:30	10–16	4.2
KCOG-012425	24, 01:41	25, 00:49	11–16	2.9
KCOG-012526	25, 01:00	26, 01:26	12–65	5.1
KCOG-012627	26, 01:33	27, 00:34	13–17	5.5
KCOG-012728	27, 00:40	28, 00:35	12–18	5.6
KCOG-012829	28, 00:42	29, 00:51	13–70	4.7
PRD-1401	03, 15:35	04, 14:35	190	6.9
PRD-1403	04, 14:40	05, 13:40	182	6.6
PRD-1405	05, 13:45	06, 12:45	154	3.7
PRD-1407	06, 12:50	07, 11:50	95	4.1
PRD-1409	07, 11:55	08, 10:55	91	6.0
PRD-1411	08, 11:05	09, 10:05	76	7.0
PRD-1413	09, 10:10	10, 08:10	67	4.8
SC-1401	10, 20:30	11, 19:30	92	8.7
SC-1403	11, 19:40	12, 18:10	76	5.7
SC-1405	12, 18:14	13, 16:45	98	4.2
SC-1407	13, 16:50	14, 15:20	116	5.1
SC-1409	14, 15:35	15, 13:35	179	6.7
SC-1411	15, 13:40	16, 11:40	213	13.8
SC-1413	16, 11:45	17, 09:45	206	6.5

Note for site acronyms: BTH (Tianjin, China), YRD (Haining, China), PRD (Zhongshan, China), SC (Deyang in Sichuan province, China) and KCOG (Korea Climate Observatory at Gosan).

Supplementary Table 2. Summary table of WS-BrC, EC and OC concentrations as well as ratios of WS-BrC/OC, WS-BrC/EC, WS-BrC/TC, EC/TC and OC/EC in PM_{2.5} during January 2014 E. Asian Campaign. Shown by mean $\pm$ standard deviation.

site	WS-BrC ($\mu\text{g m}^{-3}$)	EC ($\mu\text{g m}^{-3}$)	OC ($\mu\text{g m}^{-3}$)	WS-BrC/OC (%)	WS-BrC/EC	WS-BrC/TC (%)	EC/TC (%)	OC/EC
Beijing	9.8 $\pm$ 5.6	7.5 $\pm$ 3.9	31.4 $\pm$ 17.1	31 $\pm$ 3	1.3 $\pm$ 0.3	24.8 $\pm$ 2.0	19.7 $\pm$ 4.4	4.3 $\pm$ 1.2
BTH	14.7 $\pm$ 5.8	7.5 $\pm$ 3.4	49.3 $\pm$ 21.4	31 $\pm$ 5	2.3 $\pm$ 0.9	26.8 $\pm$ 4.3	13.4 $\pm$ 5.0	7.4 $\pm$ 2.6
Shanghai	3.1 $\pm$ 1.6	1.6 $\pm$ 0.7	6.7 $\pm$ 3.3	46 $\pm$ 6	1.9 $\pm$ 0.8	36.4 $\pm$ 6.2	20.6 $\pm$ 4.0	4.0 $\pm$ 1.0
YRD	6.1 $\pm$ 3.4	3.1 $\pm$ 1.5	18.2 $\pm$ 10.9	36 $\pm$ 4	1.9 $\pm$ 0.3	30.1 $\pm$ 3.6	15.8 $\pm$ 2.2	5.6 $\pm$ 0.9
PRD	10.9 $\pm$ 5.0	4.8 $\pm$ 1.8	26.4 $\pm$ 11.6	41 $\pm$ 7	2.3 $\pm$ 0.5	34.5 $\pm$ 5.8	15.8 $\pm$ 3.3	5.6 $\pm$ 1.3
SC	13.5 $\pm$ 4.4	4.6 $\pm$ 1.5	31.6 $\pm$ 11.4	43 $\pm$ 3	3.1 $\pm$ 1.1	37.4 $\pm$ 2.3	13.4 $\pm$ 3.8	7.2 $\pm$ 3.0
KCOG	2.2 $\pm$ 2.0	0.7 $\pm$ 0.7	3.3 $\pm$ 2.9	66 $\pm$ 4	3.3 $\pm$ 0.7	54.7 $\pm$ 4.6	17.6 $\pm$ 3.4	4.7 $\pm$ 0.9

Note for site acronyms: BTH (Tianjin, China), YRD (Haining, China), PRD (Zhongshan, China), SC (Deyang in Sichuan province, China) and KCOG (Korea Climate Observatory at Gosan).

Supplementary Table 3. Source signatures (endmembers) of radiocarbon $\Delta^{14}\text{C}$ and stable carbon $\delta^{13}\text{C}$ used in Markov-Chain Monte Carlo (MCMC) source apportionment calculations (mean $\pm$ standard deviation). According to *Andersson et al.*, (2015)^{ref. 9}.

	Biomass (C3 plants)	Liquid fossil	Coal
$\Delta^{14}\text{C}$ (‰)	112 ± 60	-1000	-1000
$\delta^{13}\text{C}$ (‰)	-26.7 ± 1.8	-25.5 ± 1.3	-23.38 ± 1.3

Supplementary Table 4. Raw data $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ of EC and WS-BrC reported for current 2014 January E. Asian campaign and other E. and S. Asian cites in previous literatures. (fraction biomass = 1 – fraction fossil).

Site	Season	Species	$\Delta^{14}\text{C}$ (-‰)	Fraction fossil (%) [*]	Fraction biomass (%) [*]	$\delta^{13}\text{C}$ (‰)	Reference
E. Asia, this campaign							
KCOG	Winter (Jan. 2014)	WS-BrC	302–554	50 ± 8	50 ± 8	22.8 ± 0.2	1
Beijing	“	WS-BrC	461–576	56 ± 3	44 ± 4	23.5 ± 0.7	“
Beijing	“	EC	772 ± 18	79 ± 2	21 ± 2	24.4 ± 0.2	“
KCOG	“	EC	625 ± 7	67 ± 7	33 ± 5	23.9 ± 0.3	“
Shanghai	“	EC	764 ± 11	79 ± 1	21 ± 1	25.8 ± 0.1	“
BTH	“	EC	741 ± 35	77 ± 3	23 ± 3	24.3 ± 0.5	“
E. Asia							
KCOG	Winter (Mar. 2011)	WS-BrC	299–575		~50	23.0 ± 0.7	4
Beijing	Winter (Jan. 2013)	WS-BrC	518 ± 38	57 ± 3	43 ± 3	23.5 ± 0.5	10
BTH	Winter (Jan. 2013)	EC	710 ± 18	74 ± 2	26 ± 2	23.8 ± 0.4	2
PRD	“	EC	650 ± 30	69 ± 3	31 ± 3	26.2 ± 0.3	“
YRD	“	EC	644 ± 24	68 ± 2	32 ± 2	25.6 ± 0.4	“
Shanghai (in YRD)	Winter 2013	OC		49 ± 2			11
Guangzhou (in PRD)	Winter 2013	OC		35 ± 7	65 ± 26		“
S. Asia							
SINH, India	Annual (2008/09)	WS-BrC	152 ± 42	24 ± 4	76 ± 4	20.8 ± 0.9	12
“	Jan. to Feb. 2008	WS-BrC	157 ± 9		76 ± 1	20.0 ± 0.3	“
“	Jan. to Feb. 2009	WS-BrC	167 ± 34		75 ± 3	20.2 ± 0.6	“
MCOH, Maldives	Annual (2008/09)	WS-BrC	47 ± 50	14 ± 4	86 ± 4	18.5 ± 0.9	“
“	Jan. to Feb. 2008	WS-BrC	62 ± 53	16 ± 5	84 ± 5	17.6 ± 0.1	“
“	Jan. to Feb. 2009	WS-BrC	89 ± 10	18 ± 1	82 ± 1	18.5 ± 0.3	“
New Delhi, India	Winter (Nov. 2010 to Feb. 2011)	WS-BrC	118 ± 40	21 ± 4	79 ± 4	24.1 ± 1.0	13
MCOH, Maldives	Winter (Feb. to Mar. 2012)	WS-BrC	30 ± 53		87 ± 5	20.8 ± 0.7	14
“	“	EC	330 ± 55		60 ± 5	25.8 ± 0.3	“
New Delhi, India	Winter (Jan. 2011)	EC	444 ± 102	50 ± 8	50 ± 8	25.7 ± 0.3	15
“	Winter (Dec.2010 to Jan. 2011)	EC	516 ± 102	56 ± 9	44 ± 9	25.8 ± 0.3	“
BCOB, Bangladesh	Winter (Jan. to Mar. 2016)	EC	463 ± 35		52 ± 3	27.2 ± 0.2	16
MCOH, Maldives	Winter (Jan. to Mar. 2016)	EC	440 ± 61		50 ± 5	25.0 ± 0.3	“
Dhaka, Bangladesh	Winter (Nov. 2013 to Feb. 2014)	EC	549 ± 72		41 ± 6	26.4 ± 0.3	17
“	“	EC	330 ± 32		60 ± 3	26.7 ± 0.3	“

Note: E. Asia: KCOG (Korea Climate Observatory at Gosan); YRD (Haining, China); PRD (Zhongshan, China); BTH (Wuqing in Tianjin, China). S. Asia: urban Delhi, central Indo-Gangetic Plain (IGP); SINH represents Sinhadag in Indian; Indian Ocean regional receptor MCOH denotes Maldives Climate Observatory at Hanimaadhoo Island; IGP outflow site BCOB: Bangladesh Climate Observatory at Bhola.

^{*}denotes all the fractions of fossil (and biomass), except the ref. 11 (due to lack of raw isotope data), were computed in this study using Markov-Chain Monte Carlo (MCMC) simulations, with the same endmembers for $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ shown in Table 3.

Supplementary Table 5. Bayesian MCMC source apportionments (biomass, coal and liq. fossil) of EC using raw isotope ($\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$) data reported for current Jan-2014 E. Asian campaign and other E. and S. Asian cites in previous literatures. In order to facilitate the comparison and consistency, the MCMC computation in this study utilized the same source endmembers for $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ (see Supplementary Table 3). Fraction values shown with mean $\pm$ standard deviation (%).

Site	Season	fraction biomass	fraction coal	fraction liquid fossil	Reference*
E. Asia this campaign					
KCOG	Jan. 2014				
	“	33 ± 2	54 ± 11	13 ± 11	1
BTH	“	23 ± 2	56 ± 13	21 ± 13	“
Beijing	“	20 ± 2	53 ± 14	27 ± 14	“
Shanghai	“	21 ± 2	22 ± 14	57 ± 15	“
E. Asia previous literatures					
BTH	Winter (Jan. 2013)	26 ± 2	62 ± 9	12 ± 9	2
YRD	“	32 ± 2	24 ± 14	44 ± 14	“
PRD	“	32 ± 2	13 ± 10	55 ± 10	“
S. Asia					
MCOH, Maldives	Winter (Feb. to Mar. 2012)	60 ± 6	20 ± 12	20 ± 12	14
New Delhi, India	Winter (Jan. 2011)	50 ± 9	22 ± 14	28 ± 15	15
New Delhi, India	Winter (Dec. 2010 to Jan. 2011)	43 ± 9	25 ± 15	32 ± 17	“
Dhaka, Bangladesh	Winter normal days (Nov. 2013 to Feb. 2014)	41 ± 7	21 ± 15	38 ± 16	17
“	“ (hartal days)	60 ± 4	17 ± 11	23 ± 11	“
BCOB, Bangladesh	Winter (Jan. to Mar. 2016)	52 ± 4	14 ± 12	34 ± 13	16
MCOH, Maldives	Winter (Jan. to Mar. 2016)	50 ± 6	26 ± 14	24 ± 14	“

Note: E. Asia: KCOG (Korea Climate Observatory at Gosan); YRD (Haining, China); PRD (Zhongshan, China); BTH (Wuqing in Tianjin, China). S. Asia: urban Delhi, central Indo-Gangetic Plain (IGP); Indian Ocean regional receptor MCOH denotes Maldives Climate Observatory at Hanimaadhoo Island; IGP outflow site BCOB: Bangladesh Climate Observatory at Bhola.

*denotes that the fractions of biomass, coal and liq. fossil were calculated by Markov-Chain Monte Carlo (MCMC) in this study using the raw ^{13}C and ^{14}C data from the corresponding reference, with the same endmembers for $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ shown in Table 3.

Supplementary Table 6. Previous studies pertaining to light absorptive properties of WS-BrC and other

Brown Carbon in source and ambient aerosols. Grey shaded denotes measurement from the same or similar experimental protocol for water-extracted BrC at 365 nm. (It should be noted that: 1. Lab and source test data are limited to the inter-comparison between different combustion sources, and direct comparison is also limited to the same method and same wavelength. 2. The optical values obtained from the source (burning) tests could be largely different from ambient aerosol samples. The real atmospheric samples are a complex mixture, from innumerable burnings of varying fuels at different burning conditions, and involved chemical-physical evolution. 3. Direct comparison of values should use the measurement from the same protocol and at the same wavelength.)

Site/source	MAC (m ² g ⁻¹)	λ (nm)	AÅE	Method (water, methanol extraction)	Reference
Laboratory & Sources test:					
HULIS	0.029–0.031	532	6–7	Spectrophotometer in water extracts	<i>Hoffer et al. (18)</i>
HULIS	0.2–3	405	1.9–5.4	Spectrophotometer in methanol extracts	<i>Lambe et al. (19)</i>
SOA	0.001–0.088	405	5.2–8.8	“	“
Biomass smoke	5.0	350	4.8	Filter-based transmittance after acetone extraction	<i>Kirchstetter et al. (20)</i>
“	0.6	550		Filter-based Spectrometer for OC	“
Wood, grass, corn	~1.0 ± 0.26 (0.52–1.47)	365		Spectrometer for WS-BrC	<i>Du et al. (21)</i>
Motorcycle	0.09–0.32	“		“	“
Biomass burning	1.6 ± 0.55	365		Spectrometer for WS-BrC	<i>Tang et al. (22)</i>
Anthracite coal	1.3 ± 0.34	“		“	“
Bituminous coal	2.0 ± 0.75	“		“	“
Vehicle	0.71 ± 0.3	“		“	“
Biomass burning	2.3 ± 1.1	“		methanol extracted BrC	“
Anthracite coal	0.88 ± 0.74	“		“	“
Bituminous coal	3.2 ± 1.1	“		“	“
Vehicle	0.26 ± 0.09	“		“	“
Bituminous coal	1.10 ± 0.16	“	9.7 ± 1.8	Spectrometer for WS-BrC	<i>Yan et al. (10)</i>
“	1.57 ± 0.78	“	7.46 ± 0.77	methanol extracted BrC	“
Bituminous coal	0.3–0.7	“	11–13	Spectrometer for WS-BrC	<i>Li et al. (23)</i>
Anthracite coal	0.9–1.0	“	7.5–11	“	“
Bituminous coal	1.2–2.8	“	7.7–11	methanol extracted BrC	“
Anthracite coal	0.9–2.4	“	8.2–12	methanol extracted BrC	“
Biomass burning/HULIS	0.79–1.56	“	6.7–8.6	Spectrometer for WS-BrC	<i>Park et al. (24)</i>
Biomass burning/HULIS	0.97–2.09		6.7–8.2	Spectrometer for WS-BrC	<i>Fan et al. (25)</i>
Coal burning/HULIS	0.63		13.6 ± 0.2	“	“
Ambient aerosols from N. America:					
Atlanta, US	0.13–0.53	365	6.0–8.0	Spectrophotometer, water extracts	<i>Liu et al. (26)</i>
“	0.27–0.41	“	4.0–6.0	Spectrophotometer, methanol extracts	“
Georgia, US	0.29–0.70	“	6.2–8.3	Spectrophotometer, water extracts	<i>Hecobian et al. (27)</i>
Atlanta, US	0.33	“	3.4 ± 0.7	particle-into-liquid sampler, water extracts based Spectrophotometer	<i>Zhang et al. (28)</i>
Pasadena, US	0.88	“	3.2 ± 1.2	“	“
Los Angeles, US	0.71	“	7.6 ± 0.5	“	<i>Zhang et al. (29)</i>
Residential area, California	0.43	400	3.0–7.4	Filter-based transmittance	<i>Kirchstetter and Thatcher (30)</i>
Ambient aerosols from E Asia:					
Beijing, China (winter)	1.79 ± 0.24	365	7.5	Spectrophotometer, water extracts	<i>Cheng et al. (31)</i>
Beijing, China (summer)	0.71 ± 0.20	365	7.1	“	“
Beijing, China	0.92	440	6.2	Sun-sky radiometer + Mie theory	<i>Wang et al. (32)</i>

Beijing, China (winter)	1.26	365	7.5	Spectrophotometer, water extracts	<i>Du et al. (21)</i>
Beijing, China (summer)	0.51	365	7.5	“	“
Xianghe, China	0.5	550	3.5	Aethalometer	<i>Yang et al. (33)</i>
Gosan, KCOG	0.7 ± 0.2	365	6.4 ± 0.6	Spectrophotometer, water extracts	<i>Kirillova et al. (4)</i>
Beijing, China (winter)	1.54 ± 0.16	365	5.3 ± 0.4	“	<i>Yan et al. (34)</i>
Beijing, China (summer)	0.73 ± 0.15	365	5.8 ± 0.5	“	“
Gosan (E. Asian outflow)	0.8–2.0	520	1.6–1.8	Aethalometer	<i>Chung et al. (35)</i>
Ambient aerosols from S Asia:					
Indian Ocean, MCOH, Maldives	0.46 ± 0.18	365	7.2 ± 0.7	Spectrophotometer, water extracts	<i>Bosch et al. (14)</i>
New Delhi (IGP) ^a	1.6 ± 0.5	365	5.1 ± 2.0	“	<i>Kirillova et al. (13)</i>
Kharagpur (IGP), India	0.78 ± 0.24	365	6.0 ± 1.1	“	<i>Bikkina and Sarin. (36)</i>
Bay of Bengal/Indian Ocean	0.45 ± 0.14	365	9 ± 3	“	<i>Bikkina and Sarin. (37)</i>
Pyramid, Nepal	~0.6	365	2.6–4.9	“	<i>Kirillova et al. (38)</i>
Kanpur (IGP), India	—	405	6.6 ± 0.3	Photo Acoustic Soot Spectrophotometry	<i>Shamjad et al. (39)</i>

Note: ^aIndo-Gangetic Plain; MCOH denotes Maldives Climate Observatory at Hanimaadhoo Island; KCOG for Korea Climate Observatory at Gosan.

Supplementary Table 7. Relative solar radiation absorbance of WS-BrC/BC (300–2500 nm) during January 2014 E. Asian campaign.

site	Relative light absorbance of WS-BrC/BC	
	(%) ^a	(%) ^b
Beijing	3.1 ± 1.0	6.2 ± 1.6
BTH	6.7 ± 3.2	13.3 ± 6.5
Shanghai	5.8 ± 1.9	11.5 ± 3.7
YRD	10.3 ± 3.0	20.6 ± 5.7
PRD	5.6 ± 1.8	11.2 ± 3.6
SC	5.6 ± 2.0	11.2 ± 4.0
KCOG	10.5 ± 4.3	21.0 ± 8.6

^aRelative light absorbance of WS-BrC/BC was calculated for the whole solar spectrum (300–2500 nm), using MAC_{WS-BrC} (365 nm, spectrophotometry), AÅ_{WS-BrC} (300–400 nm), MAC_{EC} (632 nm, thermal-optical transmission (TOT), Sunset OC-EC analyzer) and AÅ_{EC} = 1.

^bUsing AÅ_{WS-BrC}, MAC_{BC} (632 nm), AÅ_{BC} = 1 and a corrected factor of 2 for MAC_{WS-BrC} (365 nm).

Note for site acronyms: BTH (Wuqing district of Tianjin, China), YRD (Haining, China), PRD (Zhongshan, China), SC (Deyang in Sichuan Basin, China) and KCOG (Korea Climate Observatory at Gosan).

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