

Source-Dependent Brown Carbon Optical Indicators in Philippine Emission Filters

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Abstract

Multiwavelength filter analysis was used to compare source-dependent light-absorption in 150 archived Philippine emission and near-source filters. After blank correction and quality control, 128 filters spanning seven source groups were retained: charcoal, fuelwood, tricycle, jeepney, light-duty gasoline and diesel vehicles, and volcanic aerosol. Absorption was measured at 405nm (violet-blue) and 639 nm (red), and spectral contrast was quantified through the 405/639 absorption ratio, the two-point absorption ratio ($AAE_{405-639}$), and a nonnegative brown-carbon (BrC) proxy defined as the excess 405-nm BC-equivalent response over the 639-nm. All indicators differed significantly across groups (Kruskal-Wallis, $p < 0.001$). Charcoal showed the highest median AAE (2.67), followed by tricycle (1.69) and volcanic aerosol (1.37); fuelwood and jeepney medians were near zero. A stratified 10% holdout over 1,000 iterations preserved source ordering with $p < 0.05$ throughout. The results support continuous multiwavelength optical indicators for comparative screening of filters by source groups.

1 Introduction

Light-absorbing carbon-containing aerosols affect atmospheric heating, visibility, and exposure. Black carbon (BC) absorbs strongly across the visible spectrum, whereas brown carbon (BrC) generally shows stronger absorption at shorter wavelengths (Andreae and Gelencsér 2006; Bond and Bergstrom 2006; Laskin et al. 2015). Because optical response also depends on particle properties, filter substrate, and measurement method, BC-equivalent and BrC-proxy values require clear definitions of how they were calculated (Petzold et al. 2013; Alang and Aggarwal 2024).

Filter-based multiwavelength instruments allow rapid, non-destructive comparison of archived samples using the same multiwavelength approach. However, filter loading, multiple scattering, and wavelength- and substrate-dependent mass absorption cross-sections can affect attenuation and BC-equivalent estimates (Weingartner et al. 2003; Pandey et al. 2019; Presler-Jur et al. 2017). Spectral slopes and wavelength ratios are therefore useful alongside absolute absorption values.

The absorption Ångström exponent (AAE) summarizes the wavelength dependence of absorption. Values near unity are often associated with BC-dominated combustion aerosol, while larger values can indicate enhanced short-wavelength absorption. No universal AAE threshold separates BC from BrC because AAE depends on wavelength range, source mixture, instrument response, and atmospheric processing (Lack and Langridge 2013; Zotter et al. 2017). Biomass-burning aerosol also varies with fuel, combustion phase, and BC content (Saleh et al. 2014; Xie et al. 2017).

Philippine aerosol is influenced by traffic, combustion, marine aerosol, and regional transport. Metro Manila studies have reported spatially variable BC and vehicle emission factors (Alas et al. 2018; Madueño et al. 2019), while receptor studies show mixed local and regional particulate sources (Pabroa et al. 2022, 2024). Few studies have compared archived Philippine source and near-source filters within one multiwavelength optical framework.

this study evaluates optical indicators in archived charcoal, fuelwood, tricycle, jeepney, light-duty vehicle, and volcanic filters. The objectives were to harmonize the archive and apply reproducible quality control, compare 405- and 639-nm absorption behavior, test source-group differences, and assess the stability of the main source ordering using repeated stratified holdout. The analysis is intended for comparative optical screening rather than chemical identification or unique source attribution.

2 Data and Methods

2.1 Filter archive and sampling contexts

The archive contained 150 47-mm quartz-fiber filters from Philippine field and controlled-emission activities involving charcoal, fuelwood, tricycles, jeepneys, light-duty gasoline and diesel vehicles, and volcanic aerosol near Mayon Volcano. Because the campaigns differed in sampling environment, source proximity, and metadata, comparisons were made within the same optical analysis and were not treated as emission factors for the wider source groups.

Tricycle exhaust was sampled along an approximately 5.9-km Quezon City route using an Airmetrics MiniVol at 5 L min⁻¹ for approximately 15 min per collection. Jeepney and light-duty filters came from vehicle exhaust projects, biomass filters from charcoal and fuelwood combustion tests, and volcanic filters from the 2018 Mayon activity period. Jeepney and light-duty diesel records were retained as separate categories where project metadata allowed. Sampling contexts are shown in Online Resource 1 (Figs. S1-S3).

Table 1
Filter archive before and after quality control

Source category	Sampling context	Available before QC (n)	Retained after QC (n)
Charcoal	Controlled combustion	9	9
Fuelwood	Controlled combustion	9	9
Tricycle	In-use route sampling	68	53
Gasoline light-duty vehicle	Vehicle exhaust sampling	8	2
Diesel transport, combined	Jeepney and diesel light-duty vehicle sampling	33	32 (30 jeepney, 2 diesel LDV)
Volcanic aerosol	Near-source ambient sampling	23	23
Total		150	128

2.2 Multiwavelength absorption measurements

Filters were analyzed using a Multi-wavelength Absorption Black Carbon Instrument (MABI) at 405, 465, 525, 639, 870, 940, and 1050 nm (Manohar et al. 2021; Atanacio et al. 2020). The 639-nm channel served as the operational BC anchor for comparison with 405 nm because wavelength-dependent organic absorption is generally weaker at longer visible wavelengths. The 639-nm response was not treated as chemically pure BC.

For wavelength λ , attenuation was calculated as

$$ATN_{\lambda} = 100 \ln \left(\frac{I_{0,\lambda}}{I_{\lambda}} \right)$$

where $I_{0,\lambda}$ is the transmitted intensity through the blank or reference filter and I_{λ} is the intensity through the exposed filter. The absorption coefficient was calculated as

$$b_{abs,\lambda} = 10^2 \frac{A}{V} \ln \left(\frac{I_{0,\lambda}}{I_{\lambda}} \right)$$

where A is the analyzed deposit area (cm^2), V is sampled air volume (m^3), and $b_{abs,\lambda}$ is expressed in Mm^{-1} (Atanacio et al. 2020).

Absorption was converted to a wavelength-specific BC-equivalent concentration using the archive-processing mass absorption efficiency:

$$BCeq_{\lambda} = \frac{1000 b_{abs,\lambda}}{\epsilon_{\lambda}}$$

where ϵ_{λ} is the wavelength-specific mass absorption efficiency ($\text{m}^2 \text{g}^{-1}$) and $BCeq_{\lambda}$ is expressed in ng m^{-3} . $BCeq$ is used only for comparison processes using the same method.

Wavelength-specific mass absorption efficiency coefficients calculated by M.G. Cayetano were applied consistently during archive processing. The coefficient values used are provided in Online Resource 1 (Table S6), while the detailed calculation method has not yet been published. Because quartz-specific calibration was unavailable, the resulting BC-equivalent and BrC-proxy values are used only for comparison and are not interpreted as direct carbon mass.

2.3 Optical indicators

AAE between 405 and 639 nm was calculated as

$$AAE_{405-639} = - \frac{\ln(b_{abs,405}/b_{abs,639})}{\ln(405/639)}$$

The normalized 405/639 absorption ratio was retained as a direct measure of short- to long-wavelength absorption:

$$R_{405/639} = \frac{b_{abs,405}}{b_{abs,639}}$$

Spectra were normalized to 639 nm to compare spectral shape independently of absolute loading:

$$b_{abs,\lambda}^* = \frac{b_{abs,\lambda}}{b_{abs,639}}$$

AAE405–639 was calculated from the original blank-corrected absorption coefficients at 405 and 639 nm. Normalization to 639 nm was used only to compare the shape of the measured spectra.

The operational BrC proxy was defined as the nonnegative excess of the 405-nm over 639-nm BC-equivalent response:

$$BrC\ proxy = \max(BCeq_{405} - BCeq_{639}, 0)$$

The proxy is reported in ng m⁻³ BC-equivalent and is not a chemically resolved BrC mass concentration. Values were set to zero when BCeq₄₀₅ did not exceed BCeq₆₃₉.

2.4 Harmonization and quality control

Project-specific blanks were used when available; otherwise, a designated reference blank was applied. Replicate scans and duplicate MABI entries for the same sample code were averaged before source-group analysis and were not counted as independent filters.

Air volume was calculated from recorded flow and duration when needed. A 15-min fallback was used only for documented short vehicle runs with missing or zero duration. Filters were excluded for nonphysical blank-corrected transmission, missing required 405- or 639-nm values, nonpositive absorption needed for logarithms or ratios, or insufficient exposure metadata. These rules retained 128 of 150 filters (Table 1).

No adjustment was applied to equalize source-group sizes or campaign conditions. Gasoline and diesel light-duty categories (n = 2 each after QC) were retained for descriptive comparison but were not used to represent the wider vehicle fleet.

2.5 Statistical analysis and stability check

Because source-group distributions were non-normal and unequal in size, results are summarized using medians, interquartile ranges (IQRs), and observed ranges. Group differences in AAE405–639, the BrC proxy, babs₄₀₅, and the normalized 405/639 ratio were tested using Kruskal-Wallis tests, followed by Holm-adjusted pairwise comparisons. Spearman's rank correlation assessed the association between AAE and the BrC proxy. Statistical significance was evaluated at α = 0.05.

Stability was assessed with 1,000 stratified holdout iterations. In each iteration, 10% of observations from charcoal, fuelwood, tricycle, jeepney, and volcanic groups were withheld, and group medians and

the Kruskal-Wallis test were recalculated. The n = 2 light-duty groups were excluded from holdout analysis. We recorded how often $p < 0.05$ was retained and whether the source-group median ordering remained the same.

3 Results and Discussion

3.1 Retained samples and source-group optical metrics

Quality control retained 128 filters: tricycle (n = 53), jeepney (n = 30), volcanic aerosol (n = 23), charcoal (n = 9), fuelwood (n = 9), and two filters each from gasoline and diesel light-duty vehicles (Table 1). The light-duty results are descriptive because of their small sample sizes.

Source groups differed in both absorption magnitude and spectral shape (Table 2). Charcoal showed the strongest wavelength dependence, with the highest median AAE_{405–639} (2.67) and normalized 405/639 ratio (3.37), followed by tricycle (AAE = 1.69; normalized ratio = 2.17) and volcanic aerosol (AAE = 1.37; normalized ratio = 1.87). Fuelwood, jeepney, and diesel light-duty filters had median ratios near 1.0, indicating nearly wavelength-neutral absorption between 405 and 639 nm.

Table 2
Median source-group optical indicators after quality control

Source	n	$b_{abs,405}$ (Mm ⁻¹)	$b_{abs,639}$ (Mm ⁻¹)	Norm. 405/639 ratio	AAE _{405–639}	BrC proxy (ng m ⁻³ BC-equivalent)
Charcoal	9	3,122.97	902.49	3.37	2.67	237,101.87
Fuelwood	9	22,193.66	22,662.84	0.99	-0.03	0
Tricycle	53	1,478.97	692.71	2.17	1.69	74,471.54
Jeepney	30	20,543.47	20,608.82	0.99	-0.03	0
Gasoline light-duty vehicle	2	9,270.00	5,051.86	1.56	0.92	449,360.36
Diesel light-duty vehicle	2	20,441.47	21,080.50	0.97	-0.07	0
Volcanic aerosol	23	2,219.16	1,220.06	1.87	1.37	116,826.80

Absolute absorption and spectral dependence did not always vary together. Fuelwood and jeepney filters had high median absolute absorption but similar 405- and 639-nm responses, whereas charcoal showed substantially stronger absorption at 405 nm. Reporting both absolute and 639-nm-normalized spectra therefore separates filter loading from spectral shape.

3.2 Absolute and normalized absorption spectra

Median absolute spectra varied markedly among source groups (Fig. 1). Fuelwood and jeepney filters had the highest median absorption across much of the measured spectrum, while tricycle and volcanic filters were lower. These values describe the archived filter loadings and are not direct emission-factor comparisons because sampling geometries and conditions differed among campaigns.

After normalization to 639 nm, charcoal and tricycle showed the largest increase toward 405 nm, with volcanic aerosol showing a smaller increase (Fig. 2). Fuelwood, jeepney, and diesel light-duty spectra remained close to 1.0 between 405 and 639 nm. Median normalized values across all measured wavelengths are provided in Online Resource 1 (Table S1), while the operational BC-baseline visualization is shown in Fig. S4.

The charcoal pattern is consistent with short-wavelength absorption reported for some biomass-burning organic aerosol, although combustion phase and BC content strongly affect this behavior (Saleh et al. 2014; Xie et al. 2017). The nearly wavelength-neutral fuelwood median shows that a broad biomass-burning label does not imply uniformly high AAE. Transport filters also differed: tricycle spectra were steeper than jeepney and diesel light-duty spectra, potentially reflecting differences in fleet technology, maintenance, fuel-lubricant mixtures, and sampling conditions (Madueño et al. 2019).

3.3 AAE and operational BrC proxy

AAE distributions followed the normalized spectra (Fig. 3): charcoal had the highest median, followed by tricycle and volcanic aerosol, while fuelwood and jeepney medians were near zero. Gasoline and diesel light-duty distributions are shown for completeness but are not interpreted at the fleet level because $n = 2$. Median, interquartile range, and observed range for each source group are provided in Online Resource 1 (Table S2).

The BrC proxy showed a similar broad ordering. Median values were 237,101.87 ng m⁻³ BC-equivalent for charcoal, 116,826.80 for volcanic aerosol, and 74,471.54 for tricycle; fuelwood, jeepney, and diesel light-duty medians were zero. The gasoline median was high but unstable because it was based on two filters. Full distributions are reported in Online Resource 1 (Table S3).

AAE_{405–639} and the BrC proxy were strongly correlated (Spearman's $\rho = 0.801$, $p < 0.001$). The relationship is expected because both use the 405- and 639-nm responses, but the indicators are not equivalent: AAE describes spectral slope, whereas the BrC proxy also depends on absolute filter loading and wavelength-specific conversion coefficients.

Samples were not assigned to fixed AAE classes because published thresholds vary with wavelength interval, source mixture, and measurement platform, and overlapping BC and BrC absorption can bias AAE-based attribution (Lack and Langridge 2013; Zotter et al. 2017). Continuous values were therefore retained.

3.4 Group tests and holdout stability

All primary indicators differed among source groups (Kruskal-Wallis, $p < 0.001$; Table 3). AAE and the normalized 405/639 ratio produced the same rank ordering because both are based on the same fixed wavelength pair. Holm-adjusted comparisons mainly separated charcoal and tricycle, which showed stronger wavelength dependence, from the nearly wavelength-neutral fuelwood and jeepney groups. Significant pairwise differences are listed in Online Resource 1 (Table S4).

Table 3
Overall source-group tests and repeated holdout results

Analysis	Metric or outcome	Result
Kruskal-Wallis, all retained groups	AAE _{405–639}	$H = 74.36, p < 0.001$
Kruskal-Wallis, all retained groups	BrC proxy	$H = 57.27, p < 0.001$
Kruskal-Wallis, all retained groups	$b_{abs,405}$	$H = 80.65, p < 0.001$
Kruskal-Wallis, all retained groups	Normalized 405/639 ratio	$H = 74.36, p < 0.001$
Spearman correlation	AAE versus BrC proxy	$\rho = 0.801, p < 0.001$
Kruskal-Wallis, five major groups	Full-data BrC proxy comparison	$H = 53.61, p < 0.001$
Stratified 10% holdout, 1,000 iterations	Iterations with $p < 0.05$	1,000/1,000
Stratified 10% holdout, 1,000 iterations	Highest median BrC proxy	Charcoal in 1,000/1,000 iterations

The repeated holdout analysis preserved $p < 0.05$ in all 1,000 iterations, and charcoal retained the highest median BrC proxy throughout. Fuelwood and jeepney medians remained zero. These results indicate that the main five-group ordering was not driven by a small subset of filters, although the procedure does not correct campaign imbalance or provide independent external validation. Holdout median ranges are summarized in Online Resource 1 (Table S5).

3.5 Interpretation and limitations

Charcoal and tricycle filters showed stronger short-wavelength absorption than fuelwood and jeepney filters, while volcanic samples showed values between these groups. The volcanic optical response should not be interpreted as direct evidence of volcanic BrC because ash, sulfate aerosol, metals, changes in the volcanic plume, and background particles may also contribute (Stewart et al. 2022). The observed patterns are therefore most useful for comparison among source groups and guiding further analysis.

Interpretation is limited by differences in sampling conditions, unequal group sizes, the wavelength range used for AAE, and effects related to the filter material. Filter loading, scattering, and the mass

absorption efficiency used for BC-equivalent conversion can affect optical values (Weingartner et al. 2003; Presler-Jur et al. 2017; Pandey et al. 2019). Because quartz-specific calibration and information on aerosol aging were unavailable, absolute BC-equivalent and BrC-proxy values should be compared only within this study and should not be assumed to represent aerosol after longer transport and aging in the atmosphere.

The AAE, the normalized 405/639 ratio, and normalized spectra can identify filters with enhanced short-wavelength absorption, while the BrC proxy adds a loading-dependent indicator. Future work should combine these optical indicators with chemical markers, combustion metadata, substrate-specific calibration, and validation using more balanced source groups.

4 Conclusions

Multiwavelength analysis of 128 quality-controlled Philippine filters showed clear source-dependent differences in absorption magnitude and spectral shape. Charcoal had the steepest median 405- to 639-nm slope, followed by tricycle and volcanic samples, while fuelwood and jeepney medians were nearly wavelength neutral. All primary indicators differed among source groups ($p < 0.001$), and repeated stratified holdout preserved the main ordering in all 1,000 iterations. These indicators support comparative screening of filters from different source groups but do not provide chemical BrC identification or unique source attribution. Their use requires explicit reporting of wavelength, filter, calibration, and BC-equivalent assumptions.

Declarations

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Competing interests

The authors have no relevant financial or non-financial interests to disclose.

Ethics approval

Not applicable. The study analyzed archived particulate matter filter samples and did not involve human participants or animals.

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Author Contribution

K.E.S. and M.G.C. conceptualized the study and developed the methodology. K.E.S. curated the data, performed the formal analysis, prepared the figures, and wrote the original manuscript. M.G.C. supervised and validated the study and reviewed and edited the manuscript. Both authors reviewed and approved the final manuscript.

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Data Availability

The processed data supporting the findings of this study are not publicly available because the archived filter measurements and associated records originated from multiple research projects with data-use and permission requirements. The data are available from the corresponding author upon reasonable request and with permission from the relevant originating project(s).

Code availability

The Python notebooks used for quality control, statistical analysis, sensitivity testing, and figure preparation are available from the corresponding author upon reasonable request.

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Figures

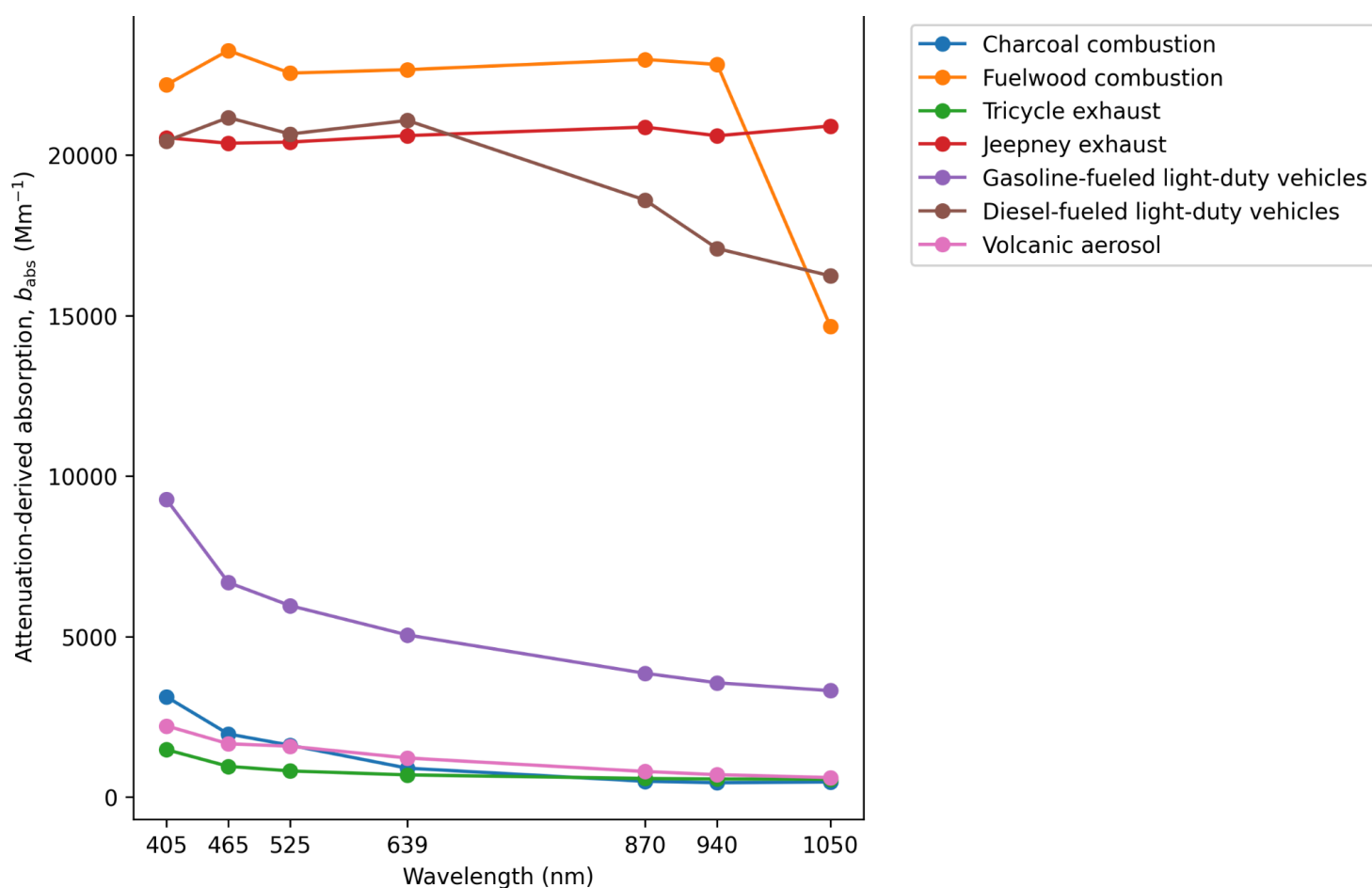


Figure 1

Median multiwavelength absorption coefficients by source group, showing the large difference in absolute filter loading among campaigns

Supplementary Files

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