Long-term trends in aerosol properties derived from AERONET measurements

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Abstract.

Over the past two decades, remarkable changes in aerosol concentrations and compositions have been observed worldwide, especially over developing countries, potentially resulting in considerable changes in aerosol properties. The Aerosol Robotic Network (AERONET) offers high precision measurements of aerosol optical parameters over about 1700 stations globally, many of which have long-term measurements for one or more decades. Here we use AERONET Level 2.0 quality assured measurements to investigate long-term trends for aerosol optical depth (AOD) and Ångström exponent (AE) trends, and quality-controlled Level 1.5 inversion products to analyze trends of absorption aerosol optical depth (AAOD) and single scattering albedo (SSA) at stations with long-term records. We also classify the aerosol properties in these sites into 6 types, and analyze the trends of each type. Results reveal decreases in AOD over the majority of the stations, except for North India and the Arabian Peninsula, where AOD increased. AE ~~also~~ (computed from the AOD within the range of 440-870 nm) decreased in Europe, eastern North America, and the Middle East, but increased over South Asia and ~~East Asia~~western North America. The decreased AE over Europe and eastern North America is likely due to decreased fine-mode anthropogenic aerosols, whereas that over the Arabian Peninsula is attributed to increased dust activities. Conversely, increased AE over North India is probably attributed to increased anthropogenic emissions and decreased dust loading. Most stations in Europe, North America, East Asia, and South Asia exhibit negative trends in AAOD, whereas Solar_Village in the Arabian Peninsula has positive trends. SSA at most stations increases and exhibits opposite trends to AAOD, but with several stations in ~~central Europe and~~ North America showing decreased SSA values. Trend analysis of different aerosol types further reveals the changes of different aerosol components that are related to AOD, AE, AAOD, and SSA trends. ~~Stronger reductions in~~ The reductions of aerosols in eastern North America mainly result from non-absorbing species. Reductions of both fine-mode absorbing species ~~than that of~~

and non-absorbing aerosols are found over Europe and East Asia, ~~whereas in eastern North America the reductions of aerosols are dominated by~~ but the reduction of absorbing species is stronger than that of non-absorbing species. Increased aerosols in Kanpur over North India should be mainly comprised of fine-mode scattering species, whereas those in Solar_Village over the Arabian Peninsula are mainly dust. Weak seasonality is found in the trends of all aerosol parameters analyzed in this work.

1 Introduction

Aerosols are pivotal in the study of climate change due to their significant effects on the climate system. Understanding the climate effects of aerosols necessitates a comprehensive recognition of their optical and microphysical properties. Variations in aerosol loading and aerosol properties can result in disparate climate impacts, underscoring the importance of accurately comprehending these changes. For example, changes in aerosol loading can directly influence the intensity of aerosol forcing, while a rise in aerosol absorption could even shift the aerosol forcing from negative to positive (Hansen et al., 1997), remarkably altering their climate effects. To quantify the contribution of aerosols to climate variability effectively, it is thus crucial to understand and quantify the long-term change of aerosol properties.

Studies using satellite observations revealed continuous reductions in the loading of aerosols and their precursors in Europe, North America, South America, and Africa in the past several decades, but increases over South Asia and Middle East, as well as increases in 2000s and decreases in 2010s over East Asia (Krotkov et al., 2016; Mehta et al., 2016; Zhao et al., 2017; de Meij et al., 2012; Fioletov et al., 2023; Gupta et al., 2022). In situ measurements also suggested negative scattering and absorption coefficient trends in majority of the stations which are mainly located in Europe and North America, and revealed ~~positive trends of aerosol scattering~~ increased scattering aerosol fraction (represented by single scattering albedo, SSA) in Asia, eastern/northern Europe, and the Arctic, and negative SSA trends in central Europe and central North America (Collaud Coen et al., 2020). As satellite observations mainly provide aerosol loading products and may have drifts in long-term calibration which impact aerosol monitoring~~and mainly provide aerosol loading products~~, and the spatial coverage of in situ measurements is quite limited, ground-based remote sensing networks provide a very accurate data source to analyze trends in multiple aerosol parameters worldwide. Xia (2011) examined 79 stations within the Aerosol Robotic Network (AERONET, Holben et al., 1998) with observations no less than six years, and found decreases in aerosol optical depth (AOD) and Ångström exponent (AE) in eastern North America and Europe. Ningombam et al. (2019) analyzed long-term AOD trends over 49 AERONET sites and 4 Sky radiometer Network (SKYNET, Takamura and Nakajima, 2004) sites, and reported decline in AOD over North-South America, Europe, the Arctic, and Australia.

However, these studies based on ground-based remote sensing data mainly focused on trends in AOD and AE, while analysis on other aerosol optical properties, such as SSA and absorption aerosol optical depth (AAOD), is still insufficient. Other studies focusing on trends of these parameters are mainly restricted to specific stations with long-term records, which is mainly because of the limited data availability of AERONET Level 2.0 data. Li et al. (2014) utilized quality-controlled AERONET Level 1.5 inversion measurements at 54 selected stations as well as Level 2.0 solar observations at 90 selected stations worldwide for the period 2000-2013 to analyze the trends of AOD, AE, SSA, and AAOD. Decreased AOD and AAOD trends, along with increased SSA trends, were consistently observed in Japan, Europe and North America. North America exhibited positive AE trends, whereas Europe showed negative AE trends. India was reported to experience increases in AOD, AE, and SSA. The Arabian Peninsula was noted for experiencing increased AOD and AAOD, with decreases in AE and SSA. Eastern China was characterized by a positive SSA trend and a negative AAOD trend, without significant changes in AOD or AE.

A decade later, many regions have experienced significant changes in aerosol loading and compositions. For example, recent studies have highlighted considerable reductions in aerosol loadings in East Asia as evidenced by AERONET measurements (Yu et al., 2022; Ramachandran and Rupakheti, 2022; Eom et al., 2022) and satellite observations (Ramachandran et al., 2020; Krotkov et al., 2016; Mehta et al., 2016; Zhao et al., 2017; Fioletov et al., 2023; Li, 2020; Gupta et al., 2022). Substantial reductions in anthropogenic emissions have been observed in eastern North America (Krotkov et al., 2016), potentially contributing to a decrease in AE. Central Australia has seen reported increases in dust activities (Shao et al., 2013), aligning with observed increases in AOD and decreases in AE (Yang et al., 2021), which might also lead to positive AAOD and negative SSA trends. Some potential variations in aerosol optical properties in certain regions were not captured by Li et al. (2014), partly due to limitations in the spatial and temporal coverages of surface stations at that time, and recent changes in aerosol loadings and compositions might lead to different or reversed trends. AERONET has now expanded from 400 to over 1700 stations globally with longer records. The AERONET algorithm has also been updated to Version 3 with numerous improvements (Giles et al., 2019; Sinyuk et al., 2020). These progresses underscore the need to update trend analysis of AERONET data to capture recent shifts in aerosol optical properties and reflect advancements in data quality and network coverage.

In this study, we analyze AERONET Level 2.0 AOD and AE observations at ~~165~~ 172 stations and Level 1.5 quality-controlled AAOD and SSA measurements at ~~74~~ 72 stations. We also made a further attempt to categorize aerosol types and analyze the trends of each type. We hope that this study can provide a more recent reference to aerosol changes globally and facilitate the assessment of aerosol climate and environmental impacts.

2 Data and Methods

2.1 AERONET Data

The AERONET is a ground-based aerosol remote sensing network, providing long-term observations of aerosol optical and microphysical properties, covering most of the continental areas around the world (Holben et al., 1998). The AERONET AOD observations are derived from direct solar radiation at several wavelength bands mainly ranging from 340 nm to 1640 nm, while other aerosol properties, including SSA and AAOD, are derived from diffuse sky radiance at four wavelengths at 440, 675, 870, and 1020 nm (Dubovik and King, 2000). The AE parameter is calculated using AOD measurements within ~~440–870~~ 440-870 nm interval (Eck et al., 1999; Giles et al., 2019). There is a series of quality assurance strategies for AERONET Level 2.0 data that ensure an AOD uncertainty of 0.01 (visible)-0.02 (UV) and an SSA uncertainty of 0.03 at AOD_{440} (AOD at 440 nm) $\sim$ 0.4 (Holben et al., 2006; Giles et al., 2019; Sinyuk et al., 2020). However, as Level 2.0 quality assurance for inversion products requires a coincident AOD exceeding 0.4 at 440 nm, many stations do not have enough data samples to produce a long-term ~~Level 2.0 inversion records~~record. Therefore, considering both the data quality and data availability, we utilize the all-point

Version 3 Level 2.0 direct measurements for AOD and AE, and quality-controlled Level 1.5 almucantar inversion products (see below for the quality control scheme) for other parameters. The uncertainty in SSA increases rapidly (exponentially) at lower AOD levels (Sinyuk et al., 2020), therefore much of the SSA retrievals in this study have larger uncertainties of ~ 0.03 to ~ 0.09. description and uncertainties of these parameters are detailed in Sect. 2.2.

The stations are selected primarily based on the availability of an extensive data record for the purpose of estimating the long-term trends of aerosol properties. The Level 1.5 almucantar inversion products are first screened based on all the Level 2.0 quality assurance criteria except for the AOD threshold, such as solar zenith angle > 50°, sky error < 5%, and coincident Level 2.0 AOD measurements. The Level 2.0 direct measurements and screened Level 1.5 almucantar inversion products are then used to calculate monthly measurements. We first remove outliers from all-point measurements, and then calculate the median of all-point measurements to represent the monthly value only if there are more than 5 all-point measurements in at least 3 different days for that month. To ensure adequate records in trend analysis, we require the data to have at least 10 years of records and no less than 8 monthly measurements for each year during the 2000-2022 period. For the years with at least 8 monthly measurements, the monthly medians are then averaged to annual and seasonal means, which are used to calculate annual and seasonal trends. Estimating a valid seasonal trend also requires at least a 10-year record. Considering polar stations often have no monthly measurements in winter, the least number of monthly medians for each year are reduced to 4 for stations at latitudes above 65 degrees. Specifically, the 2019-2022 data for Birdsville in Australia are eliminated for more accurate trend estimation, as these data are strongly biased due to a data filtering artifact in the quality assurance (QA) process of the algorithm according to Giles et al. (2019), which results in a large jump in AOD (personal communication, T. Eck). This AOD artifact is caused by erroneous time stamping of the data that is greatest at some sites in Australia due to a unique data logging system utilized there. The unnatural increase in AOD for Birdsville in 2019 can be found in Yang et al. (2021). As a result, 165 172 stations for the direct-sun observations and 74 72 stations for the inversion measurements are retained for trend analysis, covering all major continents over the world. Locations, trends and time series for all the stations could be found in the supplementary. The distributions of all the selected stations as well as the number of monthly annual mean samples at each station are presented in Fig. 1. Locations of stations mentioned in the manuscript this article are presented in Fig. 2.

Here we focus on analyzing AOD, SSA, and AAOD trends at 440 nm, which are noted as AOD_{440}, SSA_{440}, and $AAOD_{440}$, respectively. Trends for parameters at the other wavelengths are very similar and thus skipped. The AE is calculated from all AOD measurements within the 440–870 440-870 nm wavelength range (typically including 440, 500, 675, and 870 nm), and are commonly denoted as AE_{440_870}.

2.2 Mann–Kendall test and Sen's slope for trend analysisAerosol parameters

AOD represents the column aerosol extinction, directly reflecting the column loading and concentration of aerosols. AERONET Level 2.0 products provide very accurate AOD measurements at clear sky conditions, with an uncertainty of 0.01 at visible wavelengths. The patterns of AOD (Fig. 3) and AOD trends (Fig. 4) should be always kept in mind when analyzing trends of the other aerosol parameters, because uncertainties of the other parameters are closely related to AOD level (see below), whose trend reflect changes of aerosol loading.

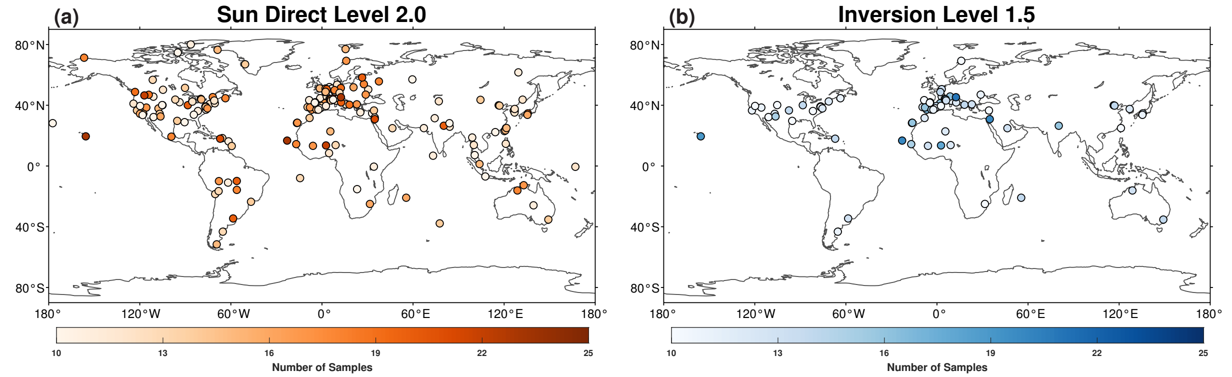

Figure 1. Locations of the stations selected for this study. (a) Level 2.0 solar stations, (b) Quality-controlled Level 1.5 almucantar stations. ~~The~~ Color coding denotes the number of monthly samples for each station~~is also displayed with different color~~.

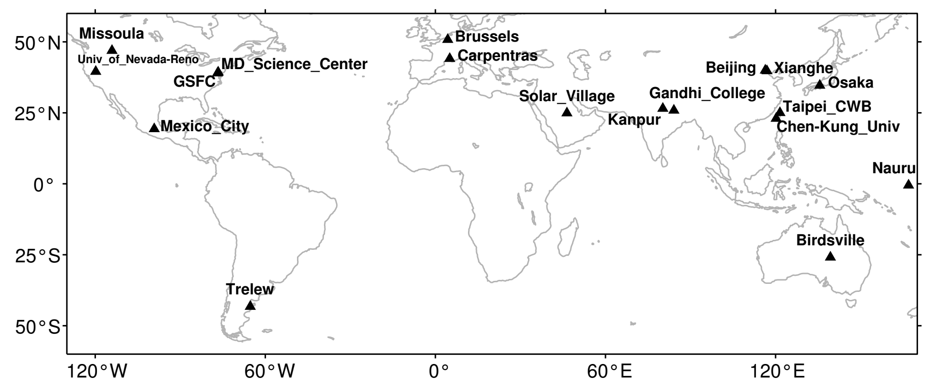

Figure 2. Locations of representative stations mentioned in the study.

The AE parameter describes the slope of the logarithm of AOD versus the logarithm of wavelength (Ångström, 1929), characterizing the wavelength dependency of AOD. AERONET AE_{440_870} products are calculated from the linear regression of AOD and wavelengths on a logarithmic scale within the range of 440-870 nm (Eck et al., 1999; Giles et al., 2019). The AE parameter closely correlates with aerosol particle size distribution, and is an indicator of aerosol components. For example, dust particles typically have AE_{440_870} values around 0.3 or lower, and the AE_{440_870} for fine-mode particles that are mostly anthropogenic, usually exceed 1.0 (Farahat et al., 2016; Giles et al., 2012; Russell et al., 2010; Dubovik et al., 2002). Therefore, AE_{440_870} can reflect the relative fraction of fine and coarse mode particles. The error in AE can be estimated by the error in

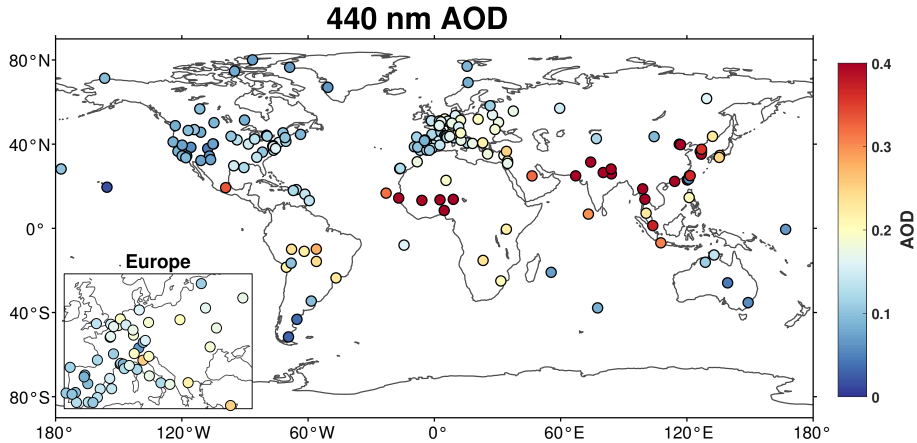

Figure 3. Mean AOD at 440 nm.

AOD as (Li et al., 2014; Kato et al., 2000):

$$\Delta \mathrm{AE} = \left[\frac{\sum_{i=1}^{n} e_i^2}{(n-2)\sum_{i=1}^{n}(\ln\lambda_i - \overline{\ln\lambda})^2} \right]^{\frac{1}{2}} \tag{1}$$

where e_i is the error of the Ångström relationship, n is the number of wavelengths λ_i used to fit the Ångström relationship, and $\overline{\ln\lambda}$ is the average of the logarithm of the wavelengths. e_i can be estimated using the relative error of AOD ($\frac{\Delta \mathrm{AOD}}{\mathrm{AOD}}$), and the uncertainty of AERONET AOD (ΔAOD) is considered as 0.01 here. According to Eq. 1, the uncertainty of AE is roughly inversely proportional to AOD, with larger errors at lower AOD conditions. Li et al. (2014) evaluated that the uncertainty of AE_{440_870} was 0.33 when $\mathrm{AOD}_{440} = 0.15$, and the uncertainty would rapidly increase to 0.56 when AOD_{440} decreased to 0.08. Eck et al. (1999) also demonstrated significant variability in AE_{440_870} for lower AOD, largely attributed to increased relative errors in AOD at these low values. These results correspond to the inverse relationship between ΔAE and AOD. Therefore, it should be noted that AE_{440_870} is highly uncertain and the AE_{440_870} trends are less robust for sites with low AOD, even if the trends are statistically significant.

AAOD and SSA together characterize the scattering and absorbing properties of aerosols. AAOD represents the total aerosol absorption optical depth, whereas SSA reflects the relative contribution of scattering to total extinction. Therefore, the AAOD trend directly reflects changes in the amount of absorbing aerosols, while the SSA trend is related to variations of both absorbing and scattering aerosols. The relationship between the two parameters can be expressed as the following equation:

$$\mathrm{AAOD} = (1 - \mathrm{SSA}) \times \mathrm{AOD} \tag{2}$$

The uncertainties of AAOD and SSA are also closely related to AOD level. AERONET implements a series of quality control criteria for Level 2.0 inversion products. Under these controls, AERONET SSA have an error of ± 0.03 when $AOD_{440} \sim 0.4$, and the error increases rapidly (exponentially) at lower AOD levels, i.e., an error of ± 0.05 when $AOD_{440} \sim 0.2$, and of ± 0.07 when $AOD_{440} \sim 0.1$ (Sinyuk et al., 2020). Therefore, although we utilize all the Level 2.0 quality assurance criteria except for the AOD threshold for AAOD and SSA data, many of the SSA retrievals in this study have larger uncertainties of ~ 0.03 to ~ 0.09 due to low AOD level. Moreover, as SSA typically varies from approximately 0.8 to 1.0 (Dubovik et al., 2002; Giles et al., 2012) , this error is remarkable when examining the variation of AAOD and SSA, i.e. a 0.03 error in SSA would lead to a 15% uncertainty. Therefore, the great uncertainties of AAOD and SSA should be kept in mind when analyzing their trends, especially for regions with low aerosol loadings.

2.3 Mann-Kendall test and Sen's slope for trend analysis

Here we use the Sen's slope combined with Mann-Kendall test to estimate the trend and its significance. The Mann-Kendall (MK) test (Mann, 1945; Kendall, 1975) is a nonparametric method to assess the significance of monotonic trends in a dataset without assuming any particular distribution. The slope of the trend k can be estimated by the median of the set of slopes (Sen, 1968):

$$k = \mathrm{Median}\left(\frac{Y_j - Y_i}{t_j - t_i}\right), \forall j > i \tag{3}$$

where Y_i and Y_j are the values of the variable at times t_i and t_j, respectively.

The significance of the trend could be tested by calculating the MK statistic:

$$S = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \mathrm{sgn}(Y_j - Y_i) \tag{4}$$

where

$$\mathrm{sgn}(x) = \begin{cases} -1 & \text{if } x < 0, \\ 0 & \text{if } x < 0, \\ 1 & \text{if } x > 0. \end{cases} \tag{5}$$

which has a normal distribution with zero mean and variance of (Li, 2020):

$$\sigma^2 = \frac{n(n-1)(2n+5) - \sum_{j=1}^{p} t_j(t_j - 1)(2t_j + 5)}{18} \tag{6}$$

The Sen's slope is a robust measurement of the trend in a dataset, and is not sensitive to outliers. As aerosol optical parameters do not follow a normal distribution, and AERONET records often have missing data, the Sen's slope is a good estimator of trends.

Table 1. Criteria of aerosol classifications defined in Lee et al. (2010).

Aerosol type	FMF_{550}	SSA_{440}	Proportion
Dust	$FMF_{550} < 0.4$	$SSA_{440} \leq 0.95$	14.4%
Mixture	$0.4 \leq FMF_{550} \leq 0.6$	/	17.2%
Non-absorbing Fine (NA)	$FMF_{550} > 0.6$	$SSA_{440} > 0.95$	22.6%
Slightly-absorbing Fine (SA)	$FMF_{550} > 0.6$	$0.9 < SSA_{440} \leq 0.95$	21.6%
Moderately-absorbing Fine (MA)	$FMF_{550} > 0.6$	$0.85 < SSA_{440} \leq 0.9$	11.4%
Highly-absorbing Fine (HA)	$FMF_{550} > 0.6$	$SSA_{440} \leq 0.85$	10.3%
Uncertain	$FMF_{550} < 0.4$	$SSA_{440} > 0.95$	2.5%

2.4 Aerosol Classification

In addition to the retrieved parameters, we also classify the observations into six aerosol types using the Fine Mode Fraction (FMF) at 550 nm and SSA at 440 nm (Lee et al., 2010). AOD and fine-mode AOD at 440, 675, 870, and 1020 nm are first interpolated to 550 nm using a second-order polynomial fit on a logarithmic scale (Eck et al., 1999). Then the FMF_{550} is calculated by AOD and fine-mode AOD at 550 nm. The classification criteria for the six aerosol types (~~Dust~~"Dust", "Mixture", ~~Mixture,~~ and four fine-mode types), as well as the proportion of each type in the total number of quality-controlled Level 1.5 all-point record, are listed in Table ~~1.~~ 1. It should be noted that sea salt aerosols ~~that typically have~~ typically having FMF_{550} below 0.4 and SSA_{440} ~~greater than 0.95 (denoted as the Uncertain~~ around 0.98 (included in the "Uncertain" type in Table 1) are not considered in ~~this study~~the analysis of aerosol type trends (Sect. 3.3), because most AERONET stations are located ~~on the mainland~~ over land where sea salt is not the predominant type, and sea salt aerosols only account for a negligible proportion (about 2.5% for "Uncertain" type).

Each quality-controlled Level 1.5 inversion all-point measurement is classified as a specific aerosol type according to the classification criteria in Table 1. ~~Since the records for~~ For each aerosol type~~are usually too few to calculate a monthly value (i.e., less than 5 measurements per month), we calculate the seasonal median values instead for the type analysis. Specifically, we~~ , we use coincident Level 2.0 AOD_{440} measurements to calculate the ~~seasonal AOD and analyse its trendfor each aerosol type~~annual AOD and analyze its trend.

3 Results

3.1 Trends for AOD and AE

The AOD_{440} trends at the ~~165~~ 172 selected AERONET stations are presented in Fig. 4. Trends surpassing the 90% significance level are marked with ~~black circles, with larger dotsdenoting trends exceeding the 95% level. A global reduction of aerosol loading is found with most stations demonstrating significant negative AOD_{440} trends, which is consistent with previous studies~~

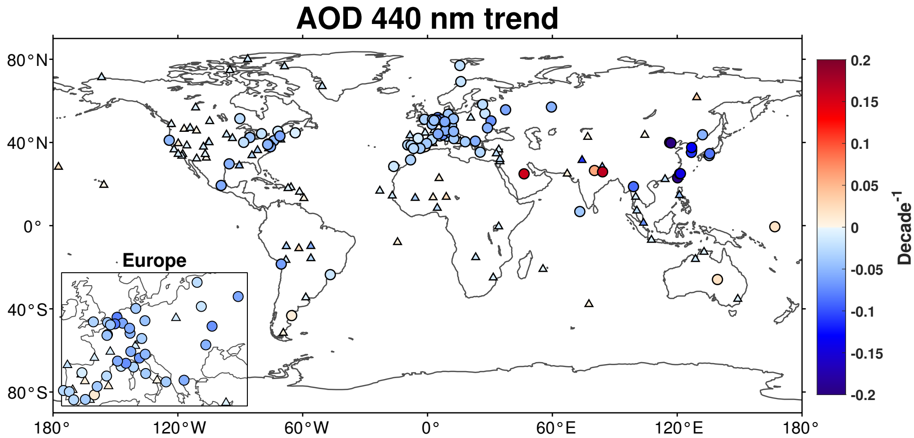

Figure 4. Trends of 440 nm AOD at AERONET stations. ~~Dots without black boundary~~ Triangles indicate trends below 90% significance level. ~~Smaller dots with black boundary~~ Dots indicate trends at 90% significance~~, and larger dots with black boundary indicate trends at 95% significance~~. The magnitude of the trend has the unit of [per decade].

~~(Li et al., 2014; Xia, 2011; Ningombam et al., 2019)~~dots. Trends below the 90% significance level are marked with triangles. The AOD$_{440}$ time series at several representative sites are ~~also~~ shown in Fig. 5. Significant negative AOD$_{440}$ trends are found for the majority of stations all over the world, demonstrating a global reduction of aerosol loading. This result is consistent with previous studies (Li et al., 2014; Xia, 2011; Ningombam et al., 2019). An increased number of stations with significant trends compared to these previous studies are observed in North America, Europe, and ~~North Africa~~the Mediterranean, likely due to spatial and temporal expansion of the network in recent years. The rates of AOD$_{440}$ reduction in western Europe (about -0.05 per decade) are not as substantial as those reported in Li et al. (2014), ~~suggesting a decelerating trend of aerosol reduction~~ which was -0.1 per decade, suggesting a decelerated aerosol reduction rate in Europe in recent years. This is also in line with the AOD$_{440}$ time series at representative European sites (Fig. 5g,h). Stations in the Arctic also exhibit coherent negative AOD$_{440}$ trends, consistent with previous studies (Breider et al., 2017; Wang et al., 2018). Since a significant proportion of aerosols in the Arctic are transported from lower latitudes, the reduction of aerosols in the Arctic is in line with the general reduction of AOD observed in the Northern Hemisphere. Strong negative AOD$_{440}$ trends are identified at more than 10 stations in East Asia and Southeast Asia, which were previously reported as exhibiting no significant trends in global studies (Li et al., 2014; Xia, 2011; Ningombam et al., 2019). The most considerable AOD$_{440}$ reductions are observed in East China, with ~~declines exceeding -0.1~~ significant declines exceeding -0.15 per decade at all the ~~five~~ four stations (Chen-Kung_Univ, XiangHe, Taipei_CWB, ~~Beijing, and Hong_Kong_PolyU) and almost~~ and Beijing) and reaching -0.2 per decade at Chen-Kung_Univ ~~, XiangHe, and Taipei_CWB. East China was reported to have increased aerosol loading in~~ and XiangHe. However, the trend of AOD$_{440}$ in East Asia is not coherent throughout the period of 2000-2022. According to the AOD$_{440}$ time series (Fig. 5a-c), AOD$_{440}$ increased in the early 2000s~~(Yoon et al., 2012; de Meij et al., 2012; Ramachandran and Rupakheti, 2022), thus the substantial AOD reductions found in this study, which were also reported in regional studies employing recent~~

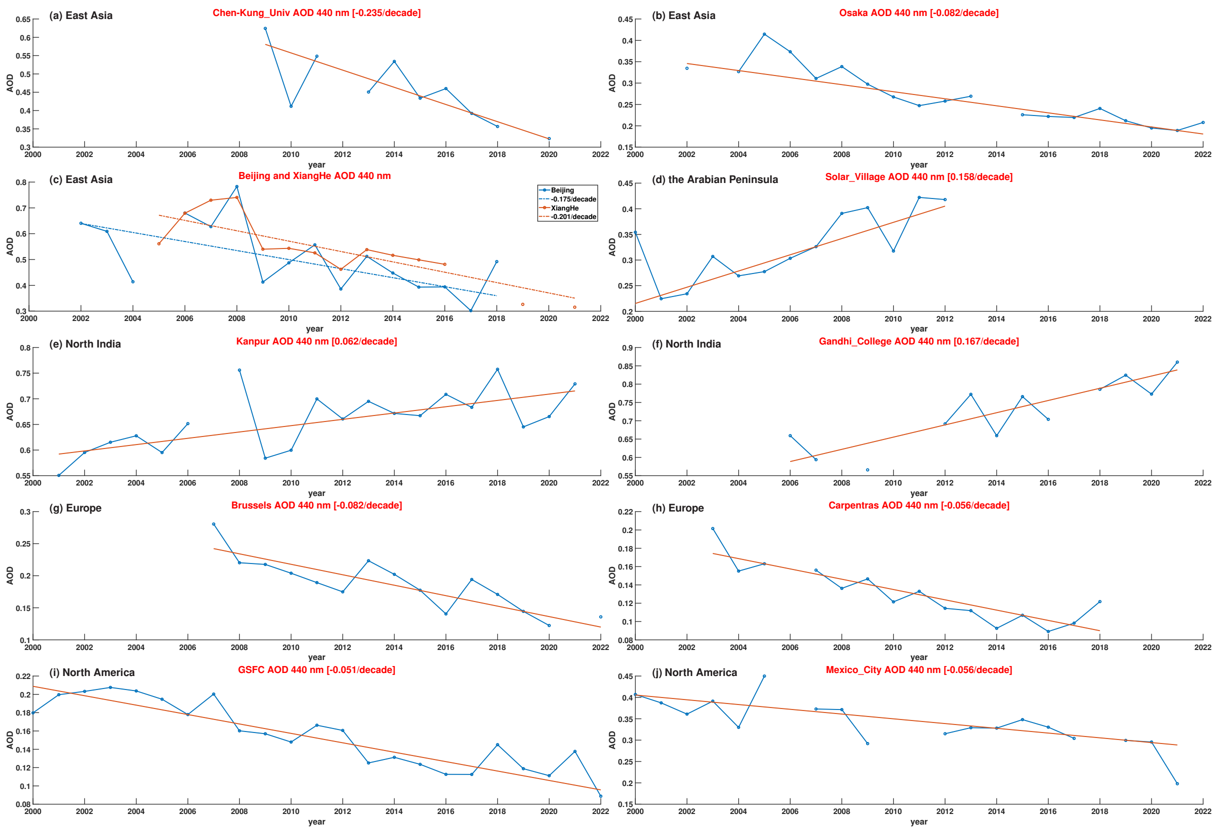

Figure 5. Time series of 440 nm AOD at several representative AERONET stations with trends at ~~95~~90% significance. (a) Chen-Kung_Univ, (b) Osaka, (c) Beijing and XiangHe, (d) Solar_Village, (e) Kanpur, (f) Gandhi_College, (g) Brussels, (h) Carpentras, (i) GSFC, (j) Mexico_City.

~~records(Ramachandran and Rupakheti, 2022; Ramachandran et al., 2020; Yu et al., 2022; Eom et al., 2022; Li, 2020; Gupta et al., 2022), have occurred mainly in the last decade. Xianghe and Beijing~~ , and decreased rapidly in the later years since around 2008, consistent with other regional aerosol trend studies (Yoon et al., 2012; de Meij et al., 2012; Ramachandran and Rupakheti, 2022; Ramachan . This result also explains why Li et al. (2014) found no significant AOD_{440} in East Asia with shorter records, as the increase of AOD_{440} in the early 2000s offset the reduction after 2008. When applying longer records, the continuous reduction of AOD_{440} after 2008 become dominant. A comparation between AOD_{440} time series of XiangHe and Beijing (Fig. 5c), two stations located very ~~near~~ close to each other in East China, ~~both~~ would further reveal that the substantial reduction of AOD_{440} mainly occurred in the later years. Both statons possess Level 2.0 records spanning ~~19 yearsfrom 2000 to 2022 (Fig. 5e).~~ a period of 17 years. However, the data record ~~in~~ for Beijing, starting in ~~2001~~2002 and ending in 2018, reveals an AOD_{440} trend of ~~-0.115~~ -0.175 per decade, whereas that ~~in Xianghe~~for XiangHe, starting in ~~2004~~2005 and ending in 2021, is more recent and exhibits a larger AOD_{440} decrease of ~~-0.179~~ -0.201 per decade, emphasizing the later years as a period of most notable AOD_{440} reduction.

Significant positive AOD_{440} trends are ~~found over Kanpur and Gandhi_College in North India,~~ mainly found over Solar_Village in the Arabian Peninsula, ~~Birdsville in Australia, two stations in South America (Trelew and CEILAP-RG), and several oceanic island stations. Note that several of these sites (Birdsville, Trelew, CEILAP-RG, and some oceanic sites) have very low AOD_{440} (typically below 0.1 for monthly values) as well as low AOD_{440} variability, therefore the results in these stations are typically more uncertain~~and over Kanpur and Gandhi_College in North India. The Level 2.0 AOD_{440} records at Solar_Village (Fig. 5d) ended in 2013, limiting current insights into aerosol properties in the Arabian Peninsula. Kanpur (Fig. 5e) has extensive records over the past two decades, exhibiting a positive AOD_{440} trend of ~~0.060~~ 0.062 per decade. This value is close to the trends calculated from different periods in previous studies (Ramachandran and Rupakheti, 2022; Li et al., 2014; Kaskaoutis et al., 2012; Kumar et al., 2022), indicating a steady increase in AOD_{440} there. Compared to previous global studies, an additional station named Gandhi_College (Fig. 5f) in northern India is observed to have a significant positive AOD_{440} trend of ~~0.149~~ 0.167 per decade, indicating a more pronounced increase in aerosol loading in this region. ~~Positive~~ Significant positive AERONET AOD_{440} trends over the other regions, such as Birdsville in Australia, Trelew in South America, and Nauru, an oceanic island station, are generally weaker, with magnitudes typically below 0.03 per decade. As these sites have very low AOD_{440} (typically below 0.1 for monthly values) as well as low AOD_{440} variability, the results in these stations are typically more uncertain. The positive AOD trend for Birdsville in Australia was confirmed by the independent research conducted by Yang et al. (2021), however this was a false trend resulting from a previously mentioned data screening anomaly. ~~Hsu et al. (2012) also suggested an increase in oceanic AOD, consistent with the widespread positive trends at oceanic stations~~ In addition to Nauru which exhibits significant positive AOD_{440} trend, some other oceanic stations worldwide also exhibit positive AOD_{440} trends, suggesting a widespread increase in oceanic aerosols, primarily sea salts. This result is consistent with Hsu et al. (2012) who also reported an increase in oceanic AOD.

~~The AE_{440_870} parameter characterizes the wavelength dependency of AOD and closely correlates with aerosol particle size distribution. Dust particles typically have AE_{440_870} values around 0.3 or lower, and the AE_{440_870} for fine-mode particles that are mostly anthropogenic, usually exceed 1.0 (Farahat et al., 2016; Giles et al., 2012; Russell et al., 2010; Dubovik et al., 2002). Therefore, AE_{440_870} can reflect the relative fraction of fine and coarse mode particles. The error in AE can be estimated by the error in AOD as (Li et al., 2014; Kato et al., 2000):~~

$$\Delta \mathrm{AE} = \left[\frac{\sum_{i=1}^{n} e_i^2}{(n-2)\sum_{i=1}^{n} (\ln \lambda_i - \overline{\ln \lambda})^2} \right]^{\frac{1}{2}}$$

~~where e_i is the error of the Ångström relationship, n is the number of wavelengths λ_i used to fit the Ångström relationship, and $\overline{\ln \lambda}$ is the average of the logarithm of the wavelengths. e_i can be estimated using the relative error of AOD ($\frac{\Delta \mathrm{AOD}}{\mathrm{AOD}}$), and the uncertainty of AERONET AOD (ΔAOD) is considered as 0.01 here. According to Eq. 1, the uncertainty of AE is roughly inversely proportional to AOD, with larger errors at lower AOD conditions. Li et al. (2014) evaluated that the uncertainty of AE_{440_870} was 0.33 when $AOD_{440} = 0.15$, and the uncertainty would rapidly increase to 0.56 when AOD_{440} decreased to 0.08.~~

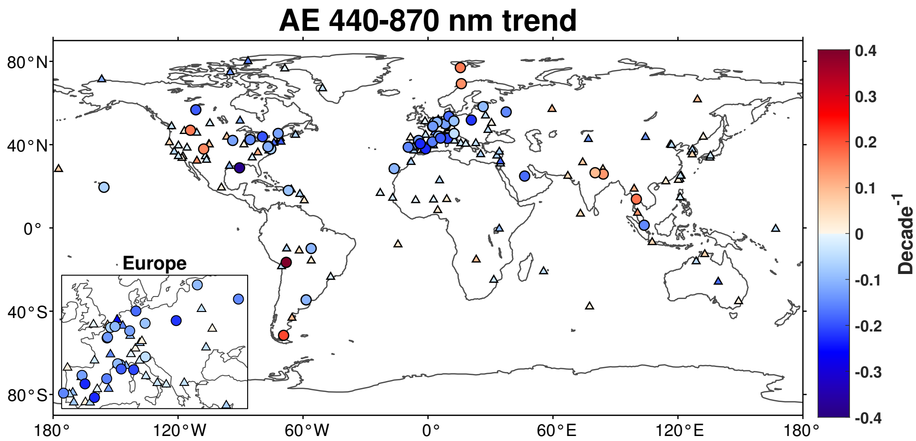

Figure 6. Same as Fig. 4, but with trends of AE.

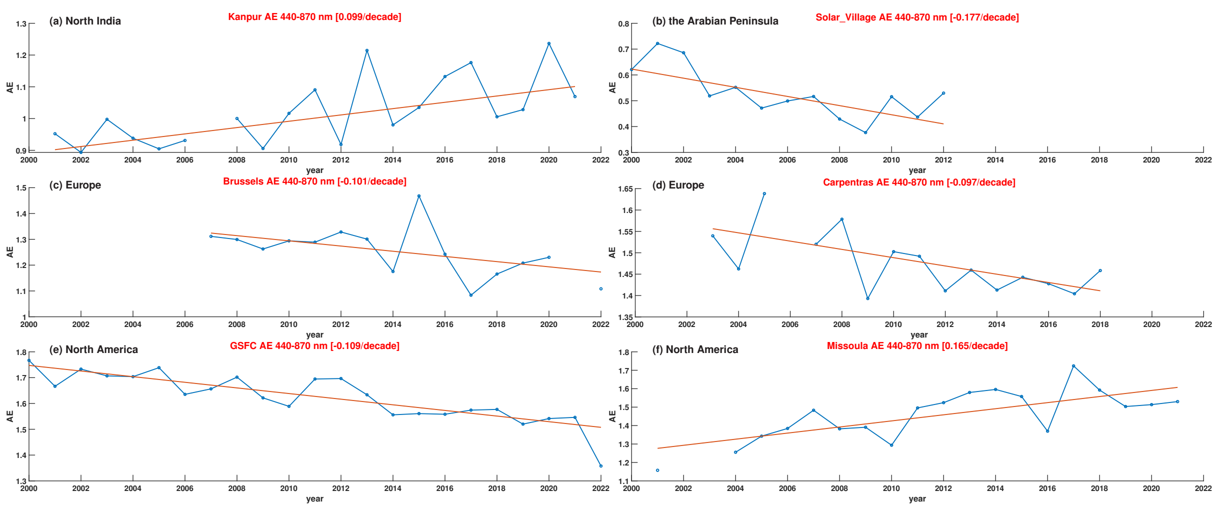

Figure 7. Time series of AE at several representative AERONET stations with trends at ~~95~~90% significance. (a) ~~Chen-Kung_Univ~~Kanpur, (b) Solar_Village, (c) ~~Kanpur~~Brussels, (d) ~~Gandhi_College~~Carpentras, (e) ~~Brussels~~GSFC, (f) ~~Carpentras, (g) GSFC, (h)~~ Missoula.

~~Eck et al. (1999) also demonstrated significant variability in AE_{440_870} for lower AOD, largely attributed to increased relative errors in AOD at these low values. These results correspond to the inverse relationship between ΔAE and AOD. Therefore, it should be noted that AE_{440_870} is highly uncertain and the AE_{440_870} trends are less robust for sites with low AOD, even if the trends are statistically significant.~~

Significant negative AE_{440_870} trends are universally found for stations across Europe, the Mediterranean, eastern North America, and the Arabian Peninsula ~~, and Middle Asia~~ (Fig. 6, Fig. 7). In contrast, stations in western North America ~~, North India , East Asia, and Southeast Asia~~ and North India mainly exhibit positive AE_{440_870} trends. The negative AE_{440_870} trends for Europe, the Mediterranean, and eastern North America are likely due to reductions in fine-mode anthropogenic aerosol and precursor emissions. In North India, considering the seasonal cycle of AE$_{440_870}$ value(Fig. 7c, d), , the positive AE$_{440_870}$ trends for Kanpur and Gandhi_College primarily result from increased fine-mode anthropogenic emissions as well as decreased coarse-mode dust loading. These shifts in anthropogenic emissions have been assessed through satellite observations and emission inventories (Pouliot et al., 2015; Szymankiewicz et al., 2021; Krotkov et al., 2016; Zhao et al., 2017; de Meij et al., 2012; Kumar et al., 2021), and the decline of dust loading over South Asia was also verified by satellite observations and AERONET measurements (Pandey et al., 2016, 2017; Ramachandran and Rupakheti, 2022; Kaskaoutis et al., 2011). The Arabian Peninsula is a well-known dust source (Ginoux et al., 2012) and the AE$_{440_870}$ values are typically low (Fig. 7b), therefore the negative AE$_{440_870}$ trend for Solar_Village is likely attributed to increased dust activities. Conversely, the increased AE$_{440_870}$ in western North America might be partly due to both increases in biomass burning aerosols and possibly diminished dust sources. These inferences align with previous studies, as Shao et al. (2013) also reported positive dust trends in the Middle East and negative trends in North America, whereas Eck et al. (2023) and Iglesias et al. (2022) revealed increases in biomass burning emissions over western North America. Over Asia , significant positive East Asia exhibits no significant AE$_{440_870}$ trendsare predominantly observed in the Korean Peninsula, the Indochina Peninsula, and the Taiwan Island, which might be linked to decreases in , indicating weak changes in the ratio of fine-mode and coarse-mode aerosols, such as the observed decline of dust transported from the mainland (Zhang et al., 2021; Kim et al., 2017; Cho et al., 2021), and is consistent with the decline in dust emissions in the mainland reported by previous studies (Wang et al., 2021a; Wu et al., 2022; Wang et al., 2021b; Zhao et al., 2018) . Notably, Beijing, Xianghe, and Hong_Kong_PolyU, despite showing substantial AOD$_{440}$ reductions, exhibit no significant AE$_{440_870}$ trends, which . Therefore, the great decrease of aersol loading in East Asia revealed in Fig. 4 might be related to similar reductions in both anthropogenic fine-mode aerosols and coarse-mode dust in these areas.

The spatial distribution of seasonal AOD$_{440}$ (Fig. 8) and AE$_{440_870}$ (Fig. 9) trends is generally similar to that of annual results. Nevertheless, the magnitude of the trends could vary by season, and certain stations might exhibit significant trends only during particular seasons. This variation is largely attributed to the seasonal patterns of aerosol emissions and meteorological conditions. For example, in Europe and North America, a greater number of stations exhibit significant AOD$_{440}$ trends in MAM (Fig. 8a), while AE$_{440_870}$ trends in DJF (Fig. 9d) are more pronounced and deviate more from the annual results. Stations in the Arctic mainly exhibit significant negative AOD$_{440}$ trends in MAM. This is because aerosol concentrations are typically higher in spring and lower in winter in the Northern Hemisphere (Fig. 5see supplementary), allowing for more substantial reductions in spring and more significant compositional variations in winter. However, AE$_{440_870}$ trends at low AOD seasons, such as the more pronounced AE$_{440_870}$ trends in winter in the Northern Hemisphere, should be treated with caution, because the uncertainty in AE$_{440_870}$ becomes large at low AOD conditions. In North India, Gandhi_College and Kanpur only exhibit significant AOD$_{440}$ trends in SON (during post-monsoon (October-November) (Fig. 8c) and DJF (winter (December-February) (Fig. 8d), while significant AE$_{440_870}$ trends predominantly occur in MAM (pre-monsoon (March-May) (Fig. 9a) and JJA (monsoon (June-September) (Fig. 9b). We can find that AE$_{440_870}$ values at these stations in North India significantly exceed 1.0 in SON and DJF (Fig. 7c, d), post-monsoon and winter, suggesting the predominance of fine-mode anthropogenic aerosols in these seasons. In contrast, AE$_{440_870}$ values in MAM and JJA pre-monsoon and monsoon start at approximately 0.5, emphasizing the dominance of coarse-mode aerosols, and rise to about 1.0 in recent years, suggesting a

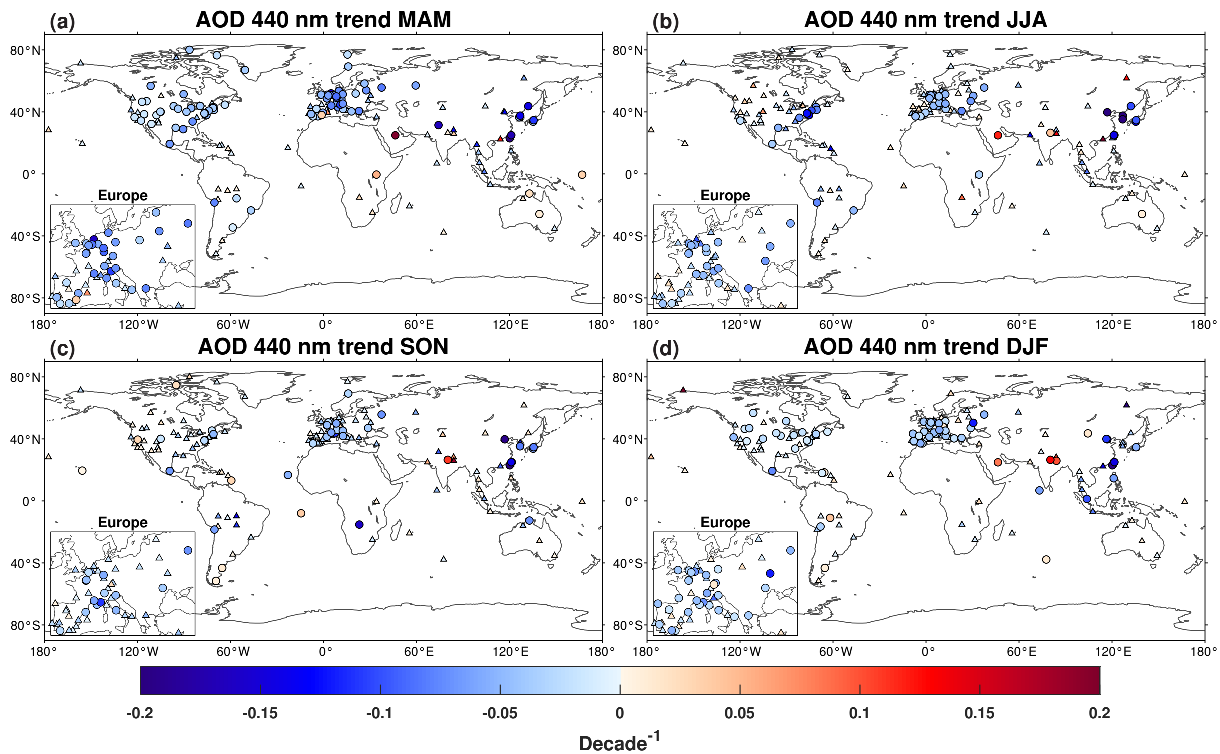

Figure 8. Seasonal trends of 440 nm AOD at AERONET stations. ~~Dots without black boundary~~ Triangles indicate trends below 90% significance level. ~~Smaller dots with black boundary~~ Dots indicate trends at 90% significance~~, and larger dots with black boundary indicate trends at 95% significance~~. The magnitude of the trend has the unit of [per decade]. (a) pre-monsoon (South Asia, March-May), peak (the Arabian Peninsula, March-June), spring (other sites, March-May). (b) monsoon (South Asia, June-September), post-peak (the Arabian Peninsula, July-October), summer (West Africa, April-October), summer(other sites, June-August). (c) post-monsoon (South Asia, October-November), autumn(other sites, September-November). (d) winter (South Asia, December-February), pre-peak (the Arabian Peninsula, November-February), Harmattan (West Africa, November-March), winter(other sites, December-February)

largely increased fraction of fine-mode aerosols. The seasonal patterns of AOD and AE in South Asia have also been verified through multi-year observations (Adhikary et al., 2007; Kaskaoutis et al., 2012). During the pre-monsoon ~~(March-May) and monsoon (June-September)~~ and monsoon seasons, higher wind speeds and stronger precipitation lead to stronger dust activities and higher wet scavenging of aerosols, whereas in the post-monsoon ~~(October-November) and winter (December-February)~~ and winter the meteorological conditions become reversed, with weaker dust activities and less efficient wet removal of aerosols occurred (Moorthy and Babu, 2006; Henriksson et al., 2011). As a result, in ~~SON and DJF~~post-monsoon and winter, the rises in anthropogenic emissions, mainly crop residue burning in post-monsoon and biofuel and fossil fuel burning in winter (Yin, 2020; Bhardwaj et al., 2015; Venkataraman et al., 2018), have a negligible impact on changes in aerosol compositions and AE_{440_870} values, but would lead to the significant positive AOD_{440} trends under less efficient wet removal. On the other hand, in ~~MAM and JJA~~pre-monsoon and monsoon, stronger wet scavenging of aerosols makes the AOD trend less pronounced, and

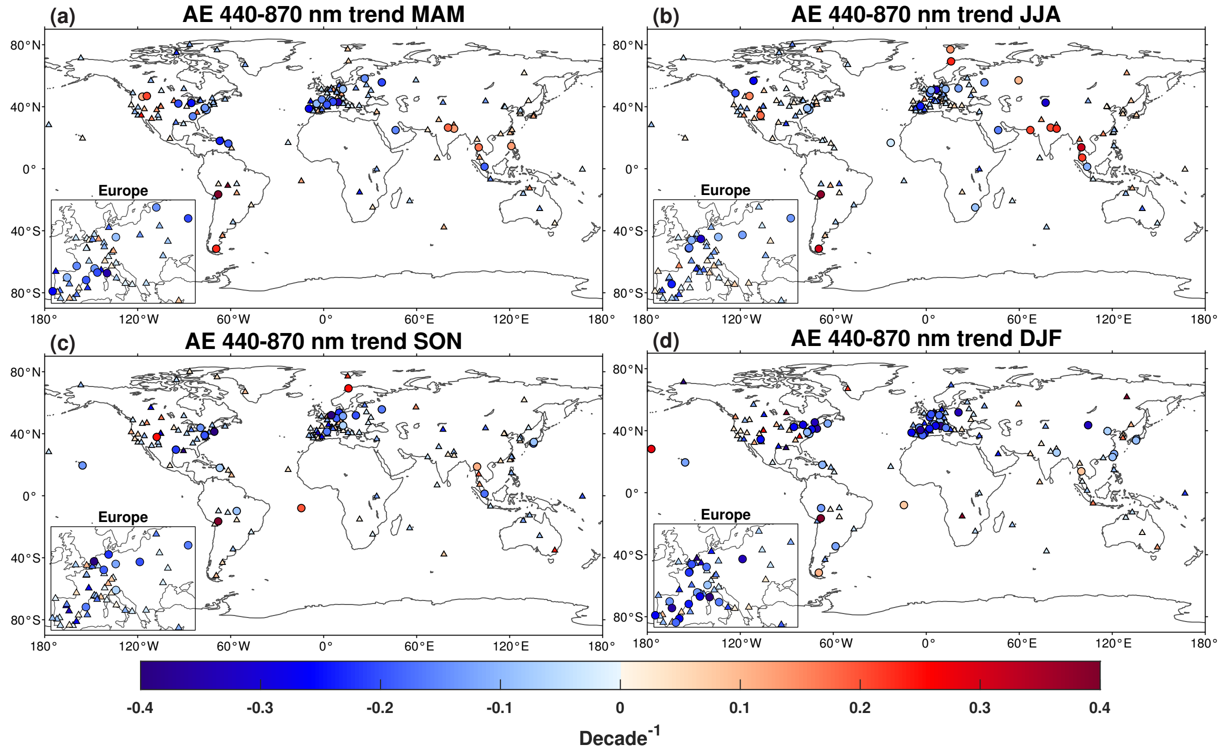

Figure 9. Same as Fig. 8, but with trends of AE.

the dominant aerosol type, dust, is mainly affected by natural variability (Kaskaoutis et al., 2012) and exhibits a negative trend (Pandey et al., 2017; Ramachandran and Rupakheti, 2022). Therefore, the increase in anthropogenic aerosols, i.e., biomass and biofuel burning emissions, fossil fuel emissions, and industry emissions (Venkataraman et al., 2018; Ramachandran and Rupakheti, 2022), does not have a significant impact on the total AOD_{440} in these two seasons, but serves to increase the fine mode fractions, leading to the insignificant AOD_{440} trends and significant positive AE_{440_870} trends.

3.2 Trends for AAOD and SSA

~~AAOD and SSA both characterize the scattering and absorption properties of aerosols. AAOD represents the total aerosol absorption optical depth, whereas SSA reflects the relative contribution of scattering to total extinction. Therefore, the AAOD trend directly reflects changes in the amount of absorbing aerosols, while the SSA trend is related to variations of both absorbing and scattering aerosols. The relationship between the two parameters can be expressed as the following equation:~~

$$\text{AAOD} = (1 - \text{SSA}) \times \text{AOD}$$

~~Before presenting the trends, it is important to acknowledge the uncertainties associated with AERONET inversion parameters. AERONET implements a series of quality control criteria for Level 2.0 inversion products. Under these controls, AERONET~~

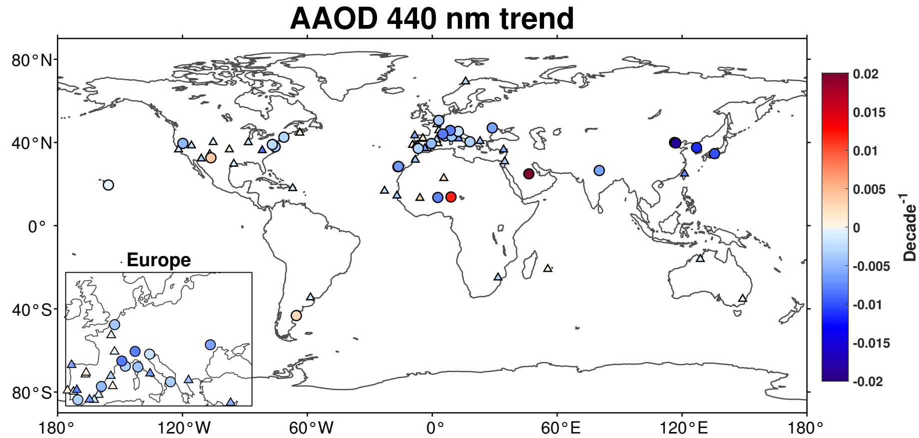

Figure 10. Same as Fig. 4, but with trends of AAOD.

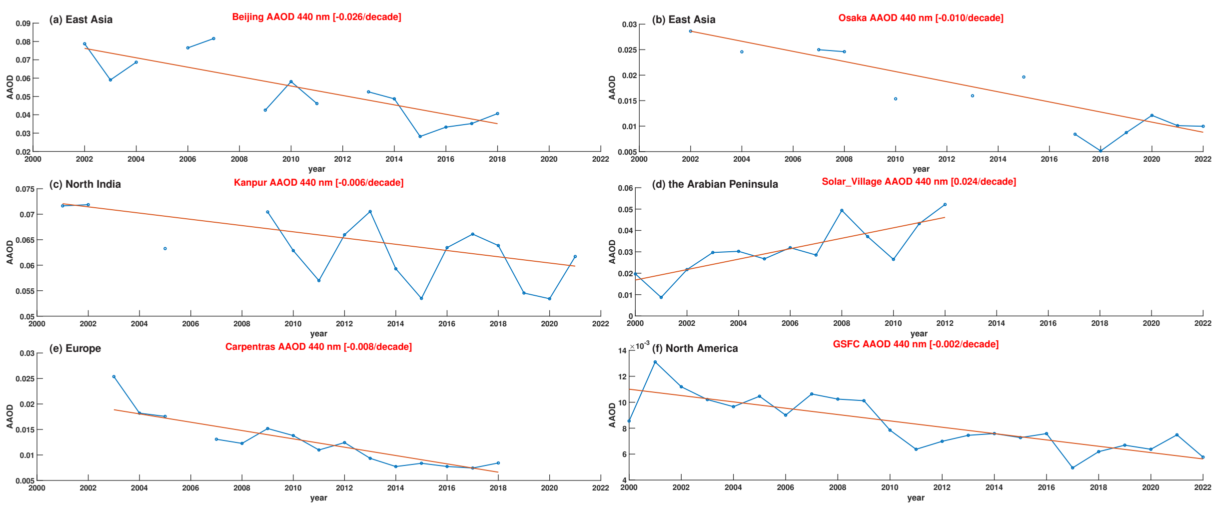

Figure 11. Time series of 440 nm AAOD at several representative AERONET stations with trends at ~~95~~90% significance. (a) Beijing, (b) Osaka, (c) Kanpur, (d) Solar_Village, (e) Carpentras, (f) GSFC.

~~SSA have an error of ±0.03 when AOD_{440} ∼ 0.4, and the error is even larger at lower AODs, i.e., an error of ±0.05 when AOD_{440} ∼ 0.2, and of ±0.07 when AOD_{440} ∼ 0.1 (Sinyuk et al., 2020). As SSA typically varies from approximately 0.8 to 1.0 (Dubovik et al., 2002; Giles et al., 2012), this error is remarkable when examining the variation of SSA and AAOD, i.e. a 0.03 error would lead to a 15% uncertainty. Therefore, the great uncertainties of these parameters should be kept in mind when analyzing trends in this section, especially for regions with low aerosol loadings.~~

Similar to AOD_{440}, significant negative $AAOD_{440}$ trends (Fig. 10, Fig. 11) are universally found for AERONET stations in the Northern Hemisphere, especially in East Asia, North India, Europe and North America, indicating reductions in absorbing species, mainly primary aerosols. Conversely, significant positive $AAOD_{440}$ trend is mainly found for Solar_Village in the

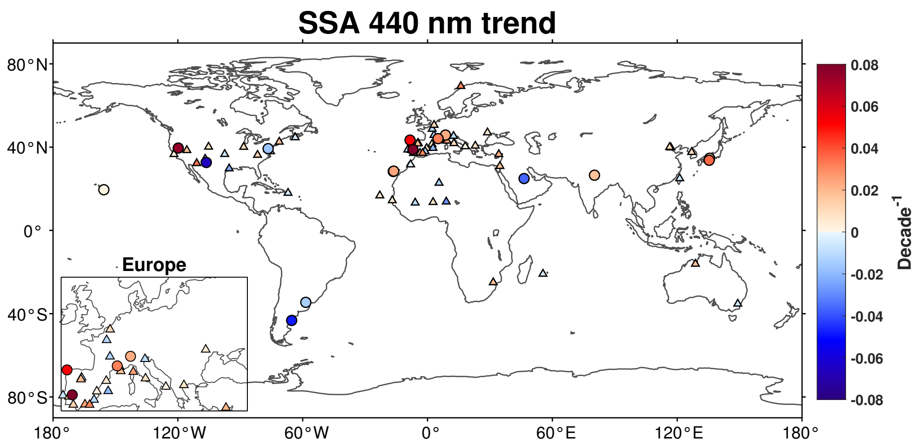

Figure 12. Same as Fig. 4, but with trends of SSA.

Arabian Peninsula (Fig. 11d), suggesting increases in absorbing aerosols. ~~Birdsville in Australia and~~ Trelew in southern South America also ~~exhibit~~ exhibits significant positive AAOD$_{440}$ trends, but the magnitude is very low and the results are relatively uncertain because the AAOD$_{440}$ are quite small~~in these sites~~. The reductions in AAOD$_{440}$ over East Asia, Europe, North India, and North America are primarily attributed to declines in anthropogenic emissions, such as reduced black carbon (BC) and/or organic carbon (OC) emissions from fossil fuels (Ramachandran and Rupakheti, 2022; He et al., 2023; Li et al., 2024), because aerosols in these regions are mainly of the Urban/Industrial type (Li et al., 2016). Decreased dust emissions discussed in the previous section might also be a potential contributor to the negative AAOD$_{440}$ trends in ~~East Asia, North India ,~~ North India and westren North America (Shao et al., 2013; Zhang et al., 2019; Wang et al., 2021b; Ramachandran and Rupakheti, 2022; Pandey et al., 2017), but the effect might not be as substantial as that of anthropogenic emissions, since dust is not the dominant type in these regions. Significant positive AAOD$_{440}$ trend for Solar_Village in the Arabian Peninsula is likely attributed to increased dust loading. As dust mainly exhibits strong absorption for short wavelengths, AAOD trends at other channels with longer wavelengths might not be that significant.

The SSA$_{440}$ trends (Fig. 12, Fig. 13) are generally opposite to the AAOD$_{440}$ trends, with exceptions in some stations in ~~central Europe and~~ North America. ~~The majority of stations~~ Stations in North India, East Asia, and Europe, ~~which also have negative AAOD$_{440}$ trends,~~ exhibit significant positive SSA$_{440}$ trends, corresponding to a decrease in the fraction of absorbing aerosols over time. ~~Remember that a rise in total aerosol loading is found for North India (Fig. 4), indicating~~ The increase of SSA$_{440}$ in North India is attributed to both the decrease in absorbing species and a more pronounced increase in scattering aerosols~~, such as sulfates, than the decrease in absorbing species~~. For East Asia and Europe, the ~~decreases in AOD$_{440}$ (Fig. 4) and AAOD$_{440}$ (Fig. 10) demonstrate possible reductions in both scattering and absorbing aerosols, while~~ positive SSA$_{440}$ trends ~~further~~ suggest stronger reductions in ~~asorbing species in these regions. Four stations in central Europe exhibit significant negative~~ absorbing species than scattering aerosols. SSA$_{440}$ trends ~~. SSA$_{440}$ trends~~ show large spatial heterogeneity in North America~~, with five stations exhibiting significant positive trends, four with significant negative trends , and four~~

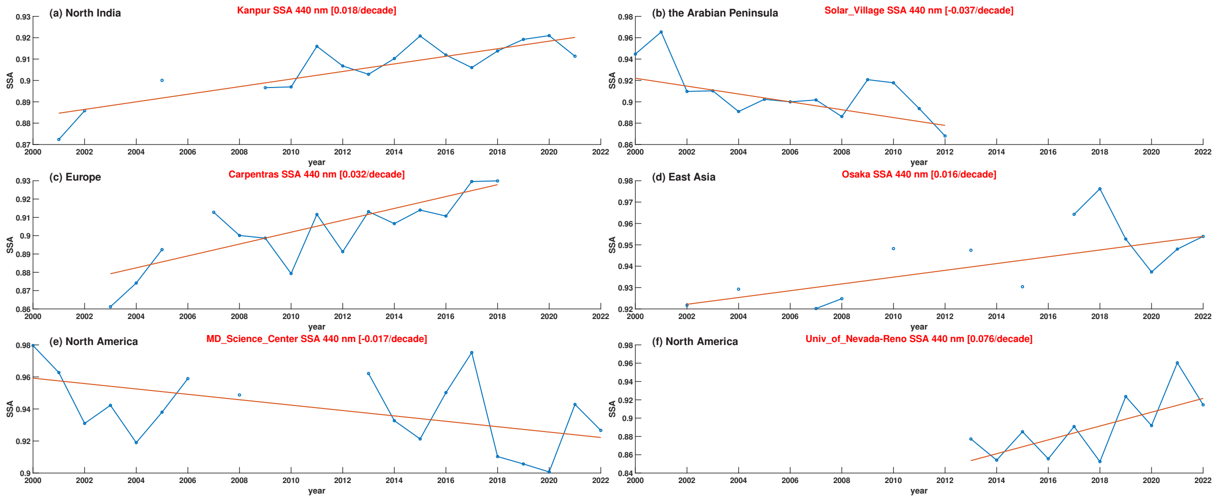

Figure 13. Time series of 440 nm SSA at several representative AERONET stations with trends at ~~95~~90% significance. (a) ~~Beijing~~Kanpur, (b) ~~Osaka~~Solar_Village, (c) ~~Kanpur~~Carpentras, (d) ~~Solar_Village~~Osaka, (e) ~~Carpentras~~MD_Science_Center, (f) ~~MD~~Univ_~~Science~~of_~~Center~~Nevada-Reno.

~~with weak negative trends not reaching the 90% significance threshold. Stations with decreased SSA_{440} over central Europe and North America exhibit only a mild decrease or even a slight increase in $AAOD_{440}$ (Fig. 10, Fig. 11f), implying that AOD_{440} reduction in these regions is mainly attributed to~~. Significant positive SSA_{440} trends are found for west coast, likely attributed to decreased absorbing aerosols. Significant negative trends are found over the center of the continent, corresponding to increase in absorbing aerosols. One station at east coast exhibits significant negative SSA_{440} trend, which is likely attributed to great reduction in scattering aerosols such as sulfates, thereby increasing the proportion of absorbing aerosols. This result aligns with Collaud Coen et al. (2020), which also found SSA reductions in ~~central Europe and~~ North America through in situ measurements, and attributed them to significant decreases in primarily scattering secondary aerosols. The substantial reductions in precursors of these scattering aerosols were also confirmed by satellite observations and emission inventories (Szymankiewicz et al., 2021; Fioletov et al., 2023; Krotkov et al., 2016; Tong et al., 2015). ~~Positive~~ However, several stations in eastern North America show positive trends which do not reaching the 90% significance threshold, emphasizing the spatial heterogeneity of SSA_{440} trends in this area. Negative SSA_{440} trend for Solar_Village (Fig. 13~~d~~b) in the Arabian Peninsula is attributed to increases in absorbing dust aerosols.

Seasonally, although some stations ~~, mainly in North America and Europe,~~ exhibit significant trends primarily during particular seasons, the spatial patterns of seasonal $AAOD_{440}$ (Fig. 14) and SSA_{440} (Fig. 15) trends are overall similar to those of annual trends. It is notable that Kanpur in North India exhibits stronger ~~and more significant~~ negative $AAOD_{440}$ trends ~~in MAM~~ during monsoon (Fig. 14~~a) and JJA (Fig. 14~~b) where dust is the dominant aerosol type, further verifying that the decreased $AAOD_{440}$ is partly attributed to the decline in dust loading. As for SSA_{440}, the positive trends for Kanpur are significant at all the four seasons, indicating that the increased anthropogenic emissions in North India are mainly scattering species.

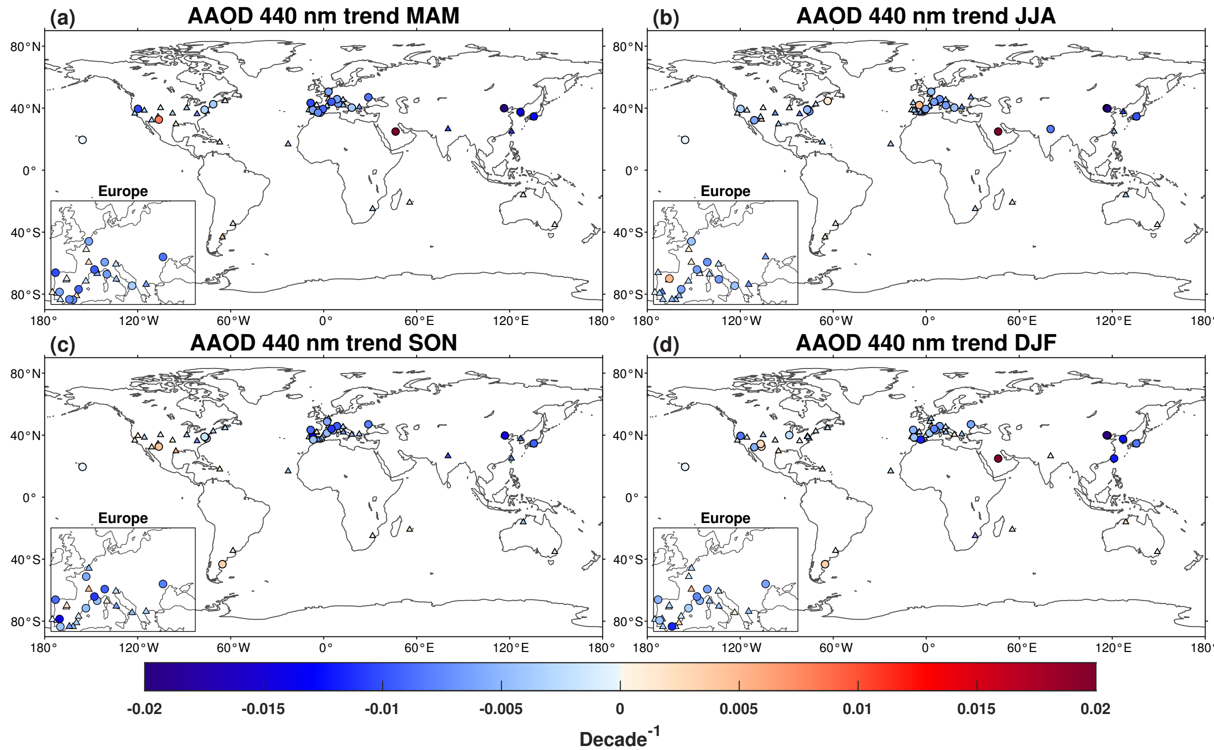

Figure 14. Same as Fig. 8, but with trends of AAOD.

3.3 Aerosol type changes

To better explain the aerosol parameter changes, we make a further attempt to classify the measurements into six aerosol types as described in Sect. ~~2.3~~2.4, and examine the long-term changes of the loadings for each type. The global AOD_{440} trends of the six aerosol types are shown in Fig. 16.

Significant positive trend for "Dust" AOD is found for Solar_Village, suggesting increased dust activities over the Arabian Peninsula, which is consistent with analysis in previous sections and other studies using satellite observations and AERONET measurements (Mehta et al., 2016; Habib et al., 2019; Sabetghadam et al., 2021; Al Otaibi et al., 2019; Li et al., 2014). We do not find significant trends over other dust sources, as dust loading can have strong decadal variability which often does not yield monotonic trends. Dust trend can also be difficult to detect when combined with fine mode anthropogenic aerosols. The "Mixture" type straddles the boundary between "Dust" type and fine-mode types and is affected by both coarse-mode and fine-mode particles. Significant negative AOD trends in the "Mixture" type are mainly found over East Asia and Europe (Fig. 16b). Since East Asia and Europe are both predominated by fine-mode aerosols (Li et al., 2016; Zhang and Li, 2019), the decreased "Mixture" aerosols are thus primarily due to reductions in fine-mode anthropogenic emissions.

The majority of stations in Europe, North America, and East Asia exhibit significant negative AOD trends in four fine-mode types (Fig. 16c-f), corresponding to the reduction in both absorbing and scattering anthropogenic emissions revealed by

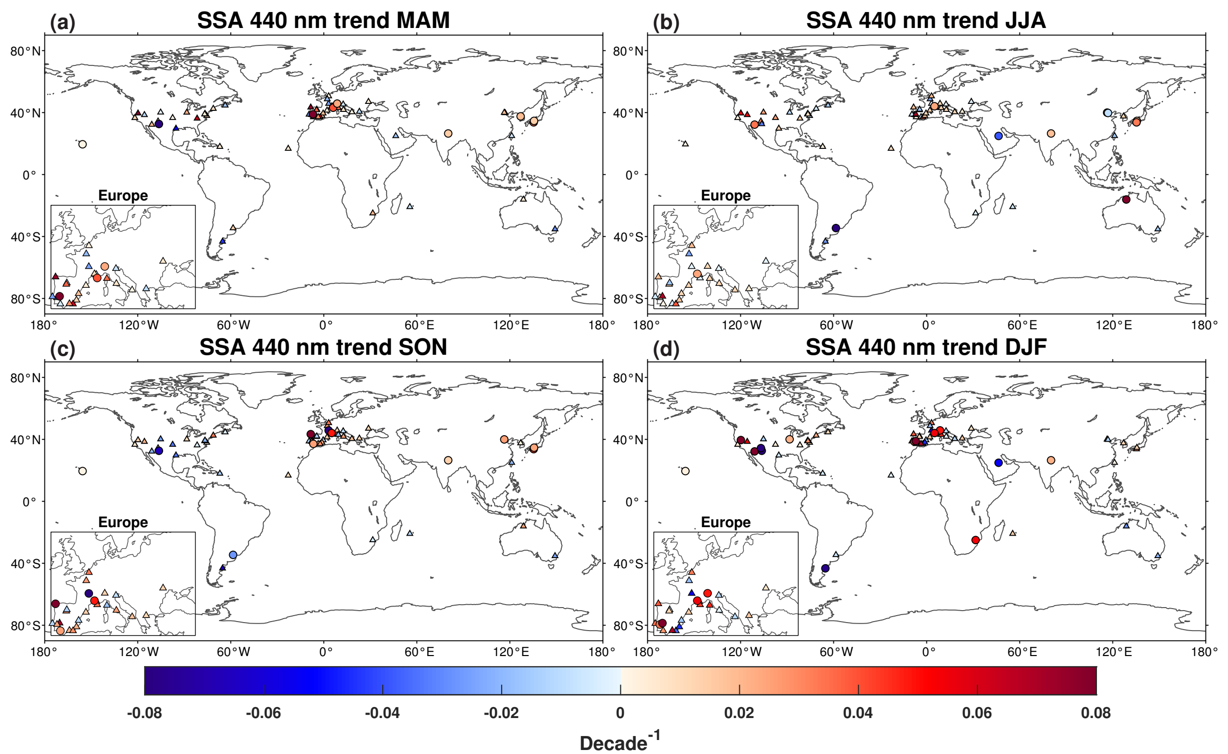

Figure 15. Same as Fig. 8, but with trends of SSA.

the reductions in AOD (Fig. 4) and AAOD (Fig. 10) in these regions. The great reduction in absorbing types (~~SA, MA and HA~~"SA", "MA" and "HA") is also the possible reason for the increase of SSA (Fig. 12). It is notable that ~~Xianghe~~ XiangHe in East Asia exhibits a significant positive trend in non-absorbing type (~~NA~~"NA") and even larger negative trends in the absorbing types, suggesting a great reduction in BC and/or OC emissions which might potentially lead to a shift in the predominance of aerosol type in pollution events (Zhang and Li, 2019). Eastern North America exhibits a greater reduction in non-absorbing aerosols than that in absorbing species, thus lead to an decrease in SSA (Fig. 12). Kanpur in North India exhibits significant positive trends on ~~SA~~ "SA" aerosols, and does not exhibit significant trends on other types. Compared to ~~MA and HA, the SA~~ "MA" and "HA", the "SA" type is more scattering with lower BC proportion. As fine mode aerosols in Kanpur are initially absorbing types (Pandey et al., 2016), the increase in ~~SA~~ "SA" loading suggests a decreased proportion of BC, making the fine mode aerosols in this region more scattering.

4 Discussion and Conclusion

In this study, we investigate trends in aerosol optical parameters using AERONET measurements. Globally, a universal decrease in AOD and AAOD, along with an increase in SSA, is observed at the majority of AERONET stations. The result generally

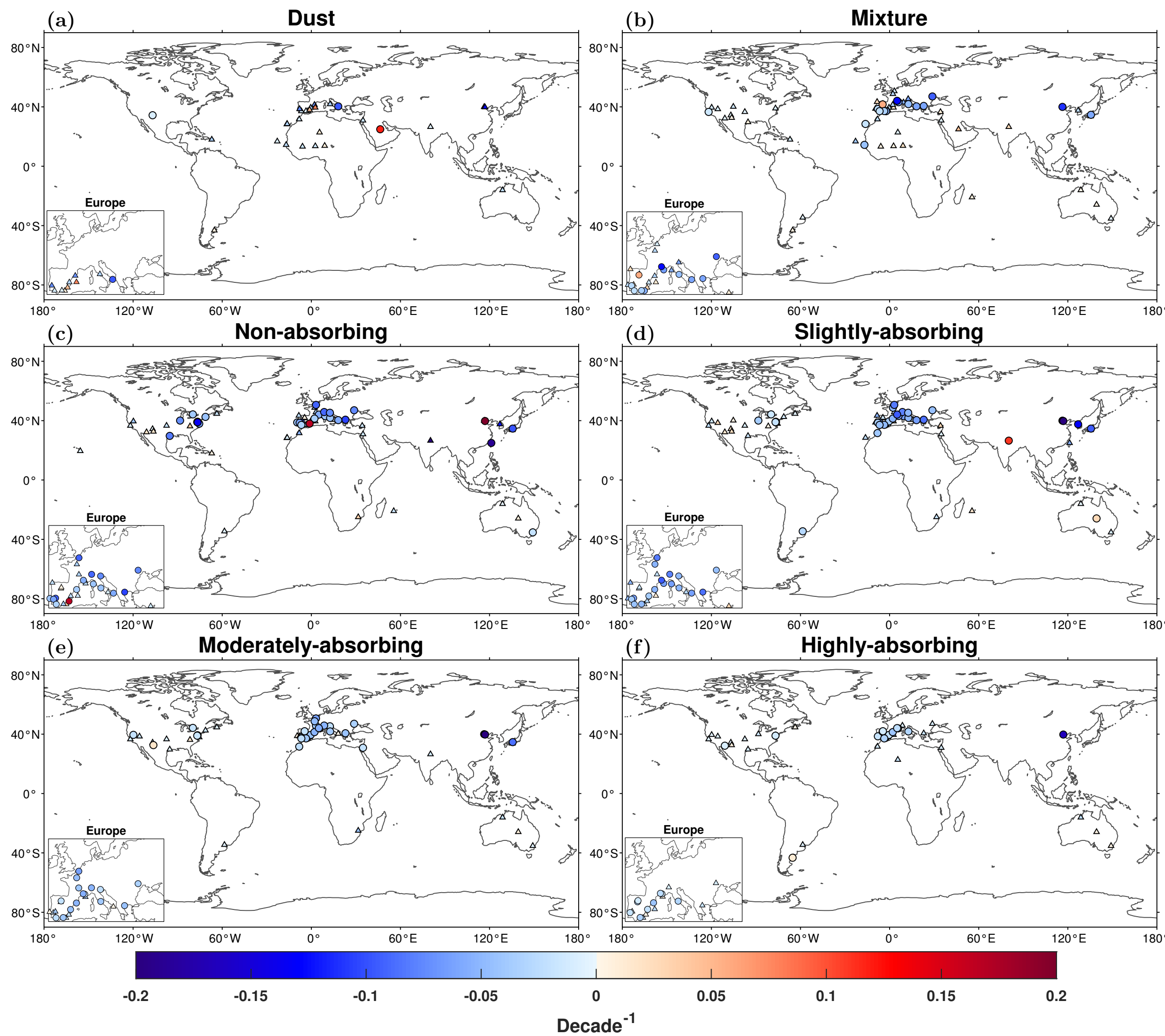

Figure 16. Same as Fig. 4, but with trends of AOD for 6 aerosol types. (a) dust, (b) Mixture, (c) Non-absorbing Fine, (d) Slightly-absorbing Fine, (e) Moderately-absorbing Fine, (f) Highly-absorbing Fine.

aligns with the previous trend analysis using AERONET Version 2 products ending in 2013 (Li et al., 2014), highlighting the continuity of these trends over time on a global scale. Although our analysis is based on measurements at ground-based stations, coherent spatial patterns over different stations could also indicate regional features, which have also been demonstrated by satellite observations, model simulations, and emission inventories (Gupta et al., 2022; de Meij et al., 2012; Fioletov et al., 2023; Mishchenko et al., 2007; Wei et al., 2021b, a; Yoon et al., 2016). ~~Similar to Li et al. (2014), no significant seasonalityis detected in the aerosol parameters examined in this work~~Although some regions, such as North India and western North America, have different seasonal and annual trends, the majority of regions do not exhibit significant seasonality. Taking advantages of longer records and improved station coverage, this study identifies more detailed regional trends and finds some new spatial patterns.

Spatially, significant negative AOD_{440} trends are universally observed across East Asia and Southeast Asia~~, which are not demonstrated by Li et al. (2014). This discrepancy indicates that the pronounced decrease in aerosols within these regions primarily occurred over the last decade, which is also supported by satellite observations and model simulations (Fioletov et al., 2023; de Me~~ ~~-~~. The most substantial reduction in AOD_{440} occurs in East China, consistent with emission inventories (Kurokawa and Ohara, 2020). ~~Correspondingly, Li et al. (2014) also reported no significant AE_{440_870} trends, while our analysis reveals significant positive AE_{440_870} trends at coastal stations, aligning with in situ measurements (Collaud Coen et al., 2020). The increase in AE_{440_870} implies a reduction in aerosol particle size, potentially due to decreased dust transportation from the mainland. Compared to Li et al. (2014), this~~ AOD_{440} time series demonstrates that the pronounced decreases in aerosols within these regions are not coherent in the whole period, with aerosol loading increased in the early 2000s and decreased in the later years around 2008, which is also supported by satellite observations and model simulations (Fioletov et al., 2023; de Meij et al., 2012; Zhao et al. . This study also finds ~~that more stations in mid-latitude East Asia exhibit~~ significant negative $AAOD_{440}$ trends and positive SSA_{440} trends for East Asia, which are mainly attributed to decreased absorbing primary aerosols, in agreement with other independent studies utilizing AERONET data (Ramachandran and Rupakheti, 2022; Ramachandran et al., 2020; Tao et al., 2017; Yu et al., 2022; Eom et al., 2022).

Coherent significant decreases of AOD_{440} and $AAOD_{440}$ found for Europe and North America are ~~consistent with Li et al. (2014), and are~~ in good agreement with satellite observations (Mehta et al., 2016; Zhao et al., 2017; Krotkov et al., 2016; Fioletov et al., 2023) as well as in situ measurements of recent aerosol absorbing and scattering trends (Collaud Coen et al., 2020). ~~These trends support the ongoing efforts in emission control throughout this century. However, while Li et al. (2014) observed smaller AOD_{440} trends in North America compared to Europe, this study reports similar and weaker trends for both~~ However, time series reveals diminished rates in the aerosol reduction in these regions. The decrease in anthropogenic emissions in Europe and North America started in the previous century and has led to a significant reduction in aerosol loading (de Meij et al., 2012; Szymankiewicz et al., 2021; Rafaj et al., 2013), resulting in a diminished rate of reduction in aerosol and aerosol precursor emissions over the last decade (Krotkov et al., 2016; Fioletov et al., 2023; Jiang et al., 2018). ~~Consequently, this potentially leads to smaller AOD_{440} and $AAOD_{440}$ trends, alongside a reduced discrepancy in AOD_{440} trends between the two regions. Additionally, the update to AERONET Version 3 and slight methodological differences in trend evaluation may also explain some inconsistencies in trend slope assessments.~~ The observed decline in AE_{440_870} and increase in SSA_{440} in Europe are also in line with ~~Li et al. (2014), while the positive~~ previous studies and suggest reductions in anthropogenic emissions (Li et al., 2014; Xia, 2011). Significant negative AE_{440_870} trends ~~found for the whole North Americaby Li et al. (2014) are mainly found in western North America in this work, with eastern North America exhibiting negative trends. This indicates that significant~~ are found for eastern North America, likely attributed to decline in anthropogenic emissions. Western North America exhibits significant positive AE_{440_870} trends which are likely related to reductions in dust emissions ~~are primarily concentrated in western North America along with~~ and increases in biomass burning emissions, ~~with the impact on eastern areas being less pronounced,~~ consistent with dust monitoring results (Aryal and Evans, 2022) and trends in western North America forest fires (Eck et al., 2023; Iglesias et al., 2022). ~~The SSA_{440} trends in this work also diverge from the coherent positive trends reported by Li et al. (2014), because we find~~ Significant negative SSA_{440} trends ~~for a small proportion of European stations and for more than half of the North American stations. This discrepancy is likely due to differences in study periods, and the trends observed in this work~~ are found for some stations over North America and align with those from in situ measurements conducted over similar periods (Collaud Coen et al., 2020), suggesting a larger decline in scattering aerosols than absorbing species.

~~The~~ Significant positive AOD_{440} ~~, AE_{440_870} and SSA_{440} trends for Kanpur~~ and AE_{440_870} trends are found for Kanpur and Gandhi_College in North India~~identified by Li et al. (2014) are corroborated in this work, suggesting increased~~ , highlighting an increase in fine-mode ~~anthropogenic aerosol loading. Unlike the report by Li et al. (2014) which found no significant~~ aerosols. We also find significant negative $AAOD_{440}$ trend and positive SSA_{440} trend ~~in AAOD~~ for Kanpur, ~~our research reveals a significant negative $AAOD_{440}$ trend, indicating recent~~ indicating decreases in absorbing aerosols in ~~the~~ this region, and we further attribute this change to both decreased anthropogenic BC emissions and decreased dust loading according to seasonal trend analysis and type analysis, consistent with previous studies (Pandey et al., 2016, 2017; Ramachandran and Rupakheti, 2022). These trends align with independent studies utilizing AERONET measurements (Ramachandran and Rupakheti, 2022; Kumar et al., 2022; and satellite observations (Ramachandran et al., 2020; Kaskaoutis et al., 2011), together verifying the alteration of aerosol compositions and suggesting that the increased aerosols are mainly scattering fine-mode species. The trends over North India exhibit strong seasonality, with significant positive AOD_{440} trends in ~~SON and DJF~~ post-monsoon and winter where anthropogenic aerosols are predominant, and decreased $AAOD_{440}$ and increased AE_{440_870} ~~in MAM and JJA~~ during pre-monsoon and monsoon where dust loading is stronger, suggesting that these seasonal trends may be associated with the seasonal cycle of aerosol emissions and meteorological conditions. ~~Furthermore, an additional site, Gandhi_College, located in North India as well, also exhibits significant positive AOD_{440} trends and positive AE_{440_870} trends, highlighting an increase in fine-mode aerosols across South Asia. The AOD_{440} and AE_{440_870} trends for both Kanpur and Gandhi College, along with $AAOD_{440}$ and SSA_{440} trends for Kanpur, align with independent studies utilizing AERONET measurements (Ramachandran and Rupakheti, 2022; Kumar and satellite observations (Ramachandran et al., 2020; Kaskaoutis et al., 2011), further verifying the increment in aerosols and the alteration of aerosol compositions in North India.~~

The AERONET products for Solar_Village end in 2013, therefore the trends of these aerosol optical parameters are the same as those reported by Li et al. (2014), with positive AOD_{440} and $AAOD_{440}$ trends, and negative AE_{440_870} and SSA_{440} trends, which is probably due to the increased dust activities in the Arabian Peninsula, and was also demonstrated in previous studies (Al Otaibi et al., 2019; Habib et al., 2019).

As a further step, we classify the aerosol observations into six types using FMF_{550} and SSA_{440}, and examine the changes in aerosol loadings of each type. The trends for different aerosol types further verify the trends of AERONET parameters and offer insights into aerosol composition changes. We only find significant positive dust loading trend in the Arabian Peninsula. Significant trends mainly concentrate on fine-mode types, with declines in both absorbing types and the non-absorbing type globally, consistent with the negative AOD_{440} and $AAOD_{440}$ trends. Spatially, the majority of stations in East Asia and Europe exhibit stronger reductions in absorbing aerosols than those in non-absorbing types, whereas in eastern North America the reduction in AOD_{440} is mainly attributed to non-absorbing species. The results can fully explain the changes in SSA_{440}, which exhibit positive trends over East Asia and Europe and negative trends over eastern North America. Significant positive ~~SA~~ "SA" loading trend found in North India suggests a decrease in BC proportion which leads to increased SSA_{440}.

This study provides insights into temporal variations in aerosol loading, optical properties, and aerosol types. Decreases in AOD across Europe, North America, and East Asia reflect the effectiveness of emission control policies implemented in these regions. For instance, there has been a significant reduction in AOD over China in the past decade due to the Air Pollution Prevention and Control Action Plan (Gupta et al., 2022; Zhao et al., 2017). Conversely, the increase of AOD over North India and the Arabian Peninsula indicates deteriorating air quality, posing potential risks to public health. The substantial changes in SSA and AAOD observed in many regions are of concern for climate models due to their critical relationship with aerosol climate effects, potentially influencing regional energy budget, atmospheric circulation, the water cycle, etc. Previous studies have indicated that failure to capture the increase in SSA over northern India in climate models likely contributed to their biases in simulating the negative precipitation trend in this region (Ying et al., 2023). Furthermore, trends in aerosol properties and types are crucial for satellite remote sensing applications, as many algorithms rely on assumed aerosol models clustered from AERONET observations. Updating these models to reflect changes in aerosol types may be necessary (Zhang et al., 2024).

It is important to note that our analysis extends through 2022, encompassing the COVID-19 pandemic. Previous studies have documented significant reductions in aerosol loading and notable changes in aerosol compositions due to decreased anthropogenic emissions in regions implementing lockdown policies, such as East Asia, Europe, and North America (Cao et al., 2021; Clemente et al., 2022; Liang et al., 2023; Sokhi et al., 2021). We observed abnormally low AOD values at certain stations during this period, including ~~Xianghe~~ XiangHe and Chen-Kung_Univ (Fig. 5a, c). This could potentially lead to a negative bias in AOD trends and contribute to discrepancies with other research on aerosol trends at these stations. However, since this period accounts for only about 10% of our total study period, and many stations lack Level 2.0 records for this time, the impact on trend analysis by COVID-19 is likely minimal at the majority of the stations.

The main purpose of this work is to update the trends in aerosol parameters with larger size of stations and longer records with respect to Li et al. (2014). We do note remarkable changes in aerosol trends over regions such as East Asia and the Southern Hemisphere, whereas patterns in other regions remain relatively stable. Most additional stations in this study are located in Europe and North America, where the distribution of stations is already dense to deduce general features of aerosol trends in these regions. We still lack insights into aerosol trends across other regions, including Asia, Africa, South America, Australia, and polar and oceanic regions where the spatial coverage of stations is insufficient, and some stations such as Solar_Village do not have Level 2.0 data in recent years. There is still need to establish more stations in Asia and the Southern Hemisphere to better capture the rapid change of aerosol properties there.

Author contributions. JL designed the research. TE, PG, BH, OD, and EL gathered the datasets and applied additional QAC to the data. ZZ selected the stations with long-term records, computed the trends, and analyzed the results. ZZ and JL prepared the manuscript draft.

YD, TE, PG, SNT, and JK reviewed and edited the manuscript. All the other co-authors contributed to the measurements of aerosol optical properties applied in this work and to the manuscript review.

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

Acknowledgements. We gratefully thank the AERONET team, especially the PIs and Co-Is and their staff of the 165 selected stations, for establishing and maintaining the sites and providing the data used in this study. The AERONET data are obtained from the AERONET website, https://aeronet.gsfc.nasa.gov/. This study is funded by the National Key Research and Development Program of China (grant no. 2023YFF0805401) and the National Natural Science Foundation of China (NSFC) Grants Nos. 42175144 and 42375121.

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