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Visibility-derived aerosol optical depth over global land from 1980 to 2021

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Abstract

Long-term and high spatial resolution aerosol optical depth (AOD) data are necessary for climate change detection and attribution. Global ground-based AOD observation stations are sparse, and satellite AOD observations have a low time frequency, as well low accuracy before 2000. In this study, AOD was derived from hourly visibility observations collected at more than 5000 stations of the Automated Surface Observing System (ASOS) over global land from 1980 to 2021. The AOD retrievals of the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua Earth observation satellite were used to train the machine learning method, and the ERA5 reanalysis boundary layer height was used as input. The predicted result has correlation coefficients of 0.54 and 0.51 with Terra MODIS satellite retrievals and AERONET ground observations. The correlation coefficients are higher at monthly and annual scales, which are 0.81 and 0.61 for the monthly and 0.91 and 0.62 for the annual, when compared with Terra MODIS and AERONET AOD, respectively. The visibility-derived AOD at ASOS stations was gridded into a 0.5-degree resolution by area-weighted ordinary kriging interpolation. Analysis of visibility-derived AOD indicates that the global mean AOD over land is 0.16, which is 0.24, 0.22, 0.11, 0.11, 0.130, and 0.12 for Africa, Asia, Europe, North America, Oceania, and South America, respectively. The mean AOD over global land, the Northern Hemisphere, and the Southern Hemisphere demonstrated decreasing trends of -0.0026/10a, -0.0018/10a, and -0.0059/10a, respectively, from 1980 to 2021. The visibility-derived AOD at station and grid scales over global land from 1980 to 2021 are available at National Tibetan Plateau / Third Pole Environment Data Center (https://doi.org/10.11888/Atmos.tpdc.300822) (Hao et al., 2023).

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https://doi.org/10.5194/essd-2023-447
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1 Introduction

Atmospheric aerosols are composed of solid and liquid particles suspended in the atmosphere. Aerosol particles are primarily discharged from the Earth's surface broadly classified into natural and anthropogenic sources (Calvo et al., 2013). They possess diverse shapes and sizes(Fan et al., 2021), optical properties, and various components (Li et al., 2022), such as inorganic salts, organic matter, metal elements and elemental carbon. Most atmospheric aerosols are concentrated in the troposphere, especially in the boundary layer (Liu et al., 2022), with a high concentration near emission sources (Kulmala et al., 2004) , and a small portion are distributed in the stratosphere with a sharp increase during large volcanic eruptions. Some aerosols from wildfires, volcanoes and sandstorms, play an important role in tropospheric aerosols. Studies have showed that 75% of volcanic eruptions inject volcanic aerosols and sulfur containing gases into the troposphere (Halmer et al., 2002), wildfire aerosols contribute up to approximately 35% of the fine particles in Europe (Barnaba et al., 2011), and dust aerosols are mainly concentrated in the middle and low troposphere (Filonchyk et al., 2018). Atmospheric aerosols severely impact the atmospheric environment and human health. They deteriorate air quality, reduce visibility, and cause other environmental issues (Wang et al., 2012; Boers et al., 2015). They affect human health or other organisms' conditions by increasing cardiovascular and respiratory disease incidence and mortality rates (Chafe et al., 2014; Yang et al., 2022). The Global Burden of Disease shows that global exposure to ambient $PM_{2.5}$ resulted in 0.37 million deaths and 9.9 million disability-adjusted life years (Chafe et al., 2014).

In addition to environmental and health impacts, aerosols are inextricably linked to climate change. Atmospheric aerosols alter the Earth's energy budget and then affect the climate (Li et al., 2022). They cool the surface and heat the atmosphere by scattering and absorbing solar radiation (Forster et al., 2007; Chen et al., 2022). Aerosols, such as black carbon and brown carbon, also absorb solar radiation (Bergstrom et al., 2007), heat the local atmosphere and suppress or invigorate convective activities (Ramanathan et al., 2001; Sun & Zhao, 2020). Aerosols also alter the optical properties and life span of clouds (Albrecht, 1989). Atmospheric aerosols strongly affect regional and global short-term and long-term climates through direct and indirect effects (McNeill, 2017).

Tropospheric aerosols are considered as the second largest forcing factor for global climate change (Li et al., 2022), and they reduce the warming due to greenhouse gases by -0.5°C (IPCC, 2021). However, aerosols are also regarded as the largest contributor to quantifying the uncertainty of present-day climate change (IPCC, 2021). The deficiency of the global descriptions of aerosol optical and microphysical properties is the primary reason for the uncertainty and the uncertainty also exists in climate models (Lee et al., 2016; IPCC, 2021). Therefore, sufficient aerosol observations are crucial. In aerosol measurements, aerosol optical depth (AOD) is often used to describe its column properties, which represents the vertical integration of aerosol extinction coefficients. AOD is an important physical quantity for estimating the content, atmospheric pollution and climatology of aerosols (Zhang et al., 2020).

The measurements of aerosols are usually divided into in-situ and remote sensing observations. In-situ observations accurately measure the mass, number concentration, shapes, compositions and scattering and absorption of aerosols by directly sampling the air (Herich et al., 2008; Laj et al., 2020). Observations from airplanes and balloons can provide vertical structure (Ziemba et al., 2013).

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Because of its accuracy, in-situ observation is often used as the benchmark for models and satellites, but its spatial representativeness is limited. Another method is ground-based lidar observation, which is an active remote sensing technology. Lidar generally emits laser and receives backscattered signals to invert the extinction coefficient of aerosols at different heights (Klett, 1985). By using the depolarization ratio, the type of aerosol, such as fine particles or dust, can also be distinguished (Bescond et al., 2013). The AOD within a certain height can be calculated by integrating the extinction coefficients; however, scattering signals are usually not received near the ground, leading to blind spots (Singh et al., 2019). At present, there are many ground-based lidar worldwide and regional networks, which provides important support in the study of vertical changes in aerosols, such as the NASA Micro-Pulse Lidar Network (MPLNET) in the early 1990s (Welton et al., 2002), the European Aerosol Research Lidar Network (EARLINET) since 2000 (Bösenberg & Matthias, 2003), the Latin American Lidar Network (LALINET) since 2013 (Guerrero-Rascado et al., 2016).

The other two passive remote sensing observations of aerosol properties are ground-based and satellite-borne remote sensing observations. Ground-based remote sensing observations supply aerosol loading data (such as AOD), by measuring the attenuation of radiation from the top of the atmosphere to the surface (Holben et al., 1998). This type of observations mainly uses weather-resistant automatic sun and sky scanning spectral radiometers to retrieve optical and microphysical aerosol properties (Che et al., 2014). The Aerosol Robotic Network (AERONET) is a popular global network composed of NASA and multiple international partners that provides high-quality and high-frequency aerosol optical and microphysical properties under various geographical and environmental conditions (Holben et al., 1998; Dubovik et al., 2000). The AERONET observations are extensively used to validate of satellite remote sensing observations and model simulations, as well as climatology study (Dubovik et al., 2002b). There are many regional networks of sun photometers, such as the Maritime Aerosol Network (MAN), which use a handheld sun photometer to collect data on the ocean and is merged into AERONET (Smirnov et al., 2009), the China Aerosol Robot Sun Photometer Network(CARSNET) (Che et al., 2009), the Canadian sub-network of AERONET (AEROCAN) (Bokoye et al., 2001), Aerosol characterization via Sun photometry: Australian Network (AeroSpan) (Mukkavilli et al., 2019), and the sky radiometer network (SKYNET) in Asia and Europe (Kim et al., 2004; Nakajima et al., 2020). Another very valuable global network is the NOAA/ESRL Federated Aerosol Network (FAN), which uses integrated nephelometers distinct from sun photometers, mainly located in areas with less human activity impact, providing regionally representative aerosol properties over 30 sites (Andrews et al., 2019).

Satellite remote-sensing is a space-based method that can provide aerosol properties worldwide. With the development of satellite remote sensing technology since 1970, aerosol distributions can be extracted with the advantage of sufficient real-time and global coverage from multiple satellite sensors (Kaufman & Boucher, 2002; Anderson et al., 2005). The Advanced Very High Resolution Radiometer (AVHRR) was the earliest sensor used for retrieving AOD over ocean (Nagaraja Rao et al., 1989). The Moderate Resolution Imaging Spectroradiometer (MODIS), on board the Terra (launched in 1999) and Aqua (launched in 2002) satellites is a popular sensor with 36 channels, which have been used for AOD retrieval over both ocean and land based on the Dark Target and the Deep Blue algorithms (Remer et al., 2005; Levy et al., 2013). The latest MODIS AOD data version is the Collection 6.1, which provides global AOD over 20 years (Wei et al., 2019a). There are also many other satellite sensors that can be used to retrieve AOD, such as the Polarization and

Directionality of the Earth's Reflectances (POLDER) during 1996-1997, 2003 and 2004-2013 (Deuzé et al., 2000), Sea-viewing Wide Field-of-view Sensor (SeaWIFS) during 1997-2007 (O'Reilly et al., 1998), the Multi-angle Imaging Spectroradiometer (MISR) on Terra since 1999 (Diner et al., 1998). The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) has also derived aerosols in the vertical direction since 2006 (Winker et al., 2009).

These measurements provide important data for studying the global and regional spatiotemporal variabilities and climate effect of aerosols. However, in-situ and ground-based remote sensing observations only provide aerosol properties with low spatial coverage. There were only 1126 ground stations worldwide in 2002 and even fewer sites were available for climate analysis (Holben et al., 1998; Chu et al., 2002), which limited aerosol climate research by spatial coverage (Bright & Gueymard, 2019). Satellite remote sensing overcomes the limitations of spatial coverage. The AVHRR has been used to retrieve AOD since 1980, but it is limited by a few channel number, low spatial resolution, and insufficient validation through ground-based observations before 2000 (Hsu et al., 2017). Many studies have only investigated the trends and distributions of aerosols after 2000 (Bösenberg & Matthias, 2003; Winker et al., 2013; Xia et al., 2016; Tian et al., 2023), because of the lack of long-term and global cover AOD products, which is the bottleneck for aerosol climate change detection and attributions.

To overcome these limitations and enrich aerosol data, alternative observation data could be utilized to derive AOD. For example, some studies used solar radiation data to infer AOD and analyze the characteristics of AOD in different regions (King et al., 1978; Vasilyev et al., 1995; Marenco et al., 1997; Qiu, 1997). There are also some studies deriving AOD based on empirical relationship between particle concentration and AOD (Xie et al., 2015; Li, 2020). These methods partially mitigate the scarcity of AOD data in spatial coverage, but it is also important to acknowledge the inherent limitation of long temporal coverage. Another more suitable alternative is atmospheric horizontal visibility, because it has the advantages of the long-term records with a large number of stations worldwide.

Atmospheric visibility is a physical quantity that describes the transparency of the atmosphere through manual and automatic observations. The automatic observations of visibility usually measure atmospheric extinction (scattering coefficient and transmissivity), including particle matter, water vapor, and gas molecules (Wang et al., 2009; Zhang et al., 2020), which makes it a favorable choice for inferring AOD. Koschmieder (1924) first proposed the relationship between the meteorological optical range and the total optical depth. Elterman (1970) futher established a formula between AOD and visibility by assuming an exponential decrease in aerosol concentration with altitude, considering the extinction of molecules and ozone to analyze air pollution, which called the Elterman model. Qiu and Lin (2001) corrected the Elterman model by considering the influence of water vapor and used two water vapor pressure correction coefficients to retrieve AOD of 16 stations in China in 1990. Lin et al. (2014) retrieved the AOD in eastern China in 2006 using visibility and aerosol vertical profiles provided by GEOS-Chem. Wu et al. (2014) and Zhang et al. (2017) parameterized the constants in the Elterman model and use satellite retrieved AOD to solve the parameters in the models at different stations, to retrive the long-term AOD in China. Zhang et al. (2020) reviewed the methods of visibility retrieval of AOD, indicating that visibility-based retrieval of AOD can compensate for the shortcomings of long-term aerosol observation data. Simultaneously, various parameters, such as station altitude, consistency of visibility data, water vapor and aerosol vertical profiles (scale height), were discussed with modified suggestions proposed. These studies have enriched AOD data regionally. Due to the similar spatial distribution of the extinction coefficient and AOD, and the proportional relationship between the reciprocal of visibility and the extinction coefficient, Wang et al. (2009) analyzed the trend of AOD using visibility-based retrivals from 1973 to 2007 over land. These studies have enriched aerosol data in some extent. At present, there are very few studies on global visibility-retrieved AOD and to analyze climatology of aerosols.

The two physical quantities of visibility and AOD have both connections and differences, making it challenging to retrieve AOD from visibility. Visibility represents the maximum horizontal visible distance near the surface, while AOD represents the total vertical attenuation of solar radiation by aerosols. The visibility of automatic observation is dependent on the local horizontal atmosphereic extinction (NOAA et al., 1998). Visibility has not a simple linear relationship with meteorological factors, such as humidity and wind speed. The vertical structure of aerosols is the greatest challenge to obtain, as it is not a simple hypothetical curve in complex terrain and circulation conditions (Zhang et al., 2020). These limitations make it more complex to derive AOD over global land. However, previous studies have shown that surface observation data can establish a link with AOD, particularly at the regional scale. Machine learning methods can effectively address complex nonlinear relationships between variables and have been widely applied in remote sensing and climate research fields. Li et al. (2021) used the random forest method to predict $PM_{2.5}$ in Iraq and Kuwait based on satellite AOD during 2001-2018. Kang et al. (2022) applied LightGBM and random forest to estimate AOD over East Asia, and the results showed a consistency with AERONET. Dong et al. (2023) derived aerosol single scattering albedo from visibility and satellite AOD over 1000 global stations. Hu et al. (2019) used a deep learning method to retrieve horizontal visibility from MODIS AOD. These studies have confirmed the ability of machine learning to effectively solve complex relationships among variables. Thus, it is feasible to derive AOD from atmospheric visibility by using the machine learning method.

In this study, we propose a machine learning method to derive AOD, where satellite AOD is the target value, and surface visibility and other related meteorological variables are the predictors. We explain the robustness of the model, validate the accuracy of the model's predictions using ground-based AOD and other observations, and analyze the climatology of AOD across land and regions. Two datasets of long-term high-resolution AOD are generated. The second part of this paper introduces the data and method. The third part is the evaluation and validation of the visibility-derived AOD, and the distribution and trends are discussed at global and regional scales. The fourth part presents the conclusions. This study is dedicated to supporting the research of aerosols in climate change detection and attribution.

2 Data and method

2.1 Study area

The study area is global land region. A total of 5032 land stations of the Automated Surface Observing System (ASOS), which is a joint surface weather observing network of the National Weather Service (NWS), the Federal Aviation Administration (FAA), and the Department of Defense (DOD) (NOAA et al., 1998). A total of 573 stations of AERONET are selected in this study

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and shown in Figure 1 (a). 12 typical regions are selected for analysis, including Eastern Europe, Western Europe, Western North America, Eastern North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and Middle East. The time range in the study is from 1980 to 2021, during which the records of ASOS stations are sufficient with a uniform spatial distribution. As shown n Figure 1 (b), the daily records have exceeded 1500 stations, and monthly and annual records have exceeded 2000 during 1980-1990. After 2000, monthly records have reached 3000, which is the foundation of gridding AOD.

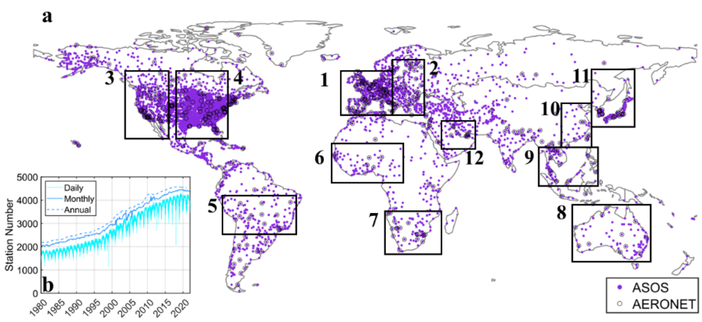

Figure 1 Study area (a) and the station number (b) with daily, monthly, and annual records in the Automated Surface Observing System (ASOS). The number of ASOS stations (filled circles) is 5032. The number of AERONET stations (empty circles) is 573. The box regions of labelled with number 1-12 are Eastern Europe, Western Europe, Western North America, Eastern North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and Middle East.

2.2 Ground-based observations

Ground-based hourly observation data from 1980 to 2021 are collected at 5032 globally distributed stations (Figure 1) from the ASOS program. From the 1960s to the 1970s, the Automated Meteorological Observing System (AMOS) and Remote Automated Weather Observing System (RAMOS) only reported objective elements, such as temperature, dew point temperature, wind (speed and direction), and pressure. With technological advancements, the ASOS was deployed and utilized in the 1980s. The automatic surface observations reduced errors associated with human involvement in data acquisition, processing, and transmission. Effective quality control methods are employed to ensure the quality of ASOS products. ASOS provided hourly and even minutely ground automatic observations, primarily for airports (NOAA et al., 1998; Dover et al., 2002).

Atmospheric visibility from ASOS is measured by the forward-scatter visibility sensor with a wavelength of 550 nm. The scattering angle of the sensor ranges from 0 to 45 degree, the sampling volume is 0.75 cubic feet and the response time is 20 seconds. The sensor provides 1-minute average visibility with the day or night indication. Hourly visibility is calculated based on the harmonic average of minutely visibility. Experiments have found that harmonic average visibility can better detect the development of some weather phenomena than arithmetic average visibility (NOAA et al., 1998). The sensor-measured visibility has a strong agreement with the human-observed during haze and homogeneous weather over a large area, even during periods when weather conditions are

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

quite variable (NOAA et al., 1998). The same algorithm is used to calculate the daily, monthly, seasonally and yearly average visibility.

$$\boldsymbol{V = n/(\frac{1}{V_1} + \frac{1}{V_2} + \cdots + \frac{1}{V_n})} \qquad \textbf{Eq. 1}$$

where V is the harmonic mean, n = 24 for the daily mean, and V_1, V_2,... V_n are the individual hourly values.

Visibility in METAR is reported in statute miles (SM). The reportable increments are: M1/4SM, 1/4SM, 1/2SM, 3/4SM, 1SM, 1 1/4SM, 1 1/2SM,1 3/4SM, 2SM, 2 1/2SM, 3SM, 4SM, 5SM, 6SM, 7SM, 8SM, 9SM and 10SM. It is noted that visibility between zero and 1/4 statute mile is reported as M1/4SM8. Visibility values of exactly halfway between reportable values are rounded down. Visibility values of 10 miles or greater are reported as10SM (NOAA et al., 1998).

In addition to hourly visibility (VIS), we also selected other automatically observed variables closely related to aerosol properties in this study. Because relative humidity influences the size and hygroscopic growth rate of particle matter, and wind speed and pressure significantly impact the transport and deposition of aerosols, relative humidity (RH), dew point temperature (DT), temperature (TMP), wind speed (WS) and sea-level pressure (SLP) are adopted. Additionally, sky conditions (cloud amount) and hourly precipitation are also selected to remove the influence of extensive cloud cover and precipitation when deriving AOD.

We processed the data as follows. The records with missing values were eliminated (Husar et al., 2000). When over 80% overcast or fog, the records of sky conditions were eliminated, though such situations occur less than 1% of the time over land (Remer et al., 2008). The records with 1-hour precipitation greater than 0.1 mm were eliminated. The records with RH greater than or equal to 90% were eliminated. We calculate the temperature dew point difference (dT). When RH is between 30% and 90%, visibility is converted to dry visibility (Yang et al., 2021c).

$$\boldsymbol{VISD = VIS/(0.26 + 0.4285 * log(100 - RH))} \qquad \textbf{Eq. 2}$$

where VISD is the dry visibility.

Daily average of variables are calculated by at least 3 hourly records with a harmonic mean for visibility (NOAA et al., 1998) and an arithmetic mean for the other variables.

2.3 Boundary layer height

The hourly boundary layer height (BLH) from 1980 to 2021 is available from the Fifth Generation European Medium-Range Weather Forecast Center (ERA5) with a resolution of 0.25° x 0.25° (https://cds.climate.copernicus.eu), which is the successor of ERA-Interim and has undergone various improvements(Hersbach et al., 2020). The atmospheric boundary layer is the layer closest to the Earth's surface and exhibits complex turbulence activities, and its height undergoes significant diurnal variation. The effects of the boundary layer on aerosols are mainly manifested in vertical distribution, concentration changes, transport, and deposition (Ackerman et al., 1995). The characteristics and variations in the boundary layer play a crucial role in regulating and adjusting the distribution of atmospheric aerosols. The boundary layer height serves as an approximate measure of the scale height for aerosols (Zhang et al., 2020). The BLH of ERA5 is considered to be

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

the more promising dataset compared to the MERRA-2, JRA-55, and NCEP-2 datasets (Guo et al., 2021). The BLH data is temporally and spatially matched with the ASOS stations. Because the inverse of visibility is proportional to the extinction coefficient and positively related to AOD (Wang et al., 2009) and the atmospheric aerosols are largely distributed in the boundary layer (Zhang et al., 2020), three variables (VISI, VISDI, VISDIB) are increased, shown in Eq. 3 :

$$\boldsymbol{VISI} = \frac{\mathbf{1}}{\boldsymbol{VIS}}, \boldsymbol{VISDI} = \frac{\mathbf{1}}{\boldsymbol{VISD}}, \boldsymbol{VISDIB} = \boldsymbol{VISDI} * \boldsymbol{BLH} \qquad \textbf{Eq. 3}$$

Thus, the Predictors (Figure 2) is composed of 11 variables: TMP, Td, dT, RH, SLP, WS, VIS, BLH, VISI, VISDI, and VISDIB.

2.4 MODIS AOD Products

Satellite daily AOD is available from the Moderate Resolution Imaging Spectroradiometer (MODIS) Level 3 Collection 6.1 AOD products of the Aqua (MYD09CMA) satellite from 2002 to 2021 and Terra (MOD09CMA) satellite from 2000 to 2021 with a spatial resolution of 0.05° x 0.05° at a wavelength of 550 nm (https://ladsweb.modaps.eosdis.nasa.gov). MOD/MYD09 has a higher spatial resolution than MOD/MYD08 (1° x 1°), which may result in a greater difference in AOD values and reduce the proximity ratio to match the same AOD value. Terra (passing approximately 10:30 am local time) and Aqua (passing approximately 1:30 pm local time) were successfully launched in December 1999 and May 2002, respectively. MODIS, carried on the Terra and Aqua satellites is a crucial instrument in the NASA Earth Observing System program, which is designed to observe global biophysical processes (Salomonson et al., 1987). The 2,330 km-wide swath of the orbit scan can cover the entire globe every one to two days. MODIS has 36 channels and more spectral channels than previous satellite sensors (such as AVHRR). The spectral range from 0.41 to 15 μm representing three spatial resolutions: 250 m (2 channels), 500 m (5 channels), and 1 km (29 channels). The aerosol retrieval uses seven of these channels (0.47–2.13μm) to retrieve aerosol characteristics and uses additional wavelengths in other parts of the spectrum to identify clouds and river sediments. Therefore, it has the ability to characterize the spatial and temporal characteristics of the global aerosol field. The MODIS aerosol product actually takes use of different algorithms for deriving aerosols over land and ocean. The Dark Target (DT) algorithm is applied to densely vegetated areas because the surface reflectance over dark-target areas was lower in the visible channels and had nearly fixed ratios with the surface reflectance in the shortwave and infrared channels (Levy et al., 2007; Levy et al., 2013). The Deep Blue (DB) algorithm was originally applied to bright land surfaces (such as deserts), and later extended to cover all cloud-free and snow-free land surfaces (Hsu et al., 2006; Hsu et al., 2013). MODIS Collection 6.1 aerosol product was released in 2017, incorporating significant improvements in radiometric calibration and aerosol retrieval algorithms. The expected errors are ± (0.05 ± 15%) for the DT retrievals over land. Higher spatial coverage is observed in August and September, reaching 86-88%. During December and January, due to the presence of permanent ice and snow cover in high-latitude regions of the Northern Hemisphere, the spatial coverage is 78-80%. Thus, challenges remain in retrieving AOD values in high-latitude regions (Wei et al., 2019a). However, visibility observations are available in high-latitude regions, thereby partially addressing the lack in these regions. In this study, the Terra and Aqua MODIS AOD are temporally and spatially matched with the ASOS stations. Aqua MODIS AOD is used as the Target, when training the model, and Terra MODIS AOD is used in the evaluation and validation of the model results, as shown in the flowchart (Figure 2).

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

2.5 Ground-based AOD

Ground-based daily AOD data are available from the Aerosol Robotic Network (AERONET) Version 3.0 Level 2.0 product at 573 stations (Figure 1), which can be downloaded from https://aeronet.gsfc.nasa.gov. The AERONET program is a federation of ground-based remote sensing aerosol networks established by NASA and PHOTONS, including many subnetworks (such as AeroSpan, AEROCAN, NEON, and CARSNET). The sun photometer (CE-318) measures spectral sun and sky irradiance in the 340-1020 nm spectral range. When the aerosol loading is low, the error is significant. When the AOD at 440 nm wavelength is less than 0.2, the error is 0.01, which is equivalent to the error of the absorption band in the total optical depth (Dubovik et al., 2002a). The total uncertainty in AOD under cloud-free conditions is less than ±0.01 for wavelength more than 440 nm, and ±0.02 for wavelength less than 440 nm (Holben et al., 1998). AERONET has three levels of AOD products: Level 1.0 (unscreened), Level 1.5 (cloud screened), and Level 2.0 (cloud screened and quality assured). Compared to Version 2, the Version 3 Level 2.0 database has undergone further cloud screening and quality assurance, which is generated based on Level 1.5 data with pre- and post-calibration and temperature adjustment and is recommended for formal scientific research (Giles et al., 2019). AERONET provides AOD products at wavelengths of 440, 675, 870, and 1020 nm. To match the MODIS AOD, the AOD measured from AERONET needs to be converted to the AOD at 550 nm using the Ångström equations (Fan & Sun, 2023).

$$\boldsymbol{\tau_\alpha(\lambda) = \beta\lambda^{-\alpha}} \qquad \textbf{Eq. 4}$$

$$\boldsymbol{\alpha = -\frac{ln\left(^{\tau(\lambda_1)}/_{\tau(\lambda_2)}\right)}{ln\left(^{\tau(\lambda_1)}/_{\tau(\lambda_2)}\right)}} \qquad \textbf{Eq. 5}$$

$$\boldsymbol{\beta = \frac{\tau(\lambda_1)}{\lambda_1{}^{-\alpha}}} \qquad \textbf{Eq. 6}$$

where $\tau_\alpha(\lambda)$ is the AOD at a wavelength of 550 nm, β is the turbidity coefficient, α is the wavelength index, and λ_1 and λ_2 are the wavelengths of the two selected channels in AERONET.

2.6 Decision Tree Regression

2.6.1 Feature selection

Although a multidimensional dataset can provide as much potential information as possible for AOD, irrelevant and redundant variables can also introduce significant noise in the model and reduce the model's accuracy and stability (Kang et al., 2021; Dong et al., 2023). Therefore, the F-test is used to search for the optimal feature subset in the Predictor, aiming to eliminate irrelevant or redundant features and select truly relevant features, which helps to simplify the model's input and improve the model's prediction ability (Dhanya et al., 2020). The F-test is a statistical test that gives an f-score(=-log(p), p represents the degree to which the null hypothesis is not rejected) by calculating the ratio of variances. In this study, we calculate the ratio of variance between the Predictors and Target, and the features are ranked based on higher values of the f-score. A greater value of f-score means that the distances between Predictors and Target are less and the relationship is closer, thus, the feature is more important. We set p=0.05. When the score is less than -log (0.05), the variable in the Predictors is not considered.

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

2.6.2 Data balance

Under good weather conditions (such as clear weather), the observed AOD values are concentrated around the average value. Under bad weather conditions (such as heavy haze, wildfires, sandstorms), the value values will vary significantly compared to the good weather conditions, and the frequency of large AOD value is low. When the AOD time series is observed under both good and bad weather conditions, the minority class is large AOD value. This is a phenomenon of data imbalance. When dealing with imbalanced datasets, because of the tendency of machine learning algorithms to perform better on the majority class and overlook the minority class, the model can be underfit (Chuang & Huang, 2023). Data augmentation techniques are commonly employed to address the issue in imbalance data, which applies a series of transformations or expansions to generate new training data, thereby increasing the diversity and quantity of the training data. The Adaptive Synthetic Sampling (ADASYN) is a data augmentation technique specifically designed to address data imbalance problem (He et al., 2008; Mitra et al., 2023). It is an extension of the Synthetic Minority Over-sampling Technique (SMOTE) algorithm (Fernández et al., 2018). The goal of ADASYN is to generate synthetic sample data for the minority class to increase its representation in the dataset. ADASYN, which adaptively adjusts the generation ratio of synthetic samples based on the density distribution of sample data, improves the dataset balance and enhances the performance of machine learning models in dealing with imbalanced data.

2.6.3 Decision Tree Regression Model

The decision tree is a machine learning algorithm based on a tree-like structure used to solve classification and regression problems. We adopt the CART algorithm to construct a regression tree by analyzing the mapping relationship between object attributes (Predictors) and object values (Target). The internal nodes have binary tree structures with feature values of "yes" and "no". In addition, each leaf node represents a specific output for a feature space. The advantages of the regression tree include the ability to handle continuous features and the ease of understanding the generated tree structure (Teixeira, 2004; Steinberg & Colla, 2009). Before training the tree model, the variables (Input) are normalized to improve model performance, and after prediction, the results are obtained by denormalization. The 10-fold cross-validation method is employed to improve the generalization ability of the model (Browne, 2000).

The core problems of the regression tree need to solve are to find the optimal split variable and optimal split point. The optimal split point of Predictors is determined by the minimum MSE, which in turn determines the optimal tree structure. We set $\mathrm{Y} = [y_1, y_2, \dots, y_N]$ as the Target. We set $\mathrm{X} = [x_1, x_2, \dots, x_N]$ as the Predictors, $x_i = (x_i^1, x_i^2, \dots x_i^n)$, $\mathrm{i} = 1,2,3 \dots, \mathrm{N}$, where n is the feature number, and N is the length of sample. We set a training dataset as $\mathrm{D} = [(x_1, y_1), (x_2, y_2), \dots, (x_N, y_N)]$.

A regression tree corresponds to a split in the feature space and the output values on the split domains. Assuming that the input space has been divided into M domains $[R_1, R_2, \dots, R_M]$ and there is a fixed output value on each R_M domain, the regression tree model can be represented as follows:

$$\boldsymbol{f(x) = \sum_{m=1}^{M} c_m I(x \in R_M)\,, m = 1, 2, \dots, M} \qquad \textbf{Eq. 7}$$

where I is the indicator function (Eq. 8).

$$\boldsymbol{I = \begin{cases} 1, x \in R_m \\ 0, x \notin R_m \end{cases}} \qquad \textbf{Eq. 8}$$

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

When the partition of the input space is determined, the square error can be used to represent the prediction error of the regression tree for the training data, and the minimizing square error is used to solve the optimal output value on each domain. The optimal value ($\widehat{c_m}$) on a domain is the mean of the outputs corresponding to all input, namely:

$$\widehat{c_m} = ave(y_i|x_i \in R_m) \qquad \textbf{Eq. 9}$$

A heuristic method is used to split the feature space in CART. After each split, all values of all features in the current set are examined individually, and the optimal one is selected as the split point based on the principle of minimum sum of the square errors. The specific step is described as follows: for the training dataset D, we recursively divide each region into two sub domains and calculate the output values of each sub domain; then, construct a binary decision tree. For example, split variable is x^j and split point is s. Then, in the domain $R_1(j,s) = [x|x^j \leq s]$ and domain $R_2(j,s) = [x|x^j > s]$, we can solve the loss function $L(j,s)$ to find the optimal j and s.

$$L(j,s) = \sum_{x_i \in R_1(j,s)} (y_i - c_1)^2 + \sum_{x_i \in R_2(j,s)} (y_i - c_2)^2 \qquad \textbf{Eq. 10}$$

When $L(j,s)$ is the smallest, x^j is the optimal split variable and s is the optimal split point for the x^j.

$$\underbrace{min}_{j,s}\left[\underbrace{min}_{c_1}\sum_{x_i \in R_1(j,s)} (y_i - c_1)^2 + \underbrace{min}_{c_2}\sum_{x_i \in R_2(j,s)} (y_i - c_2)^2\right] \qquad \textbf{Eq. 11}$$

We use the optimal split variable x^j and the optimal split point s to split the feature space and calculate the corresponding output value.

$$\widehat{c_1} = ave(y_i|x_i \in R_1(j,s)),\ \widehat{c_2} = ave(y_i|x_i \in R_2(j,s)) \qquad \textbf{Eq. 12}$$

We traverse all input variables to find the optimal split variable x^j, forming a pair (j,s). Divide the input space into two regions accordingly. Next, repeat the above process for each region until the stop condition is met. The regression tree is generated.

Therefore, the regression tree model $f(x)$ can be represented as follows:

$$f(x) = \sum_{m=1}^{M} \widehat{c_m} I(x \in R_M), m = 1, 2, \ldots, M \qquad \textbf{Eq. 13}$$

2.7 Gridding method

Kriging is a regression algorithm to model and predict (interpolate) random processes/fields based on the covariance function, which is widely used in geo-statistics (Pebesma, 2004). Ordinary Kriging is the earliest and most extensively studied form of Kriging. It is a linear estimation system applicable to any intrinsic stationary random field that satisfies the assumption of isotropy. The two key parameters of Ordinary Kriging are the semi-variogram function and the weight factors (Goovaerts, 2000). It has been widely applied in fields, such as climatology, environmental science, and agriculture (Lapen & Hayhoe, 2003; Chen et al., 2010), due to high accuracy, stability, and insensitivity to data shape and distribution. This study utilizes area-weighted ordinary kriging algorithm to estimate the unknown values of AOD at specific locations to generate gridded AOD. The longitude range is between -180° E and 180 °E, the latitude range is between -60 °N and 85 °N, and the spatial resolution is 0.5 °*0.5 °.

2.8 Evaluation metrics

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Open Access Earth System Science Data Discussions

Evaluation metrics, including Root Mean Squared Error (RMSE), Mean Absolute Error (MAE) and Pearson Correlation Coefficient (R), are used to measure the performance and accuracy of the model and gridded results.

$$\boldsymbol{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(y_i - \hat{y}_i)^2} \quad \textbf{Eq. 14}$$

$$\boldsymbol{MAE} = \frac{1}{n}\sum_{i=1}^{n}|y_i - \hat{y}_i| \quad \textbf{Eq. 15}$$

$$\boldsymbol{R} = \frac{\sum_{i=1}^{n}(y_i-\bar{y})(\hat{y}_i-\bar{\hat{y}})}{sqrt(\sum_{i=1}^{n}(y_i-\bar{y})^2\sum_{i=1}^{n}(\hat{y}_i-\bar{\hat{y}})^2)} \quad \textbf{Eq. 16}$$

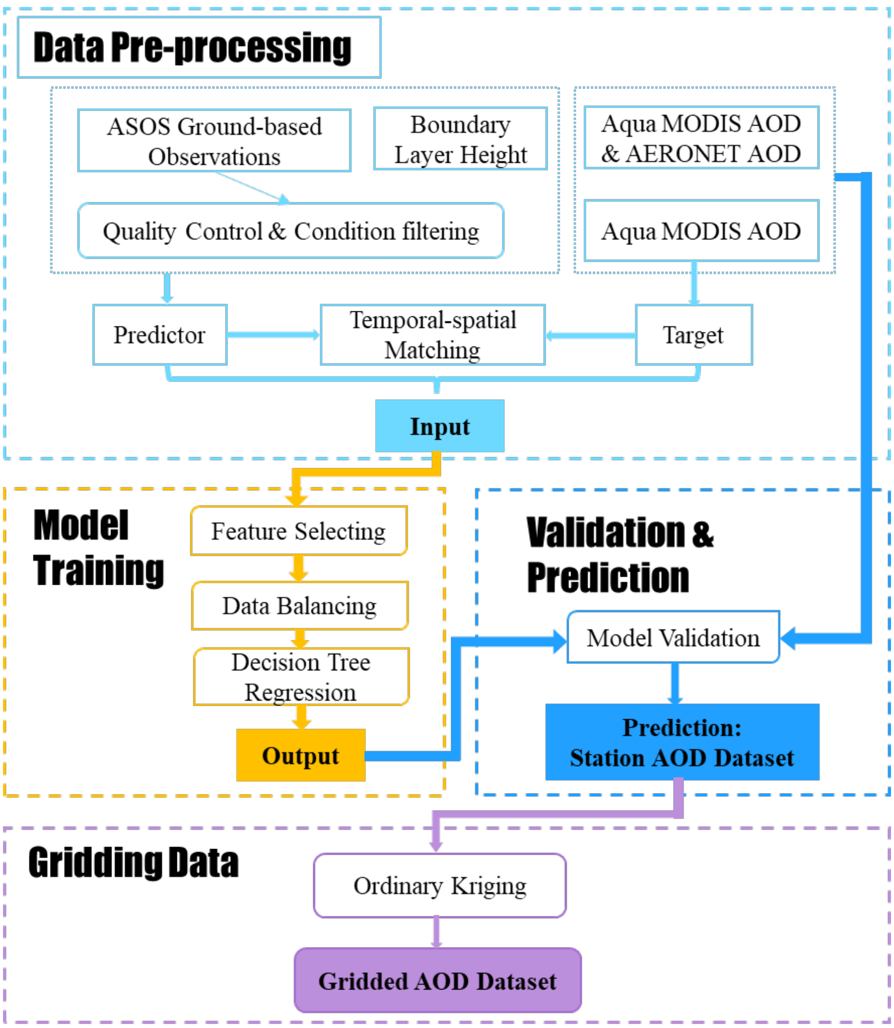

Figure 2 Flowchart for deriving aerosol optical depth (AOD).

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

2.9 Workflow

Figure 2 is the summarized flowchart and provides an overview of the structure of this study, which involves four main parts: (1) data preprocessing, (2) model training, (3) validation and prediction, and (4) data gridding.

3 Results and discussion

3.1 Examination of the model performance

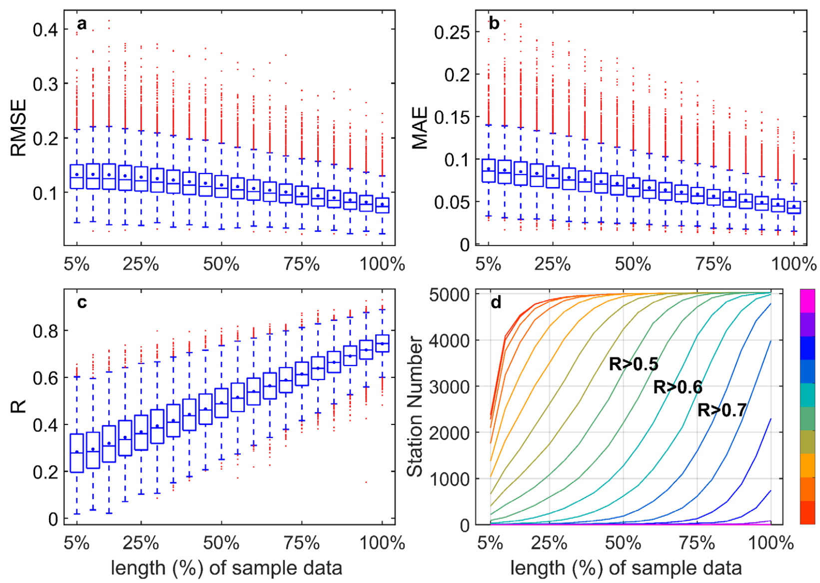

Figure 3 Boxplots of root mean squared error (RMSE) (a), mean absolute error (MAE) (b), and correlation coefficient (R) (c) between predicted values and target using different lengths of sample data (5% interval) as the training dataset, and the correlation coefficient curve (d) of the station number in the different lengths of sample data.

We build the models using different lengths of sample data (5% to 100%, with a 5% interval) by random allocation without overlap and evaluate the predictive performance of each model. Figure 3 depicts RMSE(a), MAE(b), and R (c) between the predicted values and target based on the training data of 5% to 100% sample data at a station. As the volume of the training data increases, the RMSE and MAE decrease, and the correlation coefficient increases. Compared to 5% of the sample data, the result of 100% sample data shows a decrease in RMSE by 41.1%, a decrease in MAE by 50.1%, and an increase in R by 162.3%. The relationship between the length of sample data and the model's performance is positive for each station. Figure 3 (d) shows that R of approximately 70% stations is greater than 0.5 at 50% of the sample data, while at 75%, the R of approximately 80% of stations is greater than 0.6. When 100% of the sample data is used as sample data, the R of approximately 80% of stations is greater than 0.75, and the R of about 97% is greater than 0.7. This finding indicates that the predictive capability and

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

robustness of the model increase as the amount of training data increases. It may be attributed to the model's ability to capture more complex patterns and relationships among the input by multi-year data.

3.2 Evaluation of model errors

The more sample data input, the better the model performs. Therefore, 100% of the sample data were used as training data. Figure 4 shows the spatial distribution (a-c) and frequency and cumulative frequency (d-e) of RMSE, MAE, and R of all stations. The mean values of RMSE, MAE, and R are 0.078, 0.044, and 0.75, respectively. The RMSE of 93% stations is less than 0.11, the MAE of 91% is less than 0.06, and the R of 88% is greater than 0.7. The R values in Africa, Asia, Europe, North America, Oceania, and South America are 0.763, 0.758, 0.736, 0.750, 0.759, and 0.738, respectively. Although the RMSE and MAE of a few stations are high in America and Asia, the R is still high (>0.6). Therefore, the results of the model's errors demonstrate that the model performs well on almost all stations.

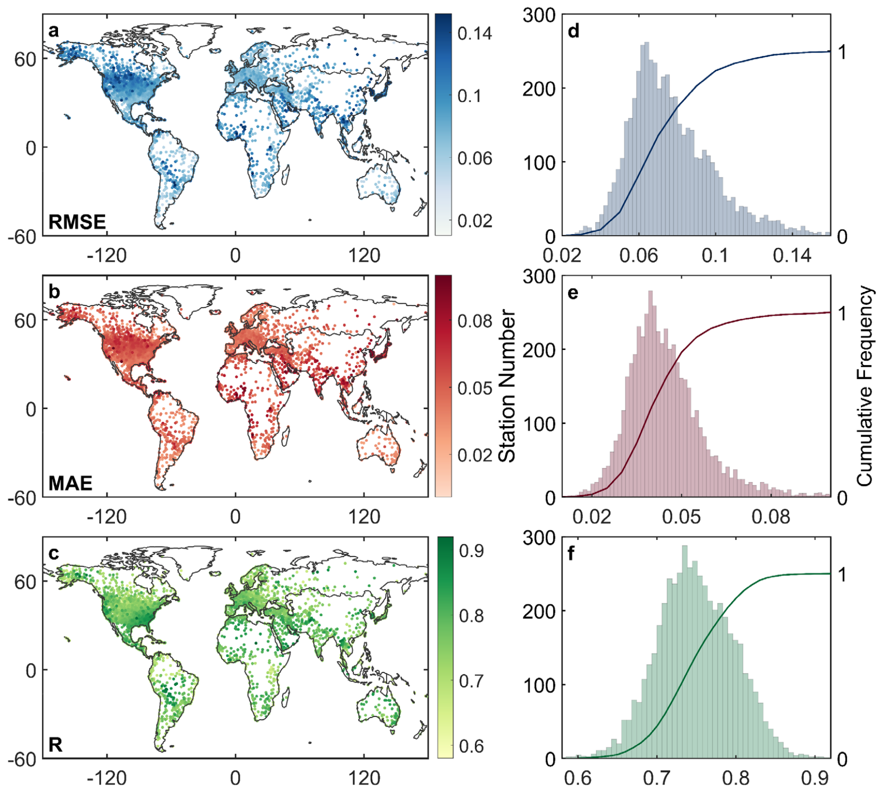

Figure 4 Spatial distribution (a-c) of root mean squared error (RMSE), mean absolute error (MAE), and correlation coefficient(R) between the model's result and target with 100% sample data. Station number (bar) and cumulative frequency (curve) (d-e) of RMSE, MAE, and R.

3.3 Validation of derived AOD against MODIS and AERONET AOD

First, the relationship among daily MODIS and AERONET AOD is evaluated. Figure 5 presents the scatter density plots (the left column) and bias probability distribution (the right column) among daily

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Aqua, Terra and AERONET AOD. The R, RMSE, and MAE of 536,998 data couples between Aqua AOD and AERONET AOD are 0.612, 0.1, and 0.093, respectively. Then, 86.8% of the data have a bias within ±0.093. The R, RMSE, and MAE of 551,462 data couples between Terra AOD and AERONET AOD are 0.602, 0.103, and 0.095, respectively. Then 86% of the data have a bias within ±0.095. The R, RMSE, and MAE of 1,896,870 data couples between Aqua AOD and Terra AOD are 0.712, 0.067, and 0.065, respectively, and the bias is within ±0.065 for 92% of the data. On the global scale, the AOD retrieved by satellites may be overestimated at low AOD levels and underestimated at high AOD levels compared to AERONET AOD. Approximately 86% of the bias values are less than the MAEs. Terra and Aqua have good consistency, although one is for morning transit and the other is for afternoon transit. Finally, 92% of the data bias are less than the MAEs. Thus, there is good consistency among them on the daily scale.

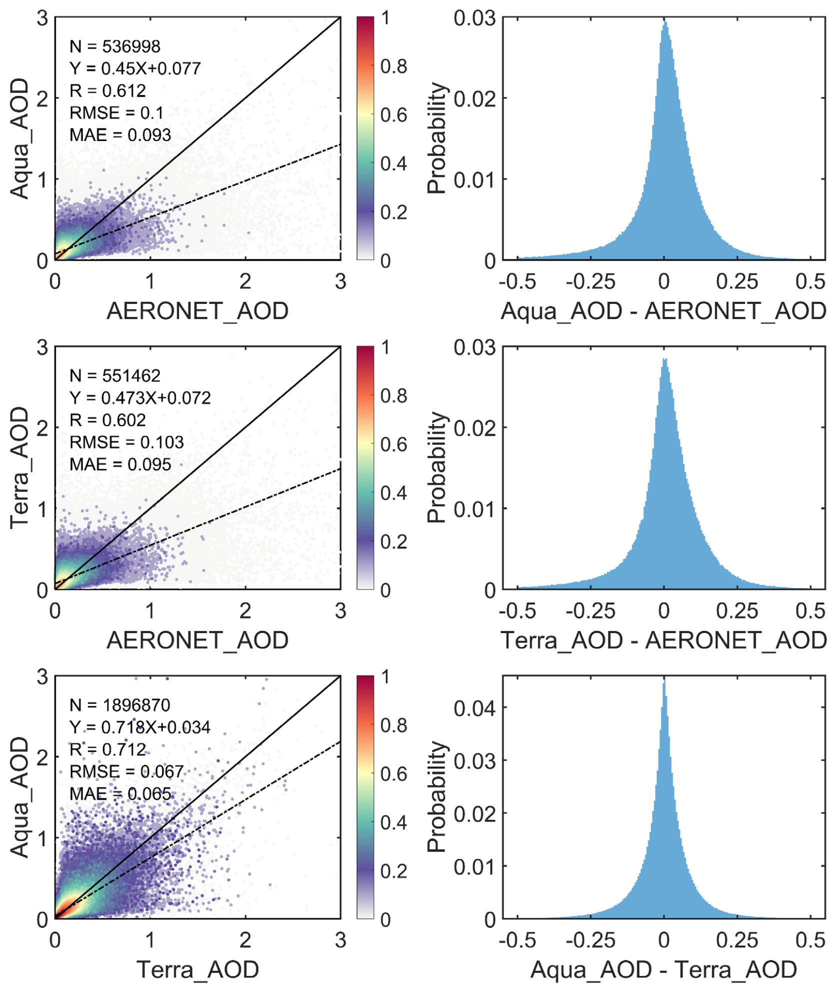

Figure 5 Scatter density plots and bias probability between Aqua AOD, Terra AOD and AERONET AOD at a daily scale. The solid black line represents the 1:1 line and the dashed black line is the

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

linear regression line.

To validate the model's predictive ability, the visibility-derived AOD (for short, VIS_AOD) is compared with other observed data for daily, monthly, and yearly scales of Aqua, Terra and AERONET AOD. Figure 6 shows the scatter density plots and probability distribution of the bias between daily VIS_AOD and Aqua AOD, Terra AOD, and AERONET AOD. The R of 15,962,757 pairs data between VIS_AOD and Aqua AOD is 0.799, higher than the R between AERONET AOD and Aqua AOD, as well as Terra AOD and Aqua AOD. The RMSE is 0.042 and the MAE is 0.044. Then, 69.7% of the data pairs have a bias within ±0.044, and 69.7% have a bias within ±0.093. The R of 17,145,578 pairs of data between VIS_AOD and Terra AOD is 0.542, the RMSE is 0.081 and the MAE is 0.078. Then, 66.8% of the data pairs have a bias within ±0.078, and 73.3% have a bias within ±0.095. The R of 334,513 data pairs between VIS_AOD and AERONET AOD is 0.514. The RMSE is 0.098 and the MAE is 0.095. Finally, 78.3% of the data pairs have a bias within ±0.095.

At the monthly and annual scales, RMSE and MAE show a significant decrease between VIS_AOD and Aqua, Terra, and AERONET AOD, and R shows a significant increase in Figure 7. The monthly RMSEs are 0.021, 0.036, and 0.048, the monthly MAEs are 0.018, 0.031, and 0.069, and the R values are 0.936, 0.808, and 0.61, respectively. The RMSE values at the annual scale are 0.012, 0.016, and 0.025, the MAE values are 0.008, 0.015, and 0.006, and the R values are 0.976, 0.0906, and 0.624, respectively. The monthly and annual R is slightly higher than those in previous studies (Wang et al., 2009; Wu et al., 2014; Zhang et al., 2017). In addition to the differences between models, it may also be related to the calculation method of daily average visibility and the consideration of boundary layer height.

Overall, the results highlighted above demonstrate a clear improvement in performance on the monthly and annual scales compared to the daily scale. However, the AERONET AOD results are slightly inferior to those of Aqua and Terra AOD, which could be caused by the representativeness of the AERONET station spatial coverage and measurement error (Holben et al., 1998). Nevertheless, the results indicate the high reliability and strong predicted capability of the model, and the visibility-derived AOD can be used for aerosol climatology.

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

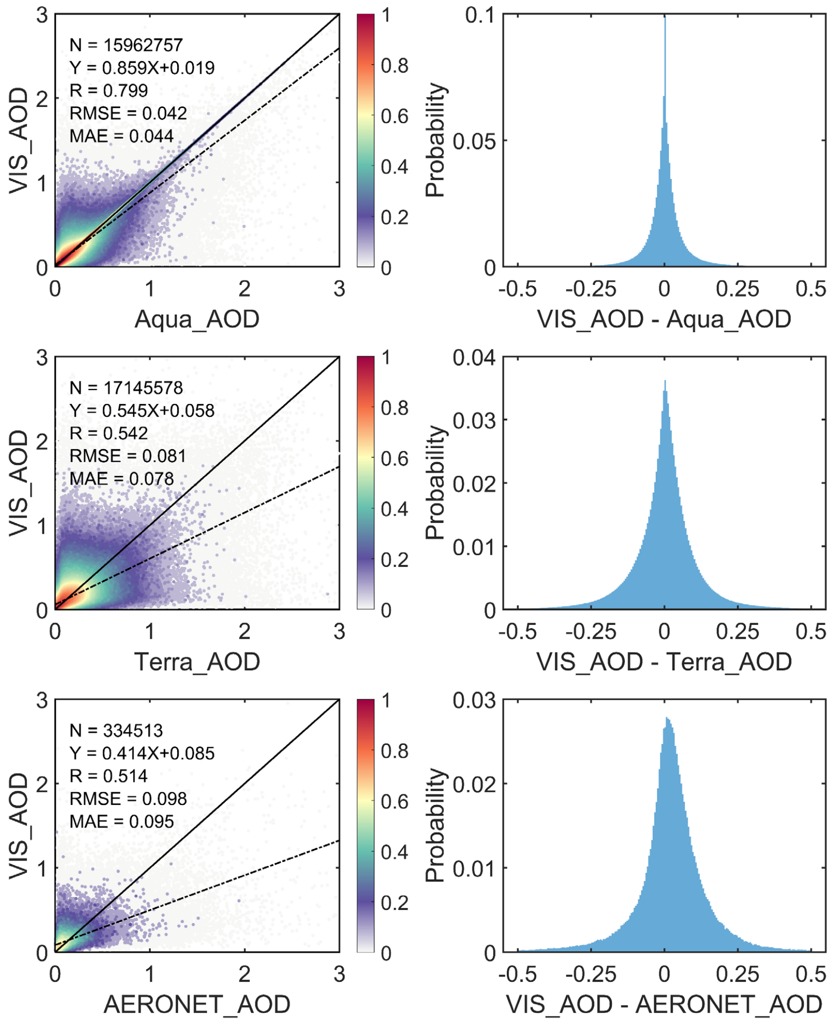

Figure 6 Scatter density plots and bias probability between predicted AOD (VIS_AOD) and Aqua MODIS AOD, Terra MODIS AOD and AERONET ground-based observations of AOD at the daily scale. The solid black line represents the 1:1 line and the dashed black line is the linear regression line.

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Open Access Earth System Science Data Discussions

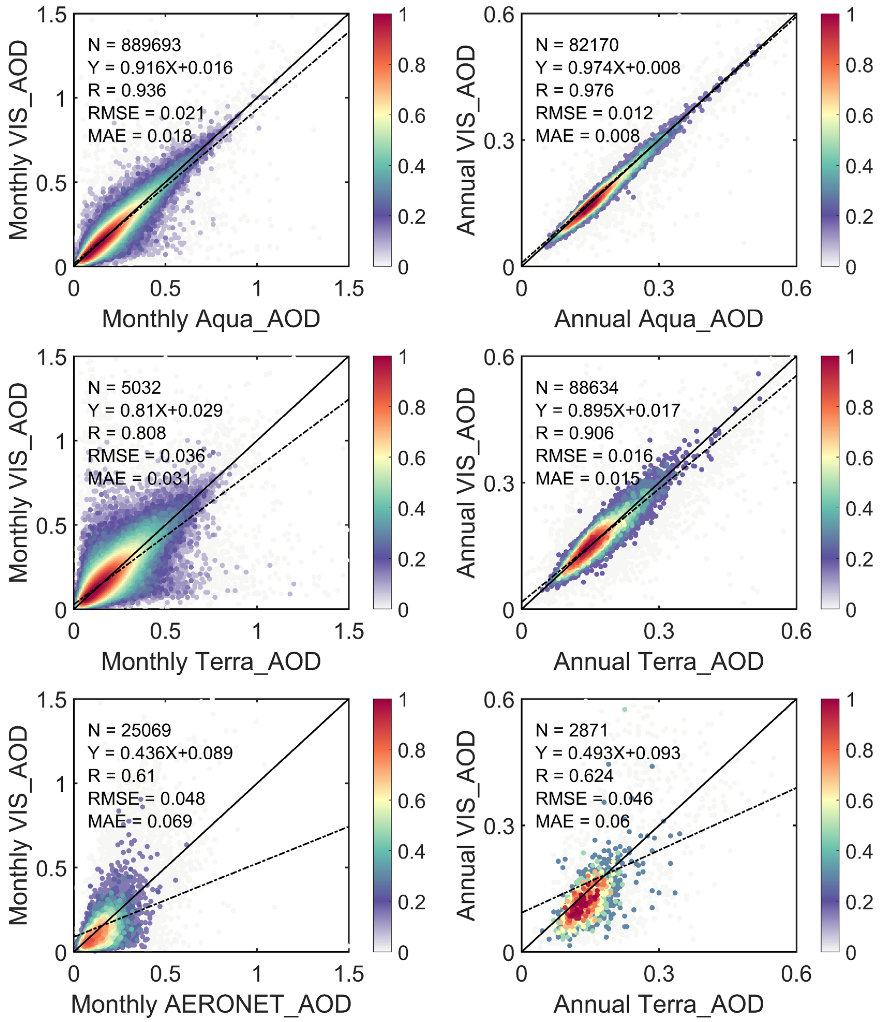

Figure 7 Scatter density plots and bias probability between VIS_AOD and Aqua MODIS AOD, Terra MODIS AOD and AERONET ground-based observations of AOD at the monthly and annual scales. The solid black line represents the 1:1 line and the dashed black line is the linear regression line.

3.4 Evaluation of gridded visibility-derived AOD

Figure 8 shows the gridded AOD based on ordinary kriging interpolation with the area-weighted method and compares the multi-year spatial, zonal, and meridional distributions of AOD with Aqua and Terra AOD from 2003 to 2021. The VIS_AOD is 0.157±0.073 over land, which is almost equal to the Aqua (0.152±0.084) and Terra (0.154±0.088) AOD values with relative biases of 3.3%, and 1.9%, respectively. In order to compare the spatial correlation, Aqua and Terra MODIS AOD are averaged to the 0.5-degree resolution. In the heatmap (Figure 9), the R of VIS_AOD and Aqua AOD

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

is 0.8, the RMSE is 0.049 with a bias of 32% compared to the mean, and the MAE is 0.008, with a bias of 5% compared to the mean. Compared to Terra AOD, the R is 0.79, and the RMSE is 0.051, with a bias of 33% compared to the mean, and the MAE is 0.005, with a bias of 3% compared to the mean. Aqua and Terra AOD are highly similar, with an R of 0.98. By comparing the zonal and meridional distributions of AOD, VIS_AOD is consistent with Aqua and Terra AOD, with the R of 0.997 and 0.99 for the zonal distribution and 0.986 and 0.9 for the meridional distribution, respectively. In the low aerosol loading region, VIS_AOD exhibits a little overestimation. Whether in meridional or zonal distribution, the peak and valley regions are basically consistent (Tian et al., 2023). Due to the limitations of satellite inversion algorithms, a bias appears on the bright surface, especially in northern North America with extensive snow cover (Levy et al., 2013). All above results suggest that the gridded AOD is highly consistent with satellite observations in spatial distribution.

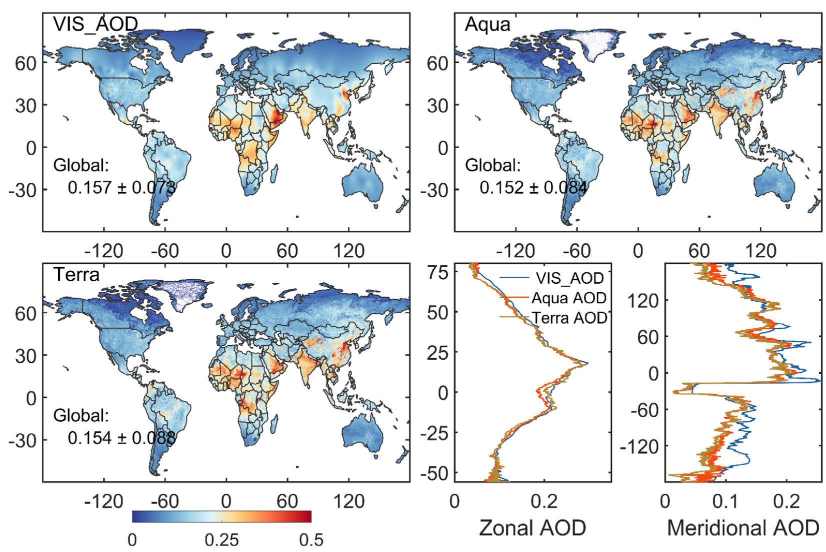

Figure 8 The spatial, zonal and meridional distributions of the multi-year mean VIS_AOD, Aqua AOD, and Terra AOD from 2003 to 2021.

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Preprint. Discussion started: 14 November 2023

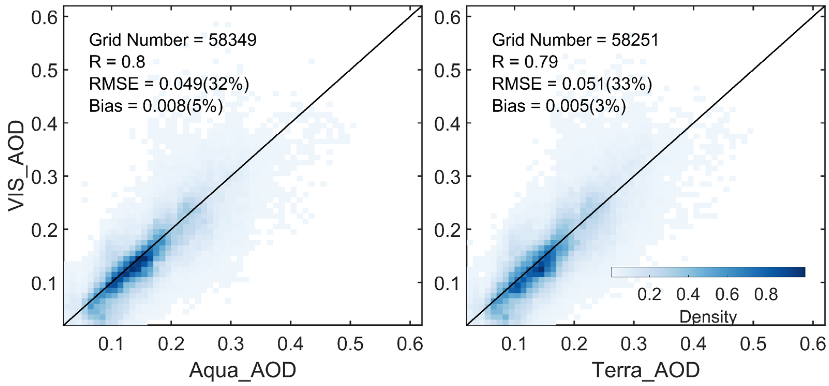


Figure 9 Heatmap of multi-year mean gridded VIS_AOD and Aqua AOD and Terra AOD during 2003-2021. Terra and Aqua AOD are averaged onto a grid of 0. 5°.

3.5 Global spatiotemporal variation of AOD in 1980-2021

The evaluation of visibility-derived AOD has demonstrated that the numerical and spatial distributions of VIS_AOD are in good agreement with the observations. Therefore, we employed VIS_AOD to analyze the spatiotemporal and seasonal distributions, and trends over land from 1980 to 2021. The analytical findings are shown as follows. The AOD mentioned below is the AOD derived from visibility.

We first analyzed the spatial distribution of multi-year average AOD over land from 1980 to 2021 and separately for the Southern Hemispheres (SH, -60-0°N) and Northern Hemisphere (NH, 0-85°N) in Figure 10 (a). The mean AOD of land, NH and SH is 0.161 ± 0.074, 0.158 ± 0.076, and 0.173 ± 0.059, respectively. The AOD values of Africa, Asia, Europe, North America, Oceania, and South America are 0.241, 0.222, 0.11, 0.111, 0.129 and 0.117, respectively. High AOD values occur in the NH, and align with the distribution of population density. Approximately 7/8 of the global population resides in the NH, with 50% concentrated at 20°N-40°N (Kummu et al., 2016), indicating a significant impact of human activities on aerosols. The highest AOD values are observed near 17°N, including the Sahara Desert, Arabian Peninsula, and southeastern India, suggesting thatin addition to anthropogenic sources, deserts also play a crucial role in aerosol emissions. Lower AOD values are found in the 25°S region of the SH, encompassing Australia, southern Africa, and southern South America, indicating lower aerosol burdens in these areas. Additionally, North America also exhibits low aerosol loading. Chin et al. (2014) analyzed the AOD over land from 1980 to 2009 with the Goddard Chemistry Aerosol Radiation and Transport model, which is similar to the visibility-derived AOD. The spatial distribution is consistent with the satellite results (Remer et al., 2008; Hsu et al., 2012; Hsu et al., 2017; Tian et al., 2023). The AOD and extinction coefficient retrieved from visibility show a similar distribution at global scale, with a correlation coefficient of nearly 0.6 (Mahowald et al., 2007). Similar global (Husar et al., 2000; Wang et al., 2009) and regional (Koelemeijer et al., 2006; Wu et al., 2014; Boers et al., 2015; Zhang et al., 2017; Zhang et al., 2020) spatial distributions have been reported.

AOD loadings exhibit significant seasonal variations worldwide, particularly over land. In this study, a year is divided into four parts: December-January-February (DJF), March-April-May (MAM),

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

June-July-August (JJA), and September-October-November (SON), corresponding to winter (summer), spring (autumn), summer (winter), and autumn (spring) in NH (SH), respectively. Figure 10 (b-e) also depicts the spatial distribution of seasonal average AOD over land from 1980 to 2021. The global AOD in DJF, MAM, JJA, and SON is 0.158±0.062, 0.162±0.081, 0.175±0.093, and 0.153± 0.07, respectively. The standard deviations of AOD in JJA and MAM are greater than those in DJF and SON. AOD exhibits seasonal changes, with the highest in JJA, followed by MAM, DJF, and SON. From 1980 to 2021, the seasonal AOD in NH is 0.152±0.064 (DJF), 0.161±0.088 (MAM), 0.176±0.09 (JJA), and 0.144±0.06 (SON), and in SH is 0.184±0.041 (DJF), 0.166±0.044 (MAM), 0.169±0.072 (JJA), and 0.19±0.06 (SON). In NH, the AOD ranking from high to low in season is summer > spring > winter > autumn. In SH, the AOD ranking from high to low in season is spring > summer > winter > autumn. The highest AOD is observed during JJA in NH, while in SH, the peak occurs during SON. The occurrence of high AOD values is highly associated with the intensification of industrial activities in Asia (JJA) (Remer et al., 2008) and Europe such as Russia (JJA), South America (SON), Southern Africa (SON), and biomass burning in Indonesia (SON) (Ivanova et al., 2010; Krylov et al., 2014), and the increased dust emissions in Middle East region related to the transport of dust from the Sahara region (Remer et al., 2008; Prakash et al., 2014). On the other hand, the lowest global AOD values are observed during autumn, which may be attributed to the influence of monsoon systems (Li et al., 2016; Zhao et al., 2019).

In addition to the spatial characteristics of AOD, the temporal variations in AOD have also been of great interest due to the significant relationship between aerosols and climate change. Figure 10 (f) shows the temporal trends of annual average AOD (** represents passing the significance test, $p<0.01$) over the global land, the SH and the NH during 1980-2021. The global land, NH, and SH trends demonstrate decreasing trends of AOD with values of -0.0026/10a, -0.0018/10a, and -0.0059/10a, respectively, with all passing the significance test with a confidence level of 95%. Notably, the declining trend is much greater in the SH than in the NH. The MODIS satellite results (including oceans) indicate trends of 0.004/10a, 0.009/10a, and -0.002/10a for the global, SH, and NH, respectively, during the period of 2003-2020. This findings suggest a growth trend in the global and NH and a declining trend in SH (Tian et al., 2023). The trend of SeaWiFS AOD was 0.0058/10a over land during 1998-2010 (Hsu et al., 2012). Our study has the same downward signal as that in previous studies. Two AOD peaks in 1983 and 1994 and two AOD valleys in 1980 and 1990 are observed before 2000. The two AOD peaks may be attributed to large volcanic eruptions, which has been confirmed by previous studies. The volcanic eruptions and their associated fires of the El Chichón volcano in Mexico in 1982 (Hirono & Shibata, 1983) and Mount Pinatubo in the Philippines in 1991(Tupper et al., 2005) resulted in elevating global AOD levels in the following years. The AOD recovery to the previous low levels after volcanic eruptions takes approximately 10 years (Chazette et al., 1995; Sun et al., 2019). This further indicates the efficiency of our data capturing the volcanic eruption emission features. also indicates that our data effectively captures this feature.

The distinct seasonal trends of AOD during 1980-2021 at the global and hemispheric scales are shown in Figure 10 (g-j). The global AOD shows a decreasing trend in all seasons (-0.002~-0.003/10a). The large declining trends are observed in JJA and SON, with decreasing trend values of -0.003/10a and -0.0029/10a, respectively. DJF and MAM follow with decreasing trend values of -0.0026/10a and -0.0022/10a, respectively, all passing the significance test ($p<0.01$). For the NH,

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

the AOD trends in different seasons are -0.003/10a (DJF), -0.0006/10a (MAM), -0.0005/10a (JJA), and -0.0034/10a (SON). DJF and SON pass the significance test ($p<0.01$), while MAM and JJA do not. In the SH, the trends are as follows: -0.0011/10a (DJF), -0.0085/10a (MAM), -0.0131/10a (JJA), and -0.0009/10a (SON). Interestingly, in contrast to the NH, MAM and JJA pass the significance test, while DJF and SON do not. The largest declining season in the NH is winter, while in the SH, it is summer. The decreasing trend in the SH is more than four times greater than that in the NH, particularly before the year 2000. While both the global and SH AOD exhibit a decreasing trend since 2005, the NH has shown a significant increase in winter AOD, leading to an overall increasing trend. Moreover, the NH shows an increasing trend of 0.004/10a from 2005 to 2021. Annual SO_2 emissions increased from 9.4 to 15.3 TgS from 2000 to 2005, which ultimately ended up as sulfate aerosols, leading to a significant increase in sulfate aerosols (Hofmann et al., 2009). More relevantly, the frequent volcanic eruptions in tropical regions from 2002 to 2006, combined with seasonal circulation patterns during winter, led to the transport of aerosol particles to higher latitudes (Hofmann et al., 2009; Vernier et al., 2011; Sawamura et al., 2012; Andersson et al., 2015).

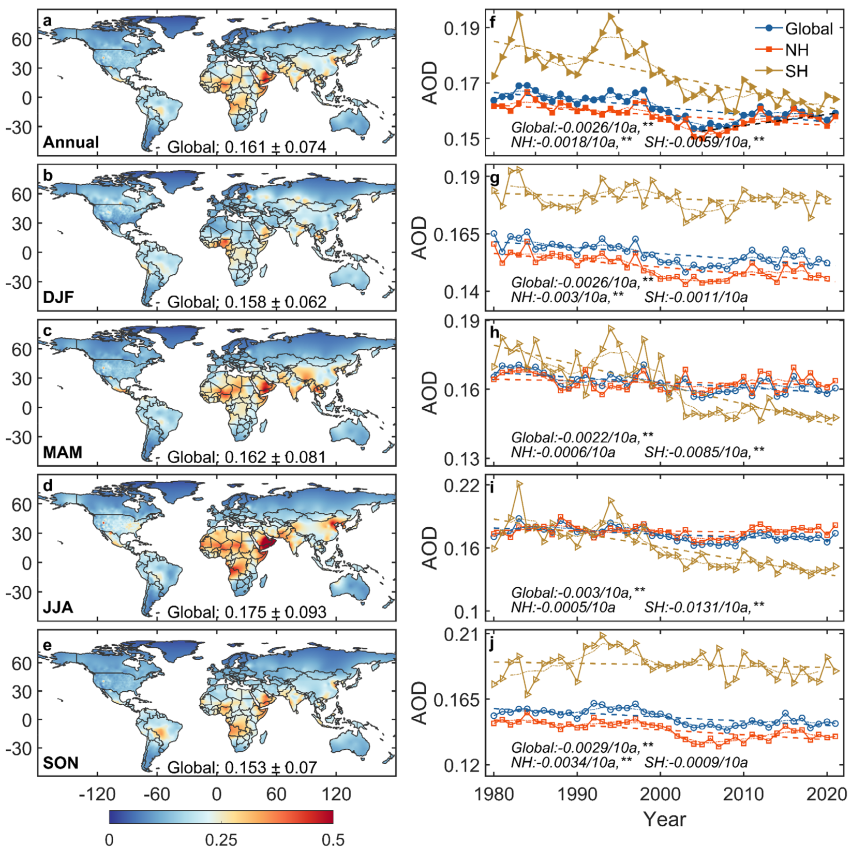


Figure 10 The multi-year averages of VIS_AOD from 1980 to 2021. Global (circle), northern hemisphere (NH,0-85°N) (triangle) and southern hemisphere (SH,0-60°S) (square) annual and seasonal AOD. The symbol, **, represents that the test passed at a significance level of 0.01. DJF

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

represents December and next January and February. MAM represents March, April, and May. JJA represents June, July, and August. SON represents September, October, and November.

3.6 Regional spatiotemporal variation in AOD during 1980-2021

The distribution of AOD over global land exhibits significant spatial heterogeneity. Large variations in aerosol concentrations exist among different regions, leading to a non-uniform spatial distribution of AOD globally. Accurately assessing the long-term trends of aerosol loading is a key for quantifying aerosol climate change, and it is crucial for evaluating the effectiveness of measures implemented to improve regional air quality and reduce anthropogenic aerosol emissions.

To analyze the spatiotemporal characteristics and trends of AOD in different regions, we selected 12 representative regions that are influenced by various aerosol sources(Wang et al., 2009; Hsu et al., 2012; Chin et al., 2014), such as desert, industry, anthropogenic emissions, and biomass burning emissions, which nearly cover the most land and are densely populated regions(Kummu et al., 2016). These representative regions are Eastern Europe, Western Europe, Western North America, Eastern North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and the Middle East, as shown in Figure 1.

We use multi-year average and seasonal average AOD to evaluate aerosol loadings (Figure 11), the annual average of monthly anomalies to analyze interannual trends (Figure 12),, and the seasonal average to analyze seasonal trends (Figure 13) in 12 regions from 1980 to 2021.

Figure 11 shows the regions with high aerosol loadings from 1980 to 2021 (multi-year average AOD > 0.2) are in West Africa, Northeast Asia, Eastern China, and the Middle East. The AOD values in Eastern North America, Central South America, South Africa, and Southeast Asia range from 0.15 to 0.2 with middle aerosol loadings. The AOD values in Eastern Europe, Western Europe, Western North America, and Australia are less than 0.15 with low aerosol loadings.

Europe is an industrial region with a low aerosol loading region, and the multi-year average AOD in Eastern Europe (0.144±0.007) is higher than that in Western Europe (0.139±0.003) during 1980-2021. Eastern Europe shows a greater downward trend in AOD (-0.0041/10a) compared to Western Europe (-0.0021/10a). The highest AOD is observed in JJA, the dry period when solar irradiation and boundary layer height increase, with Eastern Europe at 0.161 and Western Europe at 0.162, which could be due to increases in secondary aerosols, biomass burning, and dust transport from the Sahara (Mehta et al., 2016). However, there are seasonal variations. In Eastern Europe, the seasonal AOD ranking from high to low is JJA (0.161) > DJF (0.147) > MAM (0.138) > SON (0.131), while in Western Europe, it is JJA (0.162) > MAM (0.140) > SON (0.136) > DJF (0.117). The differences among seasons are larger in Western Europe. AOD in Eastern Europe shows declining trends in all seasons, while it does not pass the significance test in MAM. Among four seasons, SON has the largest decline trend of AOD (-0.0058/10a). In Western Europe, DJF, JJA, and SON exhibit declining trends of AOD that pass the significance test, while the MAM shows a significant increase trend of AOD (0.0022/10a), which may be due to eruptions of the Eyjafjallajökull volcano in Iceland in spring 2010 (Karbowska & Zembrzuski, 2016). Both Western and Eastern Europe experienced increasing trends in AOD during the period of 1995-2005, with Western Europe showing a greater increase. However, after 2000, the decline rate accelerated in both regions. The downward trend in Europe is attributed to the reduction of biomass burning,

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

anthropogenic aerosols, and aerosol precursors (such as sulfur dioxide)(Wang et al., 2009; Chin et al., 2014; Mortier et al., 2020).

North America is also an industrial region with a low aerosol loading. The average AOD values for Eastern and Western North America during 1980-2021 are 0.153±0.004 and 0.131±0.005, respectively, with the Eastern region being higher than the Western region by 0.022. From 1980 to 2021, both Eastern (-0.0021/10a) and Western North America (-0.0009/10a) show a downward trend; however, the decline in the Western region is not statistically significant. The average AOD values in DJF, MAM, JJA, and SON in Western North America are 0.1367, 0.1286, 0.1457, and 0.114, respectively, compared to 0.137, 0.145, 0.1913, and 0.138 in Eastern North America. The lowest AOD values of 12 regions during DJF and SON are observed in Western North America (Remer et al., 2008). Specifically, in the Western region, there is a consistent increasing trend during MAM (0.004/10a) from 1980 to 2021, while JJA and SON also show an increase after 2000, except for DJF (-0.0032/10a). In contrast, the AOD trends in the Eastern region remain unchanged during the period 1980-2021, except for MAM, which shows a stable increasing trend (0.0033/10a), while DJF, JJA, and SON exhibit decreasing trends (-0.0023/10a, -0.004/10a, -0.0053/10a, respectively). In the Western region, the annual mean AOD started to increase after 2005, while in the Eastern region, the increase was not significant. The upward trend may be due to low rainfall and increased wildfire activities (Yoon et al., 2014). The decrease in AOD in Eastern North America is related to the reduction of sulfate and organic aerosols, as well as the decrease in anthropogenic emissions caused by environmental regulations (Mehta et al., 2016).

Central South America is a relatively high aerosol loading region, sourced from biomass burning, especially in SON (Remer et al., 2008; Mehta et al., 2016), with a multi-year average AOD of 0.192±0.017. There is a clear downward trend (-0.01/10a) from 1980 to 2021, which is slightly greater than the trend (0.009/10a) from 1998 to 2010 (Hsu et al., 2012) and AOD decreased from 1980 to 2006 (Streets et al., 2009) and from 2001 to 2014 (Mehta et al., 2016). Although DJF (0.199) and SON (0.226) have higher values compared to MAM (0.18) and JJA (0.163), the large declining trends are observed in MAM (-0.0126/10a) and JJA (-0.0167/10a). It indicates that although AOD has decreased overall, the aerosol loading caused by seasonal deforestation and biomass combustion is still large(Mehta et al., 2016).

Africa is also one of the regions with a high aerosol loading worldwide. In West Africa, the average AOD is 0.275±0.0116 during 1980-2021, and the annual AOD shows a downward trend (-0.0008/10a, $p>0.05$). The world's largest desert (Sahara Desert) is in West Africa, with much dust aerosol discharged. AOD values in all seasons are above 0.25, with JJA (0.301) and MAM (0.3) reaching 0.3, and DJF and SON being 0.252 and 0.25 respectively. In addition to the dust source, frequent forest fires and biomass burning result in high AOD in JJA (Tian et al., 2023). The AOD in DJF (-0.0135/10a, $p<0.01$) and SON (-0.0026/10, $p>0.05$) exhibit decreasing trends, while JJA (0.0088/10a, $p<0.01$) and MAM (0.0037/10a, $p>0.05$) show an opposite trend. The multi-year average AOD in South Africa is 0.177±0.02, lower than that of West Africa. The annual mean AOD in South Africa shows a significant decrease (-0.0096/10a). The AOD values range from 0.12 to 0.2 during 2000-2009, dominated by fine particle matter from industrial pollution from biomass and fossil fuel combustion (Hersey et al., 2015). The average AOD values in DJF, MAM, JJA, and SON are 0.189, 0.162, 0.147, and 0.21, respectively. JJA (-0.0268/10a, $p<0.01$), MAM (-0.0126/10a, $p<0.01$) and SON (-0.0001/10a, $p>0.05$) exhibit a downward AOD trend, while DJF (0.0006/10a, p>0.05) shows an upward trend. AERONET and simulation results also show a decreasing trend of AOD (Chin et al., 2014).

Australia is a region with a low aerosol loading. The multi-year mean AOD is 0.127±0.014 during 1980-2021. The AOD ranges from 0.05 to 0.15 from AERONET during 2000-2021 and wildfires are an important contributor to the aerosol loading (Yang et al., 2021a). There is a downward trend of AOD (-0.0081/10a, p<0.01), which may be related to a decrease in BC and OC (Yoon et al., 2016). In addition, research has shown that the forest area in Australia has increased sharply since 2000 (Giglio et al., 2013), surpassing the forest fire area of the past 14 years. The seasonal average of AOD in MAM, JJA, SON, and DJF are 0.122, 0.108, 0.125, and 0.151. The AOD in JJA is the lowest among all seasons and regions. The highest AOD is in DJF with an increasing trend (0.0056/10a, p<0.01), while the trends during MAM, JJA and SON are -0.0096/10a (p<0.01), -0.0231/10a (p<0.01) and -0.0042/10a (p<0.01), respectively. Ground-based and satellite observations indicate that wildfires, biomass burning and sandstorms lead to high AOD in DJF and SON. The low AOD of MAM and JJA is due to a decrease in the frequency of sandstorms and wildfires and an increase in precipitation(Gras et al., 1999; Yang et al., 2021a; Yang et al., 2021b).

Asia is also a high aerosol loading area with various sources. In Southeast Asia, the multi-year average AOD is 0.177 during 1980-2021 with a downward trend of AOD (-0.0003/10a, p>0.05). It is also a biomass-burning area. The seasonal average AOD ranking from high to low is JJA (0.207) > MAM (0.183) > DJF (0.169) > SON (0.149). The trends in DJF (-0.0035/10a, p<0.05), JJA (-0.0007/10a, p>0.05) and SON (-0.0021/10a, p>0.05) are opposite to MAM (0.005/10a, p<0.01). Natural emissions were predominant in 1992 and 1997, because of the volcanic eruptions and forest fires. Southeast Asia has no clear long-term trend in estimated AOD or observed surface solar radiation (Streets et al., 2009). In Northeast Asia, the multi-year average AOD is 0. 222 during 1980-2021, with no significant temporal trend. The seasonal AOD values are 0.252 in MAM, 0.215 in DJF, 0.212 in SON and 0.209 in JJA. AOD in MAM is significantly higher than other seasons, which may be related to sandstorms in East Asia, and the reason for the high AOD in winter may be related to the low boundary layer height. The trends of AOD in DJF (-0.0025/10a, p>0.05), MAM (0.0031/10a, p>0.05), JJA (0) and SON (-0.0006/10a, p>0.05) are not significant. In Eastern China, the multi-year average AOD is 0.233, with an increasing trend (0.0071/10a, p<0.01). The seasonal average AOD ranking from high to low is JJA (0.284), MAM (0.234), SON (0.23) and DJF (0.183). The AOD trends in DJF (0.0093/10a, p<0.01), MAM (0.0092/10a, p<0.01), JJA (0.0038/10a, p>0.05) and SON (0.0065/10a, p<0.05) are all positive but the trend in JJA does not pass the significance test. We can see that there are three stages of changes in AOD: 1980-2005, 2006-2013 and 2014-2021. In the first stage, AOD increased steadily. In the second stage, AOD maintained a high level of volatility. The third stage experienced a rapid decline, reaching the level of the 1980s by 2021. The increasing trend of AOD before 2006 may be due to the significant increase in industrial activity, and after 2013, the significant decrease is closely related to the implementation of air quality-related laws and regulations, along with adjustments in the energy structure (Hu et al., 2018; Cherian & Quaas, 2020).

In the Middle East, aerosols are influenced by local deserts and aerosols transport from Africa and petroleum-related industries, resulting in high aerosol loading (Wei et al., 2019a; Wei et al., 2019b). The multi-year average AOD is 0.293, which is the highest among all 12 study regions, with an upward trend (0.0027/10a, p>0.05). The aerosol loading was higher during 1980-1990 and 2000-

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Open Access Earth System Science Data Discussions

2021 and lower during 1990-2000. The seasonal average AOD values are 0.201 in DJF, 0.319 in MAM, 0.394 in JJA, and 0.26 in SON. The trends of AOD in DJF (-0.0039/10a, p<0.05) and SON (-0.0012/10a, p>0.05) show an upward trend, while the trends in MAM (0.0067/10a, p<0.05) and JJA (0.0095/10a, p<0.01) are opposite. This increasing trend is related to sand and dust emissions (Klingmüller et al., 2016).

To summarize, there are significant differences in the spatial distribution, annual trends, and seasonal trends of AOD across different regions from 1980 to 201. The high aerosol loadings from 1980 to 2021 are in West Africa, Middle East and Asia, and low aerosol loading regions are in Europe, Western North America, and Australia. Eastern China and Middle East show an increasing trend, Southeast Asia and Northeast Asia show no significant trend, and the other regions show downward trends. However, not all regional seasonal trends are consistent with their annual trends. The results in this study have supplemented the long-term trend and distribution of AOD over land.

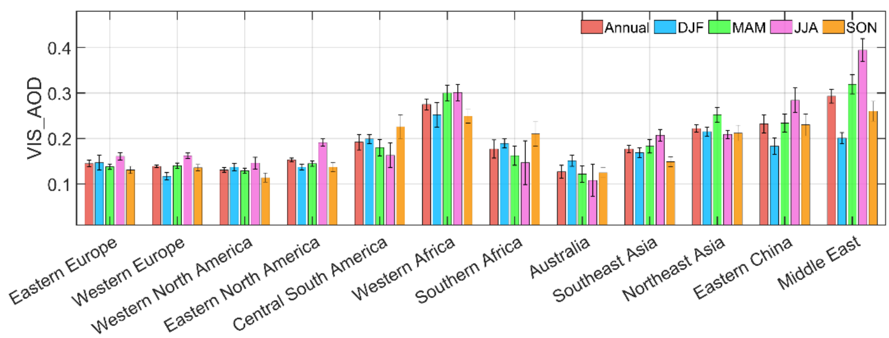


Figure 11 Annual and seasonal averages of AOD in 12 regions during 1980-2021.

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

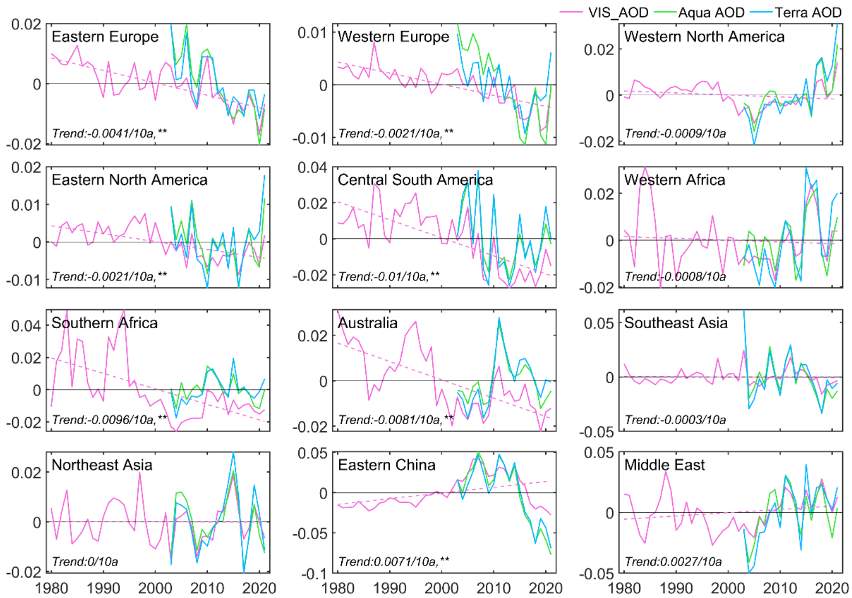
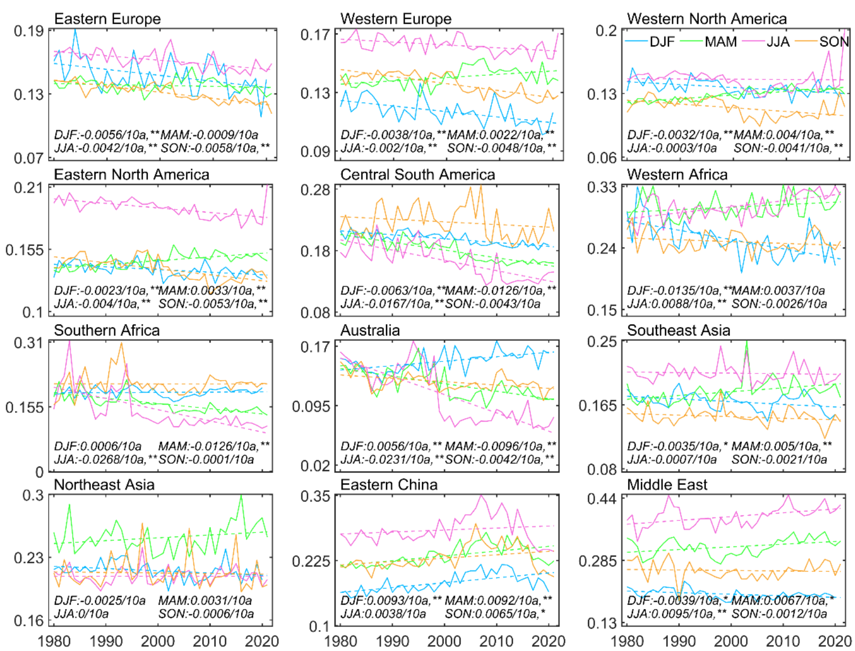


Figure 12 Annual averages of monthly anomaly gridded VIS_AOD (pink line), Aqua (green line), and Terra (blue line) MODIS AOD in 12 regions. The dotted line is the trend line. VIS_AOD has good temporal consistency with Aqua and Terra MODIS AOD from 2003 to 2021.



https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

Figure 13 Seasonal averages of gridded VIS_AOD during 1980 to 2021 in 12 regions (Eastern Europe, Western Europe, Western North America, Eastern North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and Middle East). The dotted line is the trend line.

4 Data availability

The visibility-derived AOD at station and grid scales over global land from 1980 to 2021 are available at National Tibetan Plateau / Third Pole Environment Data Center (https://doi.org/10.11888/Atmos.tpdc.300822) (Hao et al., 2023).

5 Conclusions

In this study, we employed a machine learning technique to derive AOD for over 5000 land stations worldwide, based on satellite data, visibility, and related parameters. Monthly AOD was interpolated onto a 0.5° grid using ordinary kriging with area weighting. The method was trained with Aqua MODIS AOD. The accuracy and performance of the derived AOD were assessed and validated against Terra MODIS AOD as well as AERONET ground-based observations of AOD for the corresponding stations. Evaluation of the gridded AOD was conducted using Aqua and Terra MODIS AOD. We obtained daily AOD for global land stations from 1980 to 2021, as well as monthly gridded AOD. The two datasets complement the shortcomings of AOD in terms of time scale and spatial coverage. Finally, the spatiotemporal variation in AOD was analyzed for global land, the Southern Hemisphere, the Northern Hemisphere, and 12 regions in the past 42 years. Several key findings have been obtained in this study as follows.

1. The longer the length of the training dataset is, the better the model performs. The RMSE, MAE, and R values for 100% of the training data are 0.078, 0.044, and 0.75, respectively. Increasing the training set length from 5% to 100% results in a 41.1% decrease in RMSE, a 50.1% decrease in MAE, and a 162.3% increase in the correlation coefficient. 97% of stations have a correlation coefficient above 0.7. The correlation coefficients of daily derived AOD with Aqua, Terra, and AERONET are 0.799, 0.542, and 0.514, respectively. The correlation coefficients of monthly derived AOD with Aqua, Terra, and AERONET are 0.936, 0.808, and 0.61, respectively. The correlation coefficients of the annual derived AOD with Aqua, Terra, and AERONET are 0.976, 0.906, and 0.62, respectively.

2. The gridded AOD is highly consistent with the satellite observations. The average biases of multi-year gridded AOD compared to Aqua and Terra are 3.3% and 1.9%, respectively. The spatial correlation coefficients are 0.8 and 0.79. The zonal correlation coefficients are 0.997 and 0.99, and the meridional correlation coefficients are 0.986 and 0.9.

3. From 1980 to 2021, the global, Northern Hemisphere (NH), and Southern Hemisphere (SH) AOD values are 0.161 ± 0.074, 0.158 ± 0.076, and 0.173 ± 0.059, respectively. Trends in AOD for the global, NH, and SH demonstrate a decreasing trend of -0.0026/10a, -0.0018/10a, and -0.0059/10a, respectively ($p<0.01$). The seasonal AOD ranking from high to low is JJA>MAM>DJF>SON over the global land and in the NH, while in the SH, it is DJF>JJA>MAM>SON. The largest declining trends are observed in NH summer and SH winter.

https://doi.org/10.5194/essd-2023-447
Preprint. Discussion started: 14 November 2023

4. From 1980 to 2021, regions with high aerosol loadings (AOD > 0.2) were found in West Africa, Northeast Asia, Eastern China, and the Middle East. Regions with moderate aerosol loadings (AOD between 0.15 and 0.2) are Eastern North America, Central South America, South Africa, and Southeast Asia. Eastern Europe, Western Europe, Western North America, and Australia are regions with low aerosol loadings (AOD < 0.15). Except for Northeast Asia (no trend), Eastern Asia (significant increasing trend), and the Middle East (insignificant increasing trend), other regions show an upward trend of AOD. There are also seasonal differences of AOD among regions, generally consistent with the seasonal variations in the NH or in the SH.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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