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研究課題名（和文）Light absorption properties of brown carbon aerosols in East Asia and source apportionment

研究課題名（英文）Light absorption properties of brown carbon aerosols in East Asia and source apportionment

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研究成果の概要（和文）：ブラウンカーボン（BrC）エアロゾルは、大気環境や気候変動を影響を及ぼすが、東アジア域の動態と発生源について未だよくわかっていない。本研究では、長崎県福江島において、現場観測とスカイラジオメータ観測を組み合わせて、BrCエアロゾルの光吸収特性や発生源を評価した。フィルター観測から得られたBrCの光吸収オングストローム指数AAEは、スカイラジオメータ観測から得られたものと正のSpearman相関（相関係数 0.77, p < 0.1）を示した。大気輸送モデルと衛星による火災ホットスポットを用いて、中国東部の化石燃料燃焼と野外バイオマス燃焼が高BrCイベントと関連していることがわかった。

研究成果の学術的意義や社会的意義
東アジア諸国の急速な経済成長に伴い、微小粒子状物質（PM2.5）が大量に排出されている。PM2.5中のBrCは日本や北太平洋西部に多く運ばれ、大気質の悪化やエアロゾルの化学組成を変化させる。なお、中国は、化石燃料から持続可能な資源へのエネルギー転換を進めており、今後、BrCの排出量が増加すると予測されている。また、BrCはロシアの森林火災からも排出される可能性があり、後者は2100年にはより頻繁に発生すると予測されている。この研究の結果は、東アジアにおけるBrCエアロゾルの動態と気候への影響の解明に役立つと考えられる。

研究成果の概要（英文）：Brown carbon aerosol, is one of the most understudied aerosol components for its sources and effects on climate change. Light-absorption properties of BrC aerosols were evaluated based on filter observation followed by analyses in the laboratory and SKYNET observation. After eliminating the contributions of black carbon, the absorption Angstrom exponent of BrC alone obtained from filter observations had a positive Spearman correlation (coeffcient = 0.77, p < 0.1) with that derived from sky radiometer observations but 33% higher values, indicating that the light-absorption properties of BrC were successfully captured using the two methods. Using the atmospheric transport model FLEXPART and fire hotspots obtained from the Visible Infrared Imaging Radiometer Suite product, a high-BrC event was identified, which is related to an air mass originating from regions with consistent fossil fuel combustion and sporadic open biomass burning in central East China.

研究分野：大気化学

キーワード：有機エアロゾル　物質循環　林野火災　大気輸送モデル

１．研究開始当初の背景 (Background)

Light-absorbing organic aerosol, also termed as brown carbon (BrC) aerosol, is one of the most understudied aerosol components for its sources and effects on climate change. For a long time, organic aerosols had been deemed to cause cooling on the earth's surface. Recently model studies indicated that BrC is accounting for ~1/4 of warming effect by carbonaceous aerosols at the tropopause globally. However, observation about the light absorption properties of BrC aerosols, which are fundamental for climate change prediction, is very limited, especially in East Asia.

The rapid growth of economy in East Asian countries such as China is accompanied by large emissions of fine particulate matter ($PM_{2.5}$). BrC in $PM_{2.5}$ is frequently transported to Japan and the western North Pacific, deteriorating air quality and altering aerosol chemical composition. China is promoting shift of energy use from fossil fuel to sustainable resources, to which BrC emissions are predicted to increase in the coming years. BrC could also be emitted from crop residue burning in China and forest fires in Russia, the latter is predicted to occur more frequently in 2100. However, so far it is under-examined on the temporal variations of BrC aerosols in Japan and where are they coming from.

２．研究の目的 (Purpose)

In this study, the light-absorption properties of BrC are investigated by combining in-situ filter measurements and sky radiometer observations on Fukue Island, western Japan, an outflow site of the Asian continent. We report important light-absorption properties of BrC, such as absorption Angstrom exponent (AAE) obtained from both filter observations and sky radiometer measurements. We also report potential sources of BrC using the atmospheric Lagrangian dispersion model FLEXPART together with fire hotspots obtained from the Visible Infrared Imaging Radiometer Suite (VIIRS) satellite product. The purposes of the study were to unveil the temporal variations of light absorption properties of BrC aerosols combining filter- and sky radiometer measurement (based on SKYNET observation network), and to identify the sources of BrC aerosols.

３．研究の方法 (Methods)

(1) Filter-based observations

Ambient $PM_{2.5}$ samples were collected using a continuous $PM_{2.5}$ mass and elemental concentration monitor (PX375, Horiba Ltd., Kyoto, Japan), which was deployed for the observation of particulate matter and elemental composition. Filter spots were extracted using 10 ml methanol (HPLC grade, Fujifilm Wako Corp., Japan) under ultrasonication for 30 min with brief intermittent manual shaking of the extraction vial. To remove BC and other relatively large particles such as dust that might interfere with the light absorption measurement of BrC, the extracts were then filtered through a syringe filter with a diameter of 13 mm and pore size of 0.2 μm (polytetrafluoroethylene for organics, Shimadzu Ltd., Japan). The light absorption of the extracts was then measured using a ultraviolet–visible spectrometer (U-2910, Hitachi Ltd., Japan) over the wavelength range of 200–900 nm with a resolution of 1 nm. The obtained liquid-phase absorption (A_λ) was corrected for instrumental detection bias by subtracting the mean absorption at 795–805 nm and for the field blanks. The corresponding Abs_λ of BrC was calculated using A_λ in the ultraviolet–visible light region (300–600 nm) as

$Abs_\lambda = A_\lambda \times V_l / (V_a \times L) \times \ln(10)$, (1)

where V_l (m^3) is the volume (10 ml) of methanol used for extraction, V_a is the volume of sampled air (1 m^3 h^{-1} by 8 hours for two spots in a daily sample), L is the optical path length (0.01 m), and the factor ln (10) is used to convert to the natural log for consistency with the atmospheric conditions (Hecobian et al., 2010; Xie et al., 2019). A typical curve of *Abs* of the methanol extract in the wavelength range of 300–600 nm is shown for March 2, 2018 in Fig. 1a. The figure shows that *Abs* had a clear wavelength dependence with higher absorbances at shorter wavelengths. *Abs* at a wavelength of 365 nm (Abs_{365}, mean value of 361–370 nm) was used as a representative light-absorption coefficient for BrC. The dependency of BrC light absorption on the wavelength based on the filter extraction method, $AAE_{BrC\text{-}filter}$, is determined as the slope of the linear fit of log10 (Abs_λ) versus log10 (λ). This study estimates $AAE_{BrC\text{-}filter}$ in the wavelength range of 340–500 nm.

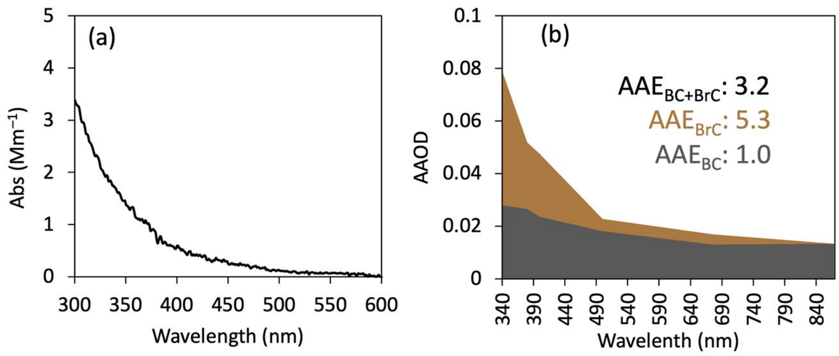

Fig. 1. Example light-absorption properties of BrC showing a dependence on wavelength, evaluated as (a) the light-absorption coefficient of methanol extracts of filter samples collected on March 2, 2018 and (b) the AAOD derived from sky radiometer observations made on January 20, 2018. The contributions of BrC were derived by assuming an AAE value of 1.0 for BC.

(2) Ground remote-sensing observations

Ground-based remote-sensing observations of aerosol optical properties were conducted using a sky radiometer, which was integrated with the SKYNET observation network. The aerosol optical depth (AOD), single-scattering albedo (SSA), Angstrom exponent (AE), volume size distribution, and refractive index were retrieved at 340, 380, 400, 500, 675, and 870 nm in 2018, using the sky radiometer analysis package of the Center for Environmental Remote Sensing (SR-CEReS) version 1. To reduce uncertainty, the retrieved data were first screened to keep only those data flagged as being unaffected by cloud and having a solar zenith angle of at least 50° and then screened to keep only those data for which the SSA was less than 0.95. The AOD and SSA data were screened to keep only those larger than their uncertainties, which were assumed as 0.01 and 0.05, respectively. The AAOD over the whole column was then derived as

$AAOD_\lambda = AOD_\lambda \times (1 - SSA_\lambda)$. (2)

This AAOD, however, incorporates contributions from all light-absorbing particles. The corresponding AAE, denoted AAETotal-SKYNET, was then estimated as the slope of the linear fit of log10 ($AAOD_\lambda$)

versus log10 (λ) in the wavelength range of 340–870 nm. Of the study period, the contributions of dust to the AAOD are assumed to be negligible. Thus, the AAOD can be assumed to be the summation of the contributions of BrC and BC:

$$AAOD_{\lambda} = AAOD_{\lambda\text{–}BrC} + AAOD_{\lambda\text{–}BC}. \quad (3)$$

A typical example of AAOD attribution is shown in Fig. 1b. Light absorption at wavelengths longer than 700 nm is known to be mostly contributed by BC. A common value of 1.0 for the AAE of BC (AAE_{BC}) has been used in fractionation studies of carbonaceous aerosols. In this study, assuming a value of 1.0 for AAEBC and a negligible contribution of BrC to $AAOD_{870\ nm}$, $AAOD_{\lambda\text{–}BC}$ at wavelengths shorter than 870 nm can be extrapolated as

$$AAOD_{\lambda\text{–}BC} = AAOD_{870\ nm} \times (\lambda/870)\ \exp\ (–AAE_{BC}). \quad (4)$$

Summarizing equations (3) and (4), $AAOD_{\lambda\text{–}BrC}$ is written as

$$AAOD_{\lambda\text{–}BrC} = AAOD_{\lambda} - AAOD_{870\ nm} \times (\lambda/870)\ \exp\ (–AAE_{BC}). \quad (5)$$

The AAE contributed solely by BrC obtained by the sky radiometer ($AAE_{BrC\text{-}SKYNET}$) is then determined as the slope of the linear fit of log10 ($AAOD_{\lambda\text{–}BrC}$) versus log10 (λ) in the wavelength range of 340–500 nm. To further reduce uncertainty, light-absorption properties based on sky radiometer observations were calculated as daily means for the period 9:00–15:00 LT.

４．研究成果 (Results)

(1) Light absorptions obtained in filter and sky radiometer measurements

$AAE_{BrC\text{-}filter}$ had a variation consistent with that of $AAE_{BrC\text{-}SKYNET}$ (Fig. 2). This agreement results from the strict screening of SKYNET retrieved data, with the potential effects of dust and other uncertainties during measurement largely being eliminated. Interestingly, $AAE_{BrC\text{-}filter}$ is 33% higher than $AAE_{BrC\text{-}SKYNET}$. This difference might relate primarily to the nature of the evaluation methods. $AAE_{BrC\text{-}filter}$ reflects the

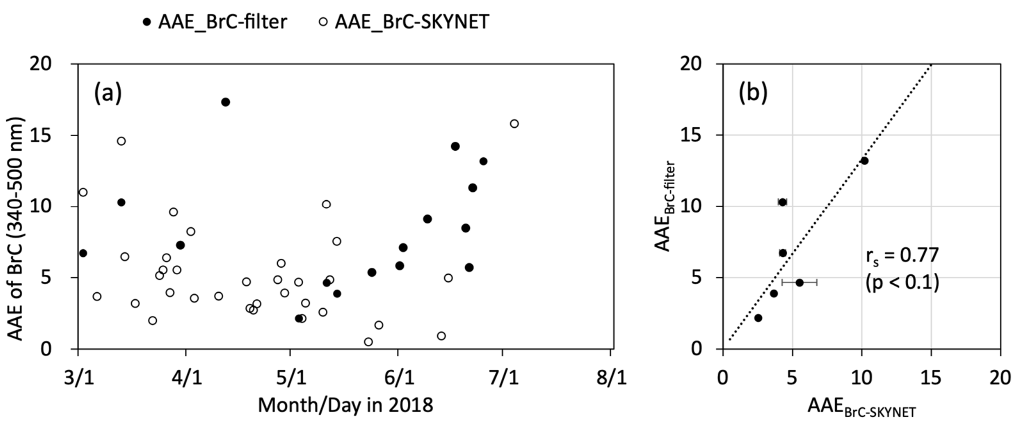

Fig. 2. $AAE_{BrC\text{-}filter}$ and $AAE_{BrC\text{-}SKYNET}$ contributed by BrC estimated for the wavelength range of 340–500 nm: (a) temporal variations in March–July 2018 and (b) correlations. The error bars in panel (b) are estimated by assuming that AAEBC has a range of 0.8–1.4. The dotted line shows the linear relations in panel (b), while r_s represents Spearman correlation coefficient as the data follow non-normal distribution.

wavelength dependence of particle absorption near the ground while $AAE_{BrC\text{-}SKYNET}$ incorporates that in the whole atmospheric vertical column. If particles with less light-absorbing potential at shorter wavelengths are dominant in higher layers of the column, $\mathrm{AAE_{BrC\text{-}SKYNET}}$ lower than $\mathrm{AAE_{BrC\text{-}filter}}$ would be estimated.

The difference between $AAE_{BrC\text{-}filter}$ and $AAE_{BrC\text{-}SKYNET}$ might also be attributed to the assumption that AAE_{BC} = 1.0 when estimating $AAE_{BrC\text{-}SKYNET}$. AAE_{BC} varies in the range of 0.8–1.4 depending on the source section, particle size, and mixing state. If the plume detected by the sky radiometer contains BC with an AAE lower than 1.0, then AAE_{SKYNET} would be underestimated.

(2) Sources of BrC during a high-BrC event

A high-BrC event was identified with elevations of light-absorbing properties in the period March 28–30, 2018. During this event, high levels of average Abs (0.80 Mm^{-1}, >70% percentile level for March–July, same for the following), $AAOD_{340nm}$ (0.12), $AAE_{BrC\text{-}filter}$ (7.29), and $AAE_{BrC\text{-}SKYNET}$ (7.58) were observed, along with a high BC concentration (0.46 μg m^{-3}). Meanwhile, the volume size distribution had a fine aerosol size prevalence peaking at <1 μm for the event. The air mass footprint of Fukue Island during this event originated from the North China Plain, where emissions from anthropogenic sources, such as the domestic burning of biomass and biofuels and fossil-fuel combustion, are discernable (Fig. 3). Meanwhile, fire hotspots were also observed in the region. These results suggest that besides anthropogenic emissions, open biomass burning on the North China Plain is an important source of high BrC in the outflow region. As the next step, further investigations would be placed to track more evidence of BrC sources, such as using molecular organic tracers of anthropogenic sources and biomass burning.

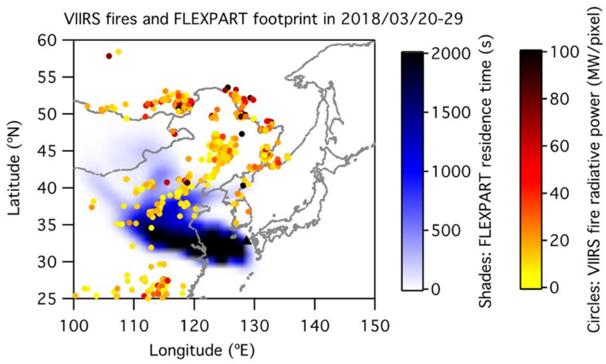

Fig. 3. Footprint of a typical high-BrC event calculated using the FLEXPART residence time during March 20–29, 2018. VIIRS fire hotspots along with fire radiative power in the same period are shown as circles. The location of the Fukue observatory is marked as triangle. Light absorption properties of BrC based on both filter samples and ground remote-sensing observations were conducted at the same location.

(3) Summary

We evaluated the light-absorption properties of BrC on Fukue Island through aerosol filter observations and sky radiometer observations. $AAE_{BrC\text{-}filter}$ and $AAE_{BrC\text{-}SKYNET}$ of BrC had a positively linear correlation, while AAE_{filter} was 33% higher than AAE_{SKYNET}. This result shows that light-absorption properties obtained from sky radiometer observations can be converted to surface values. In the future, it is recommended to investigate the vertical distributions of aerosol light absorption properties along with size ranges. A high-BrC event on March 28–30, 2018 was identified, where air masses originated from the North China Plain with discernable open biomass burning. The results of the study help to clarify the dynamics and sources of BrC in East Asia.

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オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

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〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関