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Long-Term Variation of Black Carbon Absorption Aerosol Optical Depth from AERONET Data over East Asia

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Abstract: Absorption aerosol optical depth induced by black carbon ($AAOD_{BC}$) was retrieved using the depolarization ratio and single scattering albedo provided by the Aerosol Robotic Network (AERONET) inversion products over East Asia. Our analysis considered AERONET data from six sites in East Asia that are mostly affected by anthropogenic pollution, black carbon (BC) emissions, and natural mineral dust, during the period 2001–2018. We identified a rapid reduction in total aerosol optical depth (AOD_T) of -0.0106 yr^{-1} over Beijing, whereas no considerable trend was observed at the Korean and Japanese sites. The long-term data for $AAOD_{BC}$ showed decreasing trends at all sites. We conclude that successful emission control policies were the major underlying driver of AOD_T and $AAOD_{BC}$ reductions over East Asia, particularly in China, during the study period. Values of the $AAOD_{BC}/AOD_T$ ratio revealed that, although these policies were successful, the Chinese government needs to undertake stricter measures toward reducing BC emissions. We found that $AAOD_{BC}$ follows seasonal trends, peaking in the colder months. This suggests that in East Asia, particularly in China, domestic coal burning is still of concern.

Keywords: absorption aerosol optical depth (AAOD); black carbon (BC); depolarization ratio (DPR); single scattering albedo (SSA); Asian dust

1. Introduction

Atmospheric aerosols can greatly affect the global climate by scattering and absorbing solar radiation and influencing cloud formation [1,2]. Population growth, urbanization, and economic development have considerably increased anthropogenic emissions of aerosols [3]. The incomplete combustion of fossil fuels, biomass, and carbon-containing substances emits black carbon (BC) aerosol particles, particularly in developing regions such as East Asia [4], which is one of the main source regions of not only BC and other anthropogenic pollutants, but also natural mineral dust [5–7]. China, in particular, is a major source of BC because of its large population and high fossil fuel consumption [8]. Indirect effects of BC include changing the microphysical properties of clouds, and its direct effects include temperature increase through light absorption [9–11]. High atmospheric aerosol levels over East Asia, including BC, affect not only local air quality but also the radiative balance of downwind areas [12]. Despite the availability of global atmospheric aerosol information, there is little data observed for more than 10 years available for BC concentrations in East Asia. Furthermore, there is no record of the impact of BC's light-absorption on aerosol optical depth (AOD) and its trends over time. The Aerosol Robotic Network (AERONET) is a global network of ground-based sun/sky photometers that provide long term data on aerosols, such as depolarization ratio (DPR), spectral AOD,

column single scattering albedo (SSA), particle size distribution, and absorption AOD (AAOD). Overall, AAOD defines the column aerosol loading of light-absorbing particles such as BC, carbon-containing particles, or mineral dust. Calculation of AAOD is complicated when different types of absorbing aerosol are present in a mixed aerosol plume.

Because BC is the main light absorber in the atmosphere, and also affects atmospheric chemical processes and air quality, the quantification of its specific AAOD ($AAOD_{BC}$) is critical [13]. Although several studies have investigated the contribution of BC to aerosol light absorption from AERONET columnar observations [14], few studies have determined $AAOD_{BC}$ and its optical properties. Schuster et al. [15] derived the BC concentrations and the specific absorption from AERONET measurements to describe the radiance field and absorption optical thickness. Russell et al. [13] applied SSA, absorption Ångström exponent, and AAOD from AERONET data to identify the individual shares of BC, mineral dust, and organic matter in the total AAOD. Shin et al. [14] discussed the contribution of BC-related absorption to the non-dust AAOD. To the best of our knowledge, this is the first study in which the contribution of $AAOD_{BC}$ to the total AOD has been studied over a period of 18 years in East Asia.

In this study, we retrieved the $AAOD_{BC}$ from AERONET version 3 level 2.0 data using DPR and SSA to identify its long-term trends and seasonal variations in East Asia. The work is beneficial to the analysis of areas of (and downwind of) deserts with notable contributions of dust to AAOD. Section 2 describes our methodology; Section 3 provides the results; Section 4 describes the discussion; Section 5 presents a summary and conclusion of this study.

2. Materials and Methods

2.1. AERONET Version 3 Data and Sites

AERONET sun/sky radiometers measure direct solar and sky radiation. The measurements are analyzed automatically by the AERONET inversion algorithm [16,17]. All AERONET data are stored in an online database (<http://aeronet.gsfc.nasa.gov>, AERONET, 2019) and the measurements are downloadable. AERONET instruments measure AOD at 440, 670, 870, and 1020 nm. In the absence of cloud contamination, the AOD uncertainty is estimated as 0.01 to 0.02 depending on wavelength. Additionally, the calibrated sky radiance measurements have uncertainties below 5%. AERONET inversion code provides aerosol optical properties in the total atmospheric column derived from the direct and diffuse radiation measured by AERONET sun/sky radiometers. AERONET version 3 level 2.0 inversion products provide DPRs at 440, 675, 870, and 1020 nm as well as AOD and other properties such as SSA. In addition, the outputs of inversion products provide estimates for random and possible systematic errors resulted from possible biases in measurements for most of the retrieved characteristics. SSA is used to determine the share of scattered light and the fraction of AOD related to light absorption (i.e., AAOD). The uncertainty in SSA was reported to be of the order of 0.03 [16].

This study selected AERONET sites with the following considerations. First, the sites must measure the optical properties of atmospheric aerosols generated in East Asia. Second, they must show variations in aerosol optical characteristics due to the mixing of dust and anthropogenic aerosol during long-range transport. Third, the sites should be able to provide long-term data on the variations in optical properties for at least 10 years. Figure 1 shows the locations of the six selected AERONET sites: one in China (Beijing, 39.977°N 116.381°E), three in Korea (Anmyeon, 36.539°N 126.330°E; Gwangju 35.228°N 126.843°E; and Gosan, 33.292°N 126.162°E), and two in Japan (Osaka, 34.651°N 135.591°E; and Shirahama, 33.693°N 135.357°E). Urbanization and industrialization in East Asia, specifically in China, have led to a vast emission of anthropogenic aerosols in this region. East Asian dust storms originating in the Asian desert also raise dust particles into the atmosphere. Readings at the selected sites are influenced by both anthropogenic (e.g., those from fossil fuels and biomass burning) and natural (e.g., mineral dust) aerosols [18]. The AERONET inversion algorithm is only performed for observations with AODs larger than 0.4 at 440 nm [17]. In this study, the data on particle linear DPR (PLDR), SSA, and size distribution is derived from the AERONET (level 2.0 (quality-assured), version

3) inversion products and the respective processing algorithms from 2001 to 2018 at the selected sites (<http://aeronet.gsfc.nasa.gov>, AERONET, 2019).

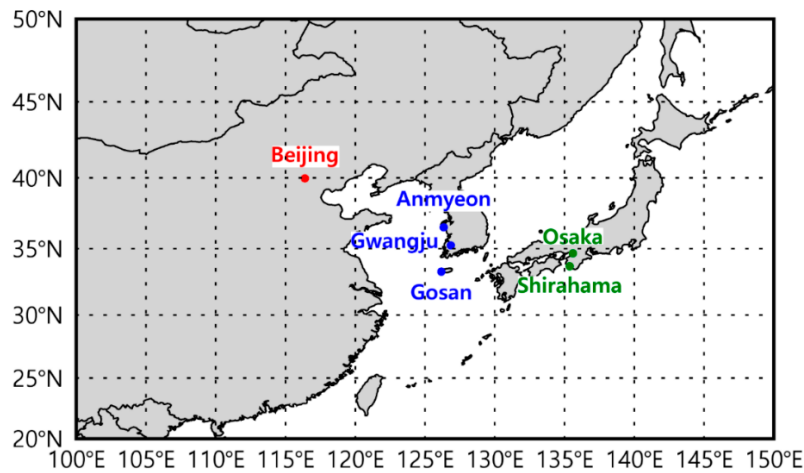


Figure 1. Locations of the six selected AERONET observation sites in East Asia.

2.2. $AAOD_{BC}$ Retrieval Methodology

Most aerosols in the atmosphere scatter light and absorb it in different levels, but BC has a strong light absorbing ability [19–22], as do dust particles [23–27]. In the absence of dust particles, BC is mainly responsible for the light absorbed by atmospheric aerosols, and BC's specific AAOD ($AAOD_{BC}$) can be calculated simply from the SSA:

$$AAOD_{BC,\lambda} = (1 - SSA_{\lambda}) \times AOD_{\lambda} \quad (1)$$

where, λ indicates wavelength. However, in the presence of dust particles, their effect must be separated to calculate $AAOD_{BC}$.

Previous researchers have found that the DPR measured by LIDAR can be used to distinguish between dust and pollution (non-dust) particles [28–30]. Noh et al. [31] confirmed that the DPR at 532 nm measured by LIDAR, and that at 1020 nm retrieved from AERONET data, were highly correlated. Therefore, we applied the method of Shin et al. [14] to separate dust and non-dust particles using the DPR at 1020 nm from AERONET data, and consequently calculate $AAOD_{BC}$ using only the SSA of pollution particles.

The particle linear DPR can be used as a parameter to obtain insight into the variation of optical and microphysical properties of particles. The AERONET version 3 level 2.0 inversion product provides particle linear DPRs (δ^p) at 440, 675, 870, and 1020 nm. Shin et al. [32] showed that any value of δ^p at 1020 nm greater than 0.30 represents pure Asian dust particles, while values below 0.02 indicate Asian anthropogenic pollution particles [33]. Values between these two thresholds (labeled δ_d^p and δ_{nd}^p , respectively) represent a mixture of dust and other aerosols such as anthropogenic pollution and non-dust particles. Therefore, the δ^p can be used to distinguish the share of pure dust and non-dust in a mixed plume [14,29]. The dust ratio (R_d) was first calculated here using δ^p values at 1020 nm from AERONET data. Shimizu et al. (2004) gave the following calculation for R_d [28]:

$$R_d = \frac{(\delta^p - \delta_{nd}^p)(1 + \delta_d^p)}{(\delta_d^p - \delta_{nd}^p)(1 + \delta^p)} \quad (2)$$

Noh et al. [31] concluded that the high correlation of δ^p at 1020 nm from AERONET version 3 inversion data, and that from LIDAR measurements, means that the AERONET data can be used to

assess the presence of dust particles. The value of R_d allows the total AOD (AOD_T) at 1020 nm to be separated into components for dust (AOD_D) and non-dust (AOD_{nd}):

$$AOD_{D,1020} = AOD_{T,1020} \times R_{d,1020} \quad (3)$$

where the subscript numbers denote the wavelength in nm. To convert $AOD_{D,1020}$ to corresponding values for other wavelengths, we used the Ångström exponent of pure Asian dust ($a_D = 0.14$) [30]:

$$AOD_{D,\lambda} = AOD_{D,1020} \times \left(\frac{1020\text{nm}}{\lambda} \right)^{a_D} \quad (4)$$

which can be used to calculate $AOD_{nd,\lambda}$:

$$AOD_{nd,\lambda} = AOD_{T,\lambda} - AOD_{D,\lambda} \quad (5)$$

Dark mineral phases like iron oxides influence the visible-light absorption of mineral dust [34–36], while BC is the main anthropogenic light absorber across the entire solar spectrum, especially in the mid-visible range [37,38]. The SSA spectral curvature can provide further information on the composition of aerosol mixtures. The SSA increases with increasing wavelength for pure mineral dust, but decreases for anthropogenic pollution or carbon-containing particles [39]. As dust and BC both absorb light in the visible range (around 440 nm), the total SSA (SSA_T) can be split into components for dust (SSA_D) and non-dust (SSA_{nd}) as follows:

$$SSA_{T,\lambda} = \frac{AOD_{D,\lambda}}{AOD_{T,\lambda}} SSA_{D,\lambda} + \frac{AOD_{nd,\lambda}}{AOD_{T,\lambda}} SSA_{nd,\lambda} \quad (6)$$

where SSA_{nd} is derived as

$$SSA_{nd,\lambda} = \left(SSA_{\lambda} - \frac{AOD_{D,\lambda}}{AOD_{T,\lambda}} SSA_{D,\lambda} \right) \times \frac{AOD_{T,\lambda}}{AOD_{nd,\lambda}} \quad (7)$$

The spectral SSA values for pure Asian dust particles at 440, 675, 870, and 1020 nm have been calculated as 0.94, 0.98, 0.98, and 0.98, respectively, using data for a source region of Asian dust; Dunhuang in Western China (40.49°N 94.95°E) [31].

From SSA_{nd} , the share of light scattered by coarse and fine non-dust particles, the non-dust AAOD can be found:

$$AAOD_{nd,\lambda} = (1 - SSA_{nd,\lambda}) AOD_{nd,\lambda} \quad (8)$$

Since aerosol light absorption in the atmosphere is dominated by BC, we assume that in the absence of dust particles the light absorption by pollution (non-dust) is mostly due to BC. As BC is not an ideal light absorber, its SSA is not equal to 0. To estimate the BC-related AAOD ($AAOD_{BC}$) we used the SSA of BC ($SSA_{BC,\lambda}$):

$$AAOD_{BC,\lambda} = AOD_{nd,\lambda} \times (1 - SSA_{nd,\lambda}) \times (1 - SSA_{BC,\lambda}) \quad (9)$$

The SSA of fresh BC has been reported as between 0.07 and 0.28 [40–42]. Values of 0.25, 0.17, 0.13, and 0.07 were used here at 440, 675, 870, and 1020 nm, respectively.

In the results section, the mean values of our findings and the specific trends for each variation are defined from 2001 to 2018 in Figures 2–6.

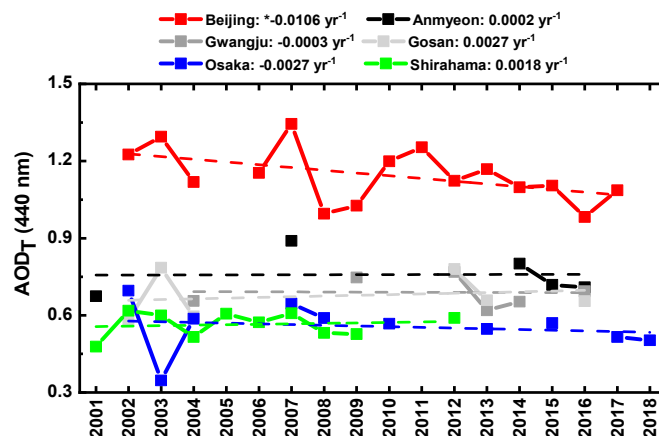


Figure 2. Variations of annual mean AOD_T at 440 nm from 2001 to 2018 at six sites. * (units AOD_T yr⁻¹).

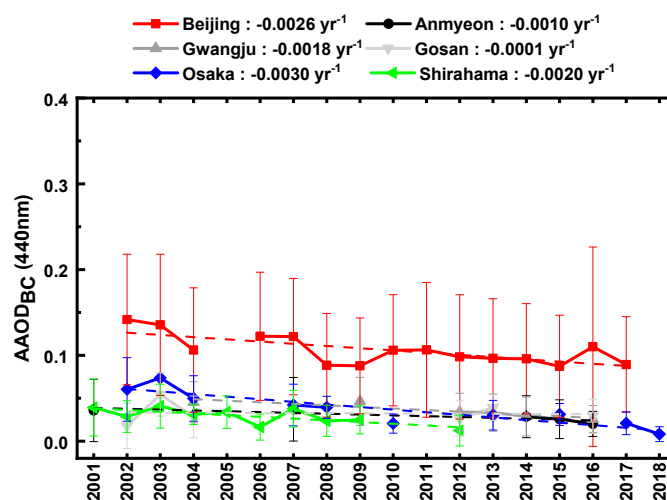


Figure 3. Variations of annual mean AAOD_{BC} at 440 nm from 2001 to 2018 at six sites. Error bars show the standard deviation of the annual averages. * (units AAOD_{BC} yr⁻¹).

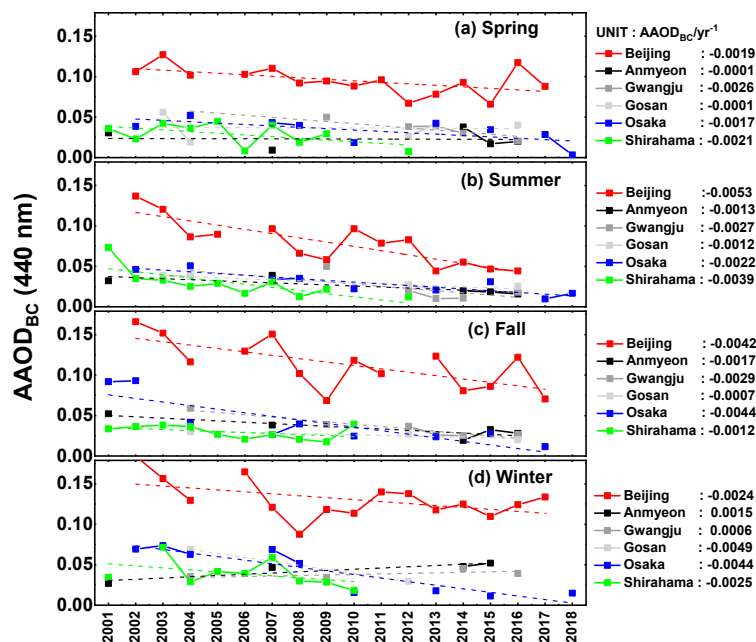


Figure 4. Variations of seasonal mean AAOD_{BC} at 440 nm in (a) spring (b) summer (c) fall, and (d) winter from 2001 to 2018 at six sites. * (units AAOD_{BC} yr⁻¹).

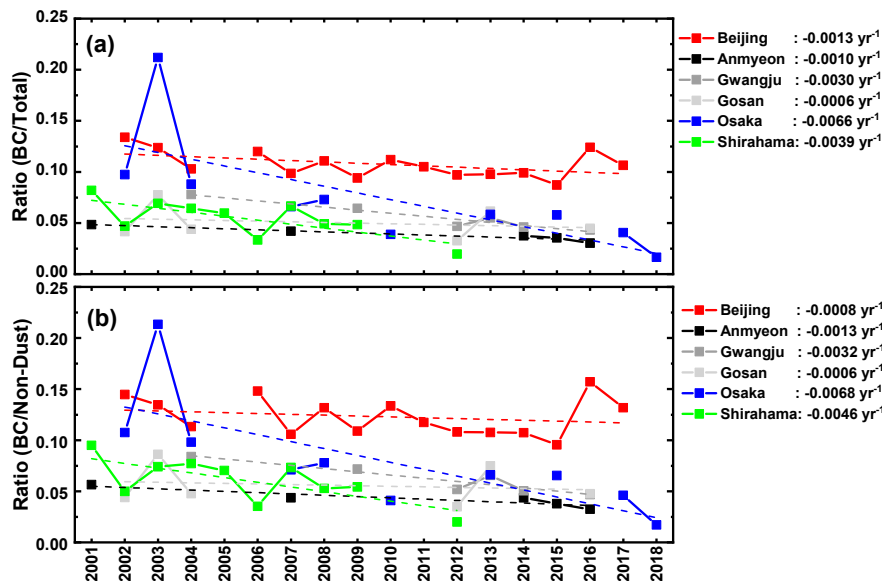


Figure 5. Variations of (a) annual $AAOD_{BC}/AOD_T$ ratio at 440 nm (b) annual ratio of $AAOD_{BC}/AOD_{nd}$ at 440 nm from 2001 to 2018 at six sites. * (units yr^{-1}).

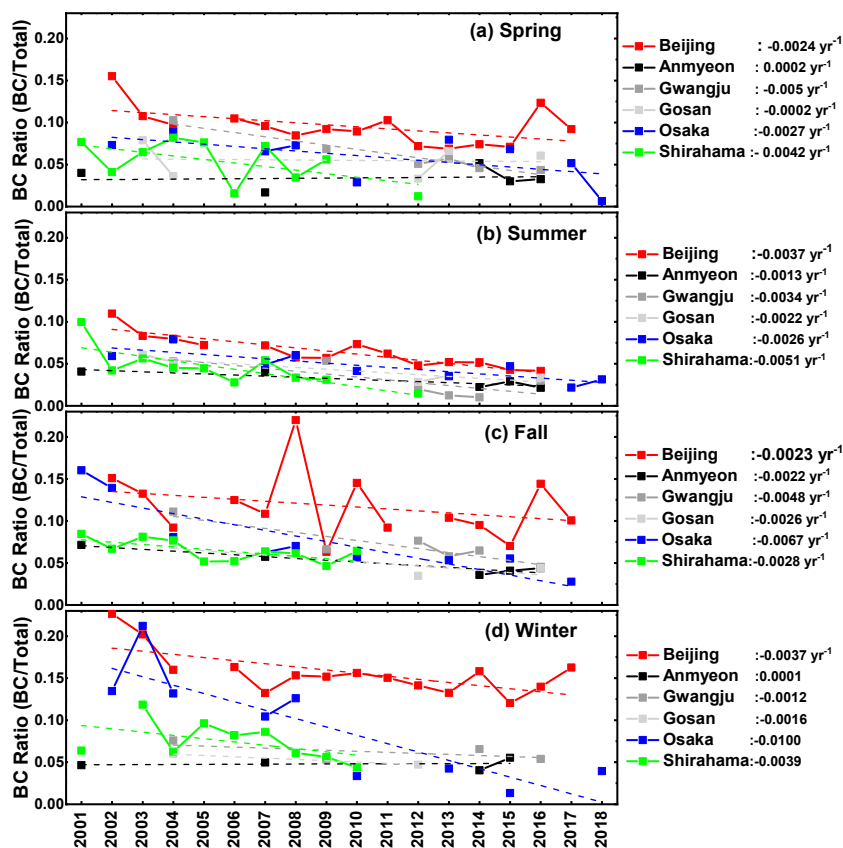


Figure 6. Seasonal variations of annual $AAOD_{BC}/AOD_T$ ratio at 440 nm in (a) spring (b) summer (c) fall, and (d) winter from 2001 to 2018 at six sites. * (units yr^{-1}).

2.3. Statistical Analysis

To the better interpretation of the $AAOD_{BC}$ trends, the average data points for each year at all sites were subjected to a one-way ANOVA test using SPSS statistics application (IBM SPSS Statistics, USA) to compare means at $p < 0.05$.

3. Results

3.1. Total Aerosol Optical Depth (AOD_T) and $AAOD_{BC}$

Table 1 summarizes the daily mean values of total and seasonal variation of AOD_T , $AAOD_{BC}$, and $AAOD_{BC}$ ratio (the ratio of $AAOD_{BC}/AOD_T$, i.e., $AOD > 0.4$) at 440 nm for the six selected stations over the period 2001–2018.

The highest values of AOD_T in Table 1 are for Beijing, whereas the Japanese sites showed the lowest (almost half those for Beijing). Considering seasonal variation, AOD_T was highest in summer at most sites, but not at Anmyeon and Gosan where it was higher in winter and fall, respectively. Beijing showed the highest total $AAOD_{BC}$ and $AAOD_{BC}$ ratio during all seasons, with values greatest in winter. All the Korean and Japanese sites showed roughly similar values of $AAOD_{BC}$ (0.02–0.04), which were clearly less than those for Beijing (0.09–0.13).

Figure 2 shows the 18-year (2001–2018) variations of the annual mean AOD_T at 440 nm for each site. Beijing showed steeply declining values ($-0.0106 AOD_T \text{ yr}^{-1}$) during the study period, whereas no considerable trends emerged for the Korean and Japanese sites, which showed annual variations between -0.0003 and $0.0027 AOD_T \text{ yr}^{-1}$. Overall, there is some seasonal variability in AOD, which is generally highest in summer (see Table 1).

Figure 3 presents the annual variation in $AAOD_{BC}$ at the six sites. Similar to AOD_T in Figure 2, Beijing showed the highest annual mean $AAOD_{BC}$, which was approximately double that at each of the Korean and Japanese sites. Unlike the annual variation of AOD_T , $AAOD_{BC}$ declined annually, not only in Beijing but at all sites, with the greatest annual decline ($-0.0030 AAOD_{BC} \text{ yr}^{-1}$) observed at Osaka. The next greatest annual decline of $-0.0026 AAOD_{BC} \text{ yr}^{-1}$ was at Beijing, where the highest $AAOD_{BC}$ of approximately 0.14 in 2002 declined to 0.09 in 2017. The annual mean $AAOD_{BC}$ at Osaka, Shirahama, and Gosan peaked in 2003. The lack of continuous observations at the Korean sites prevented observation of any coherent trend for $AAOD_{BC}$, but $AAOD_{BC}$ declined overall at these sites from 2001 to 2018.

Figure 4 shows the time series of seasonal mean $AAOD_{BC}$ and its trends for the six sites. There is a distinct seasonal trend, with higher values in winter and lower values in summer. Most sites showed annual decreases of $AAOD_{BC}$; the exceptions, Anmyeon and Gwangju, showed increases in winter values of 0.0015 and $0.0006 AAOD_{BC} \text{ yr}^{-1}$, respectively.

Table 1. AERONET data for six selected sites listing mean values and standard deviations of total and seasonal variations of AOD_T, AAOD_{BC}, and AAOD_{BC} ratio at 440 nm, over the period 2001–2018.

Station	N	AOD _T (440 nm) (Mean ± SD)					AAOD _{BC} (440 nm) (Mean ± SD)					AAOD _{BC} Ratio (440 nm) (Mean ± SD)				
		Total	Spring	Summer	Fall	Winter	Total	Spring	Summer	Fall	Winter	Total	Spring	Summer	Fall	Winter
Beijing	1313	1.15 ± 0.73	1.10 ± 0.65	1.43 ± 0.80	1.18 ± 0.80	0.96 ± 0.67	0.11 ± 0.08	0.09 ± 0.07	0.09 ± 0.06	0.12 ± 0.08	0.13 ± 0.08	0.11 ± 0.01	0.09 ± 0.06	0.07 ± 0.04	0.12 ± 0.07	0.15 ± 0.06
Anmyeon	240	0.74 ± 0.38	0.67 ± 0.30	0.78 ± 0.39	0.72 ± 0.37	0.94 ± 0.55	0.03 ± 0.03	0.03 ± 0.03	0.02 ± 0.03	0.04 ± 0.02	0.04 ± 0.03	0.04 ± 0.01	0.04 ± 0.03	0.03 ± 0.02	0.05 ± 0.02	0.05 ± 0.02
Gwangju	267	0.70 ± 0.35	0.69 ± 0.31	0.84 ± 0.40	0.54 ± 0.18	0.80 ± 0.61	0.04 ± 0.02	0.04 ± 0.02	0.03 ± 0.03	0.03 ± 0.02	0.04 ± 0.02	0.06 ± 0.01	0.06 ± 0.03	0.03 ± 0.03	0.07 ± 0.03	0.06 ± 0.03
Gosan	113	0.69 ± 0.32	0.69 ± 0.32	0.72 ± 0.27	0.73 ± 0.39	0.63 ± 0.24	0.04 ± 0.02	0.04 ± 0.03	0.03 ± 0.02	0.03 ± 0.01	0.04 ± 0.02	0.05 ± 0.02	0.06 ± 0.04	0.04 ± 0.02	0.04 ± 0.02	0.06 ± 0.03
Osaka	300	0.58 ± 0.20	0.57 ± 0.19	0.64 ± 0.21	0.53 ± 0.20	0.53 ± 0.20	0.03 ± 0.02	0.03 ± 0.02	0.03 ± 0.02	0.04 ± 0.03	0.04 ± 0.03	0.07 ± 0.05	0.06 ± 0.04	0.05 ± 0.03	0.07 ± 0.05	0.08 ± 0.07
Shirahama	296	0.57 ± 0.23	0.57 ± 0.22	0.65 ± 0.28	0.46 ± 0.13	0.51 ± 0.18	0.03 ± 0.02	0.03 ± 0.02	0.03 ± 0.02	0.03 ± 0.02	0.04 ± 0.02	0.05 ± 0.02	0.05 ± 0.04	0.05 ± 0.03	0.06 ± 0.03	0.08 ± 0.03

3.2. BC Ratio

The variations of the annual ratios of $AAOD_{BC}$ to AOD_T and $AAOD_{BC}$ to AOD_{nd} at 440 nm and their trends for 2001–2018 are shown in Figure 5a,b, respectively. Both ratios show a long-term declining trend at all sites. As mentioned in Section 3.1, Beijing showed the greatest declining trend for the annual variation of AOD_T and the second greatest for $AAOD_{BC}$ (after Osaka), with annual declines of -0.0106 and -0.0026 yr^{-1} , respectively (Figures 2 and 3). The results in Figure 5 show that Beijing had the smallest decreases in the annual ratios of both $AAOD_{BC}/AOD_T$ (after Gosan and Anmyeon) and $AAOD_{BC}/AOD_{nd}$ from 2001 to 2018. Therefore, although Beijing had sharp drops in $AAOD_{BC}$ and AOD_T , its ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ decreased only slightly. On the other hand, for the Korean and Japanese sites, although AOD_T remained largely steady, and decreases (in Osaka and Gwangju) were less than those in Beijing, rapid decreases in $AAOD_{BC}$ led the annual ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ to fall more quickly than those for Beijing.

Figure 6 compares the seasonal variation of the annual $AAOD_{BC}/AOD_T$ ratio (BC ratio) at 440 nm. It was higher during winter, and lower in summer (below 0.12) at all sites. The observed variation indicates a larger contribution of BC to AAOD in fall and winter. The BC ratio declined at all sites, other than Anmyeon (during spring and winter). The greatest rate of decrease at -0.0100 was at Osaka in the winter.

Figure 7b presents the monthly variation of BC ratio from 2001 to 2018. The highest values were in Beijing, peaking in November, December, and January; the lower values were in August. The next-highest values were at the Japanese sites (Osaka and Shirahama), while the Korean sites showed the lowest BC ratios.

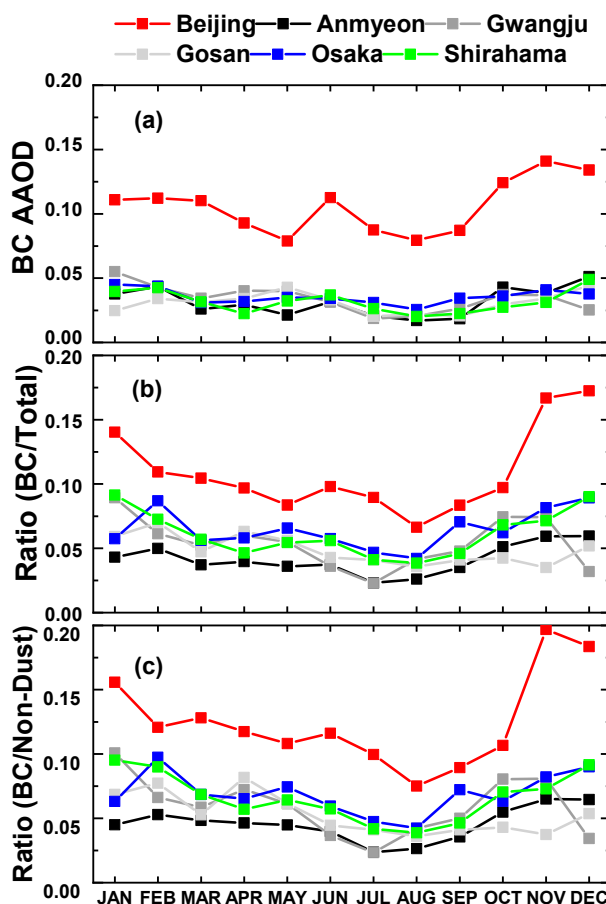


Figure 7. Monthly variations of (a) mean $AAOD_{BC}$, (b) the ratio of $AAOD_{BC}$ to AOD_T (BC ratio), and (c) the ratio of $AAOD_{BC}$ to AOD_{nd} at 440 nm from 2001 to 2018 at six sites. * (a) (units, $AAOD_{BC} \text{ yr}^{-1}$), (b) (units, yr^{-1}), and (c) (units yr^{-1}).

The monthly mean variations of the ratio of $AAOD_{BC}$ to AOD_{nd} at the six sites are shown in Figure 7c. For Beijing, this ratio increased sharply from October to February, and was lowest in late summer (August).

3.3. Statistical Analysis Results

Since $AAOD_{BC}$ trends are the main findings of this study, a one-way ANOVA test was applied to the average data sets at all six sites to further test that if the trends are statistically significant or not. Table 2 summarizes the statistical results of the One-way ANOVA. Among all the selected sites, Beijing and Anmyeon with the p -values of 0.128 and 0.358 did not show a significant difference between the average data points.

Table 2. Results of the ANOVA test for variations of annual mean $AAOD_{BC}$ at 440 nm from 2001 to 2018 at six sites using SPSS statistics application (IBM SPSS Statistics, USA).

Station	p -Value *
Beijing	0.128
Anmyeon	0.358
Gwangju	0.023
Gosan	0.032
Osaka	0.00
Shirahama	0.016

* Means were considered significant at $p < 0.05$.

4. Discussion

According to Figure 2, Beijing showed steeply declining values of AOD_T during the study period, whereas no considerable trends were found for the Korean and Japanese sites. Yu et al. [43] and Kim et al. [44] reported similar seasonal variations of AOD in Beijing and Gosan, respectively. The higher values and high declining trend of AOD in China have been reported previously [45–47]. Yu et al. [48] compared AOD values in East Asia during 2001–2005, and found higher values at Chinese sites than at Korean or Japanese sites. Khan et al. [49] reported a decreasing trend in AOD at 440 nm over the North China Plain, with Beijing showing the highest values from 2001 to 2018; however, some studies have reported that the annual mean AOD has remained constant or increased over Northeast Asia and China [50–52]. Nam et al. [50] found the highest annual mean AOD at North China sites and lower values at Korean and Japanese sites. A significantly increasing trend in AOD has been found in North and Northeast China, but no significant trends were identified at sites in Korea (Gwangju) and Japan (Shirahama and Osaka) between 2004 and 2015.

The higher AODs in China and its downwind regions during spring are attributed to dust plumes transported from deserts in Mongolia and China, which coincide with increasing local sources during this season [53,54]. In summer, AOD peaks at most sites due to increased moisture and enhanced formation of secondary aerosols [55–57]. Beijing's higher annual mean AOD in comparison to other sites is mainly due industrial pollution and its combination with mineral dust and other anthropogenic particles [58]. This is also associated with high emissions of different chemical species, such as burning fossil fuel (including coal) for transportation, industry, and domestic heating during cold seasons [59]. Furthermore, the declining trend of AOD in Beijing can be explained by improving air pollution control measures and the implementation of strict governmental policies on air pollution [60,61]. The lower variation of annual mean AOD at Korean and Japanese sites is attributable to lower air pollution emissions in comparison to China. The lack of continuous observations and limited data for the Korean and Japanese sites is another possible reason for their lower AOD values.

In Figure 3, Beijing showed the highest annual mean $AAOD_{BC}$; besides, $AAOD_{BC}$ declined overall at the Korean and Japanese sites from 2001 to 2018. The trends for Anmyeon and Beijing are not statistically significant; however, the higher values of $AAOD_{BC}$ were found in the Beijing site.

This non-significant trend can result from the higher emissions of BC over China. In the case of the Anmyeon site, the non-significant trend might be due to the few observations and insufficient data sets over this site. Sun et al. [62] found decreasing trends in both $AAOD_{BC}$ and AOD from 2008 to 2017 in China. The variation of AAOD in China from 1980 to 2017 was mostly attributed to the variation of $AAOD_{BC}$. Other studies have also reported declining trends of BC concentration in Beijing, which may explain the declining trend of $AAOD_{BC}$ in this region [8,21]. The reduction of BC emissions in China appears to result from governmental clean air actions (e.g., Regional Emission inventory in Asia (REAS) version 3.1 and Multi-resolution Emission Inventory version 1.3 (MEIC1.3)), reduction of coal combustion (both industrial and residential), traffic limitation, and gasoline consumption restrictions [63].

Cho et al. [64] compared the contribution of BC to carbonaceous aerosol AAOD at background sites (Gosan, Anmyeon, Shiraham, Fukuoka, and Baengnyeong) and polluted urban regions (Beijing, Gwangju, Osaka, Xianghe, Xuzhou, Taihu, Hongkong, Seoul, and Chiba). They found that BC made a greater contribution to carbonaceous aerosol AAOD in polluted urban regions than in the background cities. Our study found the highest annual mean $AAOD_{BC}$ in China, the largest source of BC in East Asia [65,66]. Liquid fuels such as kerosene, gasoline, and diesel have been found to contribute significantly to BC pollution in Beijing in recent years [67]. The high $AAOD_{BC}$ in China is mainly due to carbon-containing pollutants emitted from industry and other human activities associated with the country's growing economy and population density over the last three decades. Transportation also accounts for significant emissions of BC in Beijing and the neighboring regions. We found lower values of $AAOD_{BC}$ in rural and coastal regions, such as Anmyeon, Gosan, and Shirahama, compared with metropolitan cities such as Beijing, Osaka, and Gwangju. This is related to lower BC emissions from diesel engines for transportation, industrial, and residential use.

According to Figure 4, there is a distinct seasonal trend for $AAOD_{BC}$, with higher values in winter and lower values in summer. The BC emissions from residential heating accounted for the generally higher winter values of $AAOD_{BC}$. The highest mean $AAOD_{BC}$ (approximately 0.17) was observed at Beijing in winter 2002; this value had dropped to 0.05 by summer 2016. The mean values in Korea and Japan were consistently lower (less than 0.10) than those observed in China over all seasons. Several studies have reported similar seasonal trends of BC concentrations (lowest in summer, highest in winter) at Chinese sites [68,69]. Zhuang et al. [70] found considerable seasonal variations in BC emissions over East Asia, predominantly driven by changing domestic and residential emissions. Kang et al. [71] attributed increased AAOD over the North China Plain and the Korean Peninsula during winter to seasonal increases of BC and organic carbon (OC) emissions. Incomplete combustion of fossil fuel for domestic heating is a significant source of BC and OC [72], and the strong light-absorbing properties of BC partly explain the increased $AAOD_{BC}$ during winter.

Figure 5a,b shows the variations of the annual ratios of $AAOD_{BC}$ to AOD_T and $AAOD_{BC}$ to AOD_{nd} at 440 nm and their trends for 2001–2018, respectively. The $AAOD_{BC}$ in Japan and Korea fell more greatly or more quickly than in Beijing, despite AOD_T remaining stable at those sites. We attributed the difference in the ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ between Beijing and the Korean and Japanese sites to Beijing's lower and/or slower reduction in BC emissions during the study period. Figure 5b shows that Beijing's ratio of $AAOD_{BC}/AOD_{nd}$ declined at a rate of 0.0008 yr^{-1} , the lowest among the six sites. Hence, it can be concluded that Beijing's decrease in BC emissions relative to the other pollution particles was lower than that at the Korean and Japanese sites. This might be attributable to the delayed implementation of pollution control or fuel substitution policies, or a shift of, or even increase in, residential coal use and emissions from vehicles burning liquid fuel in Beijing compared to the Korean and Japanese sites. In other words, although BC emission control policies in China seem to have been successful during the past 18 years, BC still needs to be further reduced.

The ratio of $AAOD_{BC}/AOD_{nd}$ showed an almost identical annual variation to the ratio of $AAOD_{BC}/AOD_T$ in the Korean and Japanese sites. Polluted urban regions, such as Osaka, showed higher $AAOD_{BC}/AOD_{nd}$ ratios during the study period than background cities, such as Anmyeon

and Gosan, owing to higher BC concentrations. Another distinctive feature of Figure 5 is that Beijing generally had the highest values for both annual ratios from 2001 to 2018 (except for 2003). Beijing's severe urban haze that contains notable amounts of BC may account for its high $AAOD_{BC}/AOD_T$ ratio. China has been reported to emit nearly a quarter of global anthropogenic BC due to its high usage of coal, oil, and biofuel [73,74].

Figure 6 represents that the seasonal variation of the annual $AAOD_{BC}/AOD_T$ ratio (BC ratio) at 440 nm was higher during winter, and lower in summer at all sites. The common trend in BC ratio across the sites suggests similar reductions in BC emissions associated with the implementation of emission control measures over East Asia. A seasonal trend with summer minima and winter maxima was superimposed on this downward trend at all sites (except for Anmyeon). Regardless of the seasonal variation, Beijing showed the highest BC ratios as it had the highest BC emissions among the selected sites. Due to a lack of long-term regional-scale observational data, no consistent trend was identifiable for the Korean sites specifically in winter, but Gwangju had higher BC ratios than the other Korean sites (Anmyeon and Gosan) during all seasons. This is connected to Gwangju being a metropolitan city and a regional industrial center, with much coal-burning and other anthropogenic emissions that negatively affect air quality. Among the Japanese sites, Osaka had a higher BC ratio than Shirahama. Osaka is Japan's second largest urban area and has contributed greatly to Japan's economy and rapid industrialization over recent decades, which may explain its high BC ratio. Note that in addition to local emissions, the Korean and Japanese sites might be affected by aerosol loads transported from China by strong westerly winds [75].

According to Figure 7b, the monthly variation of BC ratio represents the highest values in Beijing, peaking in November, December, and January, and the lower values in August from 2001 to 2018. Summers in Beijing are rainy, and account for about 70% of the city's rainfall. The frequent rain probably lowered the concentration of BC, and consequently its AAOD, over Beijing in summer. The BC sources from October to March in Beijing are unlikely to be forest fires because of the cold weather. The increased BC ratio in this period was instead attributed to changes in major emission sources, specifically residential emissions. Qin et al. [67] suggested that transportation is the dominant emission source of BC in Beijing, with similarly high emissions in all seasons, while residential BC emissions greatly increase in winter. The Korean and Japanese sites showed lower BC ratios (below 0.10) than Beijing. The limited observation data for the Korean and Japanese sites may have resulted in them showing lower BC ratios. The highest values were in cold seasons (October to March), and the lowest values were during warmer seasons (June to September).

Figure 7c shows that the monthly mean variations of the ratio of $AAOD_{BC}$ to AOD_{nd} increased sharply from October to February, and was lowest in late summer (August) for Beijing. This suggests a higher contribution of BC to AAOD during the colder months (November to January) than at other times of the year. The average ratio of $AAOD_{BC}$ to AOD_{nd} at 440 nm during 2001–2018 varied between 0.10 and 0.02 at the Korean and Japanese sites, with the Japanese sites showing higher values than those in Korea. Values were highest in December and January, and lowest in July and August. The residential sector was likely the main reason the ratio of $AAOD_{BC}$ to AOD_{nd} increased during the colder months over these regions.

It is worth noting that in this study some errors arise from SSA_D and SSA_{BC} . As was mentioned in the materials and methods section, the value of 0.94 was used for SSA of East Asian dust at 440 nm [31], while in some other studies the value of 0.92 was applied for SSA_D over Korea sites [48]. Besides, the value of 0.25 was applied for SSA_{BC} at 440 nm; however, in some other studies, this value was reported to be 0.25 ± 0.13 [14]. In this study, the value of 0.25 ± 0.13 for SSA_{BC} can cause negligible changes in $AAOD_{BC}$ values.

5. Conclusions

The long-term results for AOD_T show that Beijing had the highest values, but its values are decreasing. The lower values in Korea and Japan showed no significant trend. This suggests that

Beijing had higher loads of anthropogenic emissions from transportation, residential, and industrial sectors, which combined with naturally occurring mineral dust. Our analysis of $AAOD_{BC}$ revealed a decrease at all sites from 2001 to 2018, and the highest values were in Beijing. The $AAOD_{BC}$ varied seasonally, being greatest in winter and lowest in summer due to seasonal differences in BC emissions. Comparison of the annual ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ and their trends over the selected sites showed that Beijing had the lowest rate of decrease for both ratios, despite it showing the greatest rate of decrease for annual variation of AOD_T and the second greatest for $AAOD_{BC}$ (after Osaka). The annual ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ in the Korean and Japanese sites decreased more rapidly than those in Beijing, despite these sites showing largely steady AOD_T , because $AAOD_{BC}$ decreased. This suggests that, although BC emissions have reduced over China during the past 18 years, further reductions are needed. The monthly variations of $AAOD_{BC}$, and the ratios of $AAOD_{BC}/AOD_T$ and $AAOD_{BC}/AOD_{nd}$ increased during the cold months and in June, because of elevated BC emissions for domestic heating and biomass burning, respectively. Overall, this study is a valuable contribution, and particularly useful for the analysis of regions with high BC emissions that mix with desert dust and anthropogenic pollutants to affect downwind areas.

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Data Availability: The data used in this study are freely available through the AERONET portal at <https://aeronet.gsfc.nasa.gov/>.

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