

# 1 **Visibility-derived aerosol optical depth over global land from 1980 to** 2 **2021**

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## 11 **Abstract**

12 Long-term and high spatial resolution aerosol optical depth (AOD) data are essential for climate  
13 change detection and attribution. Global ground-based AOD observation stations are sparsely  
14 distributed, and satellite AOD observations have a low temporal frequency, as well low accuracy  
15 before 2000 over land. In this study, AOD is derived from hourly visibility observations collected  
16 at more than 5000 meteorological stations over global land from 1980 to 2021. The AOD retrievals  
17 of the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua Earth  
18 observation satellite are used to train the machine learning model, and the ERA5 reanalysis  
19 boundary layer height is used to convert the surface visibility to AOD. Comparisons with  
20 independent dataset show that the predicted AOD has correlation coefficients of 0.54 and 0.55 with  
21 Terra MODIS satellite retrievals and AERONET ground observations at daily time scale. The  
22 correlation coefficients are higher at monthly and annual scales, which are 0.81 and 0.61 for the  
23 monthly and 0.91 and 0.65 for the annual, compared with Terra MODIS and AERONET AOD,  
24 respectively. The visibility-derived AOD at station scale is gridded into a 0.5° grid by ordinary  
25 kriging interpolation. The mean visibility-derived AOD over the global land (-60°N-85°N), the  
26 Northern Hemisphere, and the Southern Hemisphere are 0.161, 0.158, and 0.173, with a trend of -  
27 0.0026/10a, -0.0018/10a, and -0.0059/10a from 1980 to 2021. For the regional scale, the mean (trend)  
28 of AOD are 0.145 (-0.0041/10a), 0.139 (-0.0021/10a), 0.131 (-0.0009/10a), 0.153 (-0.0021/10a),  
29 0.192 (-0.0100/10a), 0.275 (-0.0008/10a), 0.177 (-0.0096/10a), 0.127 (-0.0081/10a), 0.177 (-  
30 0.0003/10a), 0.222 (-0.0000/10a), 0.232 (0.0071/10a), and 0.255 (0.0096/10a) in Eastern Europe,  
31 Western Europe, Western North America, Eastern North America, Central South America, Western  
32 Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India. The  
33 visibility-derived AOD at station and grid scales over global land from 1980 to 2021 are available  
34 at National Tibetan Plateau / Third Pole Environment Data Center  
35 (<https://doi.org/10.11888/Atmos.tpdc.300822>) (Hao et al., 2023).

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37 depth over global land (1980-2021). National Tibetan Plateau / Third Pole Environment Data

## 39 **1 Introduction**

40 Atmospheric aerosols are composed of solid and liquid particles suspended in the atmosphere.  
41 Aerosol particles are directly emitted into the atmosphere or formed through gas-particle  
42 transformation (Calvo et al., 2013), with diverse shapes and sizes (Fan et al., 2021), optical  
43 properties, and various components (Liao et al., 2015; Zhang et al., 2020; Li et al., 2022). Most  
44 atmospheric aerosols are concentrated in the troposphere, especially in the boundary layer (Liu et  
45 al., 2022), with a high concentration near emission sources (Kulmala et al., 2004) , and a small  
46 portion are distributed in the stratosphere. Atmospheric aerosols severely impact the atmospheric  
47 environment and human health. They deteriorate air quality, reduce visibility, and cause other  
48 environmental issues (Wang et al., 2012; Boers et al., 2015). They impair human health or other  
49 organisms' conditions by increasing cardiovascular and respiratory disease incidence and mortality  
50 rates (Chafe et al., 2014; Yang et al., 2022). The Global Burden of Disease shows that global  
51 exposure to ambient PM<sub>2.5</sub> resulted in 0.37 million deaths and 9.9 million disability-adjusted life  
52 years (Chafe et al., 2014).

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

61 Tropospheric aerosols are considered as the second largest forcing factor for global climate change  
62 (Li et al., 2022), and they reduce the warming due to greenhouse gases by -0.5°C (IPCC, 2021).  
63 However, aerosols are also regarded as the largest contributor to quantifying the uncertainty of  
64 present-day climate change (IPCC, 2021). The uncertainties are caused by the deficiencies of the  
65 global descriptions of aerosol optical properties (such as scattering and absorption) and  
66 microphysical properties (such as size and component), and the impact on cloud and precipitation,  
67 further affecting the estimation of aerosol radiative forcing (Lee et al., 2016; IPCC, 2021). Therefore,  
68 sufficient aerosol observations are crucial. In aerosol measurements, aerosol optical depth (AOD)  
69 is often used to describe its column properties, which represents the vertical integration of aerosol  
70 extinction coefficients. AOD is an important physical quantity for estimating the content,  
71 atmospheric pollution and climatology of aerosols (Zhang et al., 2020).

72 AOD data usually from ground-based and satellite-borne remote sensing observation. They have  
73 both advantages and disadvantages. Ground-based lidar observation is an active remote sensing  
74 technology. Lidar generally emits laser and receives backscattered signals to invert the extinction  
75 coefficient of aerosols at different heights (Klett, 1985). By using the depolarization ratio, the type  
76 of aerosol, such as fine particles or dust, can be distinguished (Bescond et al., 2013). The AOD  
77 within a certain height can be calculated by integrating the extinction coefficients; however,  
78 scattering signals are usually not received near the ground, leading to blind spots (Singh et al., 2019).

79 At present, there are many ground-based lidar worldwide and regional networks, which provides  
80 important support of vertical changes in aerosols, such as the NASA Micro-Pulse Lidar Network  
81 (MPLNET) in the early 1990s (Welton et al., 2002), the European Aerosol Research Lidar Network  
82 (EARLINET) since 2000 (Bösenberg and Matthias, 2003), the Latin American Lidar Network  
83 (LALINET) since 2013 (Guerrero-Rascado et al., 2016).

84 Ground-based remote sensing observations supply aerosol loading data (such as AOD), by  
85 measuring the attenuation of radiation from the top of the atmosphere to the surface (Holben et al.,  
86 1998). This type of observations mainly uses weather-resistant automatic sun and sky scanning  
87 spectral radiometers to retrieve optical and microphysical aerosol properties (Che et al., 2014). The  
88 Aerosol Robotic Network (AERONET) is a popular global network composed of NASA and  
89 multiple international partners that provides high-quality and high-frequency aerosol optical and  
90 microphysical properties under various geographical and environmental conditions (Holben et al.,  
91 1998; Dubovik et al., 2000). The AERONET observations are extensively used to validate satellite  
92 remote sensing observations and model simulations, as well as climatology study (Dubovik et al.,  
93 2002b). There are many regional networks of sun photometers, such as the Maritime Aerosol  
94 Network (MAN), which use a handheld sun photometer to collect data over the ocean and is merged  
95 into AERONET (Smirnov et al., 2009), the China Aerosol Robot Sun Photometer Network  
96 (CARSNET) (Che et al., 2009), the Canadian sub-network of AERONET (AEROCAN) (Bokoye et  
97 al., 2001), Aerosol characterization via Sun photometry: Australian Network (AeroSpan)  
98 (Mukkavilli et al., 2019), and the sky radiometer network (SKYNET) in Asia and Europe (Kim et  
99 al., 2004; Nakajima et al., 2020). Another very valuable global network is the NOAA/ESRL  
100 Federated Aerosol Network (FAN), which uses integrated nephelometers distinct from sun  
101 photometers, mainly located in remote areas, providing background aerosol properties over 30 sites  
102 (Andrews et al., 2019).

103 Satellite remote-sensing is a space-based method that can provide aerosol properties worldwide.  
104 With the development of satellite remote sensing technology since 1970s, aerosol distributions can  
105 be extracted with the advantage of sufficient real-time and global coverage from multiple satellite  
106 sensors (Kaufman and Boucher, 2002; Anderson et al., 2005). The Advanced Very High Resolution  
107 Radiometer (AVHRR) is the earliest sensor used for retrieving AOD over ocean (Nagaraja Rao et  
108 al., 1989). The Moderate Resolution Imaging Spectroradiometer (MODIS), on board the Terra  
109 (launched in 1999) and Aqua (launched in 2002) satellites is a popular sensor with 36 channels,  
110 which have been used for AOD retrieval over both ocean and land based on the Dark Target and the  
111 Deep Blue algorithms (Remer et al., 2005; Levy et al., 2013). The latest MODIS AOD data version  
112 is the Collection 6.1, which provides global AOD over 20 years (Wei et al., 2019). There are also  
113 many other satellite sensors that can be used to retrieve AOD, such as the Polarization and  
114 Directionality of the Earth's Reflectances (POLDER) during 1996-1997, 2003 and 2004-2013  
115 (Deuzé et al., 2000), Sea-viewing Wide Field-of-view Sensor (SeaWiFS) during 1997-2007  
116 (O'Reilly et al., 1998), the Multi-angle Imaging Spectroradiometer (MISR) on Terra since 1999  
117 (Diner et al., 1998). The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) has also  
118 derived aerosols in the vertical direction since 2006 (Winker et al., 2009).

119 These measurements provide important data for studying the global and regional spatiotemporal  
120 variabilities and climate effect of aerosols. However, ground-based remote sensing observations  
121 only provide aerosol properties with low spatial coverage. There were only 1126 ground stations

122 worldwide in 2002 and even fewer sites were available for climate analysis (Holben et al., 1998;  
123 Chu et al., 2002), which limited aerosol climate research by spatial coverage (Bright and Gueymard,  
124 2019). Satellite remote sensing overcomes the limitations of spatial coverage. The AVHRR has been  
125 used to retrieve AOD since 1980, but it is limited by a few channel number, low spatial resolution,  
126 and insufficient validation through ground-based observations before 2000 (Hsu et al., 2017). Many  
127 studies have only investigated the trends and distributions of aerosols after 2000 (Bösenberg and  
128 Matthias, 2003; Winker et al., 2013; Xia et al., 2016; Tian et al., 2023), because of the lack of long-  
129 term and global cover AOD products, which is the bottleneck for aerosol climate change detection  
130 and attributions.

131 To overcome these limitations and enrich aerosol data, alternative observation data could be utilized  
132 to derive AOD. Atmospheric horizontal visibility is a suitable alternative (Wang et al., 2009; Zhang  
133 et al., 2020), because it has the advantages of the long-term records with a large number of stations  
134 worldwide.

135 Atmospheric visibility is a physical quantity that describes the transparency of the atmosphere  
136 through manual and automatic observations, and the automatic observations of visibility usually  
137 measure atmospheric extinction (scattering coefficient and transmissivity). Koschmieder (1924)  
138 first proposed the relationship between the meteorological optical range and the total optical depth.  
139 Elterman (1970) further established a formula between AOD and visibility by assuming an  
140 exponential decrease in aerosol concentration with altitude, considering the extinction of molecules  
141 and ozone to analyze air pollution, which called the Elterman model. Qiu and Lin (2001) corrected  
142 the Elterman model by considering the influence of water vapor and used two water vapor pressure  
143 correction coefficients to retrieve AOD of 16 stations in China in 1990. Wang et al. (2009) analyzed  
144 the trend of AOD using visibility-based retrievals from 1973 to 2007 over land. Lin et al. (2014)  
145 retrieved the AOD in eastern China in 2006 using visibility and aerosol vertical profiles provided  
146 by GEOS-Chem. Wu et al. (2014) and Zhang et al. (2017) parameterized the constants in the  
147 Elterman model and use satellite retrieved AOD to solve the parameters in the models at different  
148 stations, to retrieve the long-term AOD in China.

149 Zhang et al. (2020) reviewed the methods of visibility retrieval of AOD, indicating that visibility-  
150 based retrieval of AOD can compensate for the shortcomings of long-term aerosol observation data.  
151 Simultaneously, various parameters, such as station altitude, consistency of visibility data, water  
152 vapor and aerosol vertical profiles (scale height), were discussed with modified suggestions  
153 proposed. These studies have enriched AOD data regionally. These studies have enriched aerosol  
154 data in some extent. At present, there are very few studies on global visibility-retrieved AOD and to  
155 analyze climatology of aerosols.

156 The two physical quantities of visibility and AOD have both connections and differences, making it  
157 challenging to retrieve AOD from visibility. Visibility represents the maximum horizontal visible  
158 distance near the surface, while AOD represents the total vertical attenuation of solar radiation by  
159 aerosols. The visibility of automatic observation is dependent on the local horizontal atmospheric  
160 extinction (NOAA et al., 1998). Visibility has not a simple linear relationship with meteorological  
161 factors. The vertical structure of aerosols is the greatest challenge to obtain, as it is not a simple  
162 hypothetical curve in complex terrain and circulation conditions (Zhang et al., 2020). These  
163 limitations make it more complex to derive AOD. Machine learning methods can effectively address

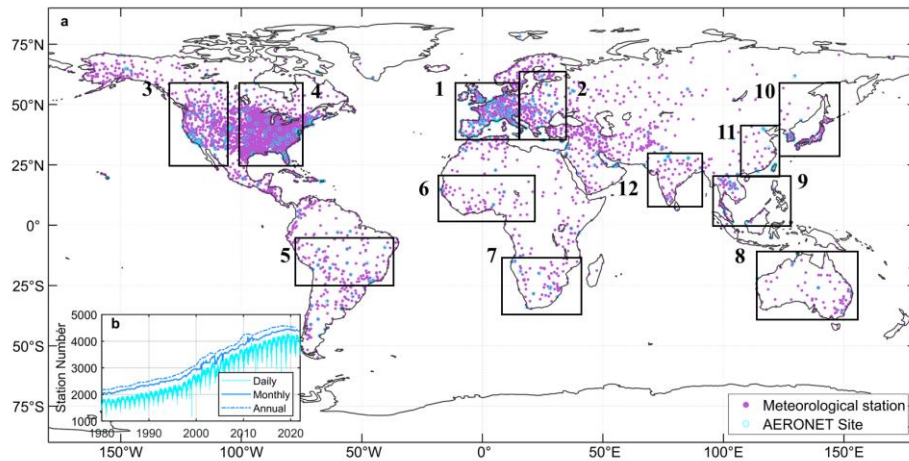
164 complex nonlinear relationships between variables and have been widely applied in remote sensing  
165 and climate research fields. Li et al. (2021) used the random forest method to predict PM<sub>2.5</sub> in Iraq  
166 and Kuwait based on satellite AOD during 2001-2018. Kang et al. (2022) applied LightGBM and  
167 random forest to estimate AOD over East Asia, and the results showed a consistency with  
168 AERONET. Dong et al. (2023) derived aerosol single scattering albedo from visibility and satellite  
169 AOD over 1000 global stations. Hu et al. (2019) used a deep learning method to retrieve horizontal  
170 visibility from MODIS AOD. These studies have confirmed the ability of machine learning to  
171 effectively solve complex relationships among variables. And previous studies are mostly  
172 conducted at the regional or national scale, and few studies at the global scale. Thus, it is feasible to  
173 derive AOD from atmospheric visibility over global land by using the machine learning method.

174 In this study, we propose a machine learning method to derive AOD, where satellite AOD is the  
175 target value, and visibility and other related meteorological variables are the predictors. We explain  
176 the robustness of the model, validate the model's predictions using ground-based AOD and  
177 independent satellite retrievals, and analyze the mean and trend of AOD across land and regions.  
178 Two datasets of long-term high-resolution AOD are generated. The Section 2 introduces the data  
179 and method. The Section 3 is the evaluation and validation of the visibility-derived AOD, and the  
180 distribution and trends are discussed at global and regional scales. The Section 4 presents the  
181 conclusions. This study is dedicated to supporting the research of aerosols in climate change  
182 detection and attribution.

## 183 **2 Data and method**

### 184 **2.1 Study area**

185 The study area is global land. A total of 5032 meteorological stations and 395 AERONET sites are  
186 selected in this study, shown in Figure 1. Twelve regions are selected for special analysis, including  
187 Eastern Europe, Western Europe, Western North America, Eastern North America, Central South  
188 America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China,  
189 and India. The time range of the study is from 1980 to 2021, during which the records of  
190 meteorological stations are sufficient with a uniform spatial distribution. As shown in Figure 1, the  
191 daily records have exceeded 1500 stations, and monthly and annual records have exceeded 2000  
192 during 1980-1990. After 2000, monthly records have reached 3000, which is the foundation of  
193 gridding AOD.



194

195 **Figure 1** Study area (a) and the meteorological station number (b) with daily, monthly, and annual  
 196 records. The number of meteorological stations (filled circles) is 5032. The number of AERONET  
 197 sites (empty circles) is 395. The box regions of labelled with number 1-12 are Eastern Europe,  
 198 Western Europe, Western North America, Eastern North America, Central South America, Western  
 199 Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India.

## 200 2.2 Meteorological data

201 The ground hourly data from 1980 to 2021 is collected from 5032 automated meteorological stations  
 202 of airports over land. Automated surface observations reduce errors associated with human  
 203 involvement in data collection, processing, and transmission. The data can be downloaded at  
 204 <https://mesonet.agron.iastate.edu/ASOS>. The data is extracted from the Meteorological Terminal  
 205 Aviation Routine Weather Report (METAR). The World Meteorological Organization (WMO) sets  
 206 guidelines for METAR reports, including report format, encoding, observation instruments and  
 207 methods used, data accuracy, and consistency. These requirements ensure consistency and  
 208 comparability of METAR reports globally. International regulations can be referenced at  
 209 [https://community.wmo.int/en/implementation-areas-aeronautical-meteorology-](https://community.wmo.int/en/implementation-areas-aeronautical-meteorology-programme)  
 210 programme. Among them, over 1,000 stations belong to the Automated Surface Observing System  
 211 (ASOS), and others are sourced from airport reports around the world.

212 The daily average visibility is calculated using harmonic mean. Experiments have found that  
 213 harmonic average visibility can better detect the weather phenomena than arithmetic average  
 214 visibility (NOAA et al., 1998). The visibility is calculated using the extinction coefficient, which is  
 215 directly proportional to the reciprocal of visibility (Wang et al., 2009). Harmonious average  
 216 visibility can capture the process of visibility decline more quickly. Therefore, daily visibility will  
 217 have greater representativeness:

$$218 \quad V = n / \left( \frac{1}{v_1} + \frac{1}{v_2} + \dots + \frac{1}{v_n} \right) \quad \text{Eq. 1}$$

219 where V is the harmonic mean visibility,  $n = 24$  for the daily visibility, and  $V_1, V_2, \dots, V_n$  are the  
 220 individual hourly visibility.

221 In addition to hourly visibility (VIS), other variables closely related to aerosol properties are selected,

222 including relative humidity (RH), dew point temperature (DT), temperature (TMP), wind speed  
223 (WS) and sea-level pressure (SLP). Temperature affects atmospheric stability and the rate of  
224 secondary particle formation, and humidity influences the size and hygroscopic growth, and wind  
225 speed and pressure significantly impact the transport and deposition. Sky conditions (cloud amount)  
226 and hourly precipitation are also selected to remove the records of extensive cloud cover and  
227 precipitation.

228 We processed the data as follows. The records with missing values are eliminated (Husar et al.,  
229 2000). When over 80% overcast or fog, the records of sky conditions are eliminated, though such  
230 situations occur less than 1% of the time over land (Remer et al., 2008). The records with 1-hour  
231 precipitation greater than 0.1 mm are eliminated. We calculate the temperature dew point difference  
232 (dT). When the RH is greater than 90%, it is impossible to distinguish whether it is fog or haze, or  
233 both, and even precipitation. The records with RH greater than or equal to 90% are eliminated.  
234 When the RH is less than 30%, the dilution effect of aerosols is very low or even negligible. When  
235 RH is between 30% and 90%, visibility is converted to dry visibility (Yang et al., 2021c):

$$236 \quad \text{VISD} = \text{VIS} / (0.26 + 0.4285 * \log(100 - \text{RH})) \quad \text{Eq. 2}$$

237 where VISD is the dry visibility.

238 Daily averages of variables are calculated by at least 3 hourly records.

### 239 **2.3 Boundary layer height**

240 The hourly boundary layer height (BLH) from 1980 to 2021 is available from the Fifth Generation  
241 reanalysis of the European Medium-Range Weather Forecast Center (ERA5) with a resolution of  
242  $0.25^\circ \times 0.25^\circ$  (<https://cds.climate.copernicus.eu>), which is the successor of ERA-Interim and has  
243 undergone various improvements (Hersbach et al., 2020). The atmospheric boundary layer is the  
244 layer closest to the Earth's surface and exhibits complex turbulence activities, and its height  
245 undergoes significant diurnal variation. The effects of the boundary layer on aerosols are mainly  
246 manifested in vertical distribution, concentration changes, transport, and deposition (Ackerman et  
247 al., 1995). The characteristics and variations in the boundary layer play a crucial role in regulating  
248 and adjusting the distribution of atmospheric aerosols. The boundary layer height serves as an  
249 approximate measure of the scale height for aerosols (Zhang et al., 2020).

250 Compared to observations of 300 stations over world from 2012 to 2019, the BLH of ERA5 was  
251 underestimated by 131.96m. Compared with the underestimated MERRA-2 (166.35m), JRA-55  
252 (351.49m), and NECP-2 (420.86m), the BLH of ERA5 was closest to the observations (Guo et al.,  
253 2021). The BLH hourly data is temporally and spatially matched with the meteorological data before  
254 calculating the daily average.

255 Because the inverse of visibility is proportional to the extinction coefficient and positively related  
256 to AOD (Wang et al., 2009), we calculated the reciprocal of visibility (VISI) and the reciprocal of  
257 dry visibility (VISDI). Due to the influence of boundary layer height on the vertical distribution of  
258 particles (Zhang et al., 2020), we calculated the product (VISDIB) of the reciprocal of dry visibility  
259 and BLH. Therefore, the Predictor (Figure 2) is composed of 11 variables (TMP, Td, dT, RH, SLP,  
260 WS, VIS, BLH, VISI, VISDI, and VISDIB).

### 261 **2.4 MODIS AOD Products**

262 Satellite daily AOD is available from the Moderate Resolution Imaging Spectroradiometer (MODIS)  
263 Level 3 Collection 6.1 AOD products of the Aqua (MYD09CMA) satellite from 2002 to 2021 and  
264 Terra (MOD09CMA) satellite from 2000 to 2021 with a spatial resolution of  $0.05^\circ \times 0.05^\circ$  at a  
265 wavelength of 550 nm (<https://ladsweb.modaps.eosdis.nasa.gov>). MOD/MYD09 has a higher  
266 spatial resolution than MOD/MYD08 ( $1^\circ \times 1^\circ$ ), which may result in a greater difference in AOD  
267 values and reduce the proximity ratio to match the visibility-derived AOD at station scale. Terra  
268 (passing approximately 10:30 am local time) and Aqua (passing approximately 1:30 pm local time)  
269 were successfully launched in December 1999 and May 2002, respectively.

270 MODIS, carried on the Terra and Aqua satellites is a crucial instrument in the NASA Earth  
271 Observing System program, which is designed to observe global biophysical processes  
272 (Salomonson et al., 1987). The 2,330 km-wide swath of the orbit scan can cover the entire globe  
273 every one to two days. MODIS has 36 channels and more spectral channels than previous satellite  
274 sensors (such as AVHRR). The spectral range from 0.41 to  $15\mu\text{m}$  representing three spatial  
275 resolutions: 250 m (2 channels), 500 m (5 channels), and 1 km (29 channels). The aerosol retrieval  
276 algorithms use seven of these channels ( $0.47\text{--}2.13\mu\text{m}$ ) to retrieve aerosol characteristics and uses  
277 additional wavelengths in other parts of the spectrum to identify clouds and river sediments.  
278 Therefore, it has the ability to characterize the spatial and temporal characteristics of the global  
279 aerosol field.

280 The MODIS aerosol product actually takes use of different algorithms for deriving aerosols over  
281 land and ocean. The Dark Target (DT) algorithm is applied to densely vegetated areas because the  
282 surface reflectance over dark-target areas was lower in the visible channels and had nearly fixed  
283 ratios with the surface reflectance in the shortwave and infrared channels (Levy et al., 2007; Levy  
284 et al., 2013). The Deep Blue (DB) algorithm was originally applied to bright land surfaces (such as  
285 deserts), and later extended to cover all cloud-free and snow-free land surfaces (Hsu et al., 2006;  
286 Hsu et al., 2013). MODIS Collection 6.1 aerosol product was released in 2017, incorporating  
287 significant improvements in radiometric calibration and aerosol retrieval algorithms.

288 The expected errors are  $\pm (0.05 \pm 15\%)$  for the DT retrievals over land. Higher spatial coverage is  
289 observed in August and September, reaching 86-88%. During December and January, due to the  
290 presence of permanent ice and snow cover in high-latitude regions of the Northern Hemisphere, the  
291 spatial coverage is 78-80%. Thus, challenges remain in retrieving AOD values in high-latitude  
292 regions (Wei et al., 2019). However, visibility observations are available in high-latitude regions,  
293 thereby partially addressing the lack in these regions.

294 In this study, the Terra and Aqua MODIS AOD are temporally and spatially matched with the  
295 meteorological stations. Aqua MODIS AOD is used as the Target, when training the model, and  
296 Terra MODIS AOD is used in the evaluation and validation of the model results, as shown in the  
297 flowchart (Figure 2).

## 298 **2.5 Ground-based AOD**

299 Ground-based 15-minute AOD data are available from the Aerosol Robotic Network (AERONET)  
300 Version 3.0 Level 2.0 product at 395 stations (Figure 1), which can be downloaded from  
301 <https://aeronet.gsfc.nasa.gov>. The AERONET program is a federation of ground-based remote  
302 sensing aerosol networks established by NASA and PHOTONS, including many subnetworks (such  
303 as AeroSpan, AEROCAN, NEON, and CARSNET). The sun photometer (CE-318) measures  
304 spectral sun and sky irradiance in the 340-1020 nm spectral range. When the aerosol loading is low,



305 the error is significant. When the AOD at 440 nm wavelength is less than 0.2, the error is 0.01,  
 306 which is equivalent to the error of the absorption band in the total optical depth (Dubovik et al.,  
 307 2002a). The total uncertainty in AOD under cloud-free conditions is less than  $\pm 0.01$  for wavelength  
 308 more than 440 nm, and  $\pm 0.02$  for wavelength less than 440 nm (Holben et al., 1998). AERONET  
 309 has three levels of AOD products: Level 1.0 (unscreened), Level 1.5 (cloud screened), and Level  
 310 2.0 (cloud screened and quality assured). Compared to Version 2, the Version 3 Level 2.0 database  
 311 has undergone further cloud screening and quality assurance, which is generated based on Level 1.5  
 312 data with pre- and post-calibration and temperature adjustment and is recommended for formal  
 313 scientific research (Giles et al., 2019). AERONET provides AOD products at wavelengths of 440,  
 314 675, 870, and 1020 nm. The AOD at 440nm and the Ångström index at 440-675nm are used for  
 315 AOD at 550 nm not provided by AERONET, as shown in Eq. 3. AERONET AOD, as the ‘true’  
 316 value, is the average of at least two times within 1 hour ( $\pm 30$  minutes) of Aqua transit time (Wei et  
 317 al., 2019).

$$318 \quad \tau_{550} = \tau_{440} \left( \frac{550}{440} \right)^{-\alpha} \quad \text{Eq. 3}$$

319 where  $\tau_{440}$  and  $\tau_{550}$  are the AOD at a wavelength of 440nm and 550 nm, and  $\alpha$  is the Ångström  
 320 index.

321 The matching conditions between AERONET sites and meteorological stations are (1) a distance of  
 322 less than  $0.5^\circ$  (2) at least three years of observation. Finally, a total of 395 pairs were matched.

## 323 **2.6 Decision Tree Regression**

### 324 **2.6.1 Feature selection**

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

### 337 **2.6.2 Data balance**

338 When it is clear, the AOD value is small, the variability of AOD is small ( $AOD < 0.5$ ), and the data is  
 339 concentrated near the mean value. When heavy pollution, the AOD value is large ( $AOD > 0.5$ ). Compared  
 340 to clear sky, the AOD sequence will show "abnormal" large values with low frequency, which is the  
 341 imbalance of AOD data. When dealing with imbalanced datasets, because of the tendency of machine  
 342 learning algorithms to perform better on the majority class and overlook the minority class, the model  
 343 can be underfit (Chuang and Huang, 2023). Data augmentation techniques are commonly employed to

344 address the issue in imbalance data, which applies a series of transformations or expansions to generate  
345 new training data, thereby increasing the diversity and quantity of the training data.

346 The Adaptive Synthetic Sampling (ADASYN) is a data augmentation technique specifically designed to  
347 address data imbalance problem (He et al., 2008; Mitra et al., 2023). It is an extension of the Synthetic  
348 Minority Over-sampling Technique (SMOTE) algorithm (Fernández et al., 2018). The goal of ADASYN  
349 is to generate synthetic sample data for the minority class to increase its representation in the dataset.  
350 ADASYN, which adaptively adjusts the generation ratio of synthetic samples based on the density  
351 distribution of sample data, improves the dataset balance and enhances the performance of machine  
352 learning models in dealing with imbalanced data.

353 The processing of imbalanced data includes (1) AOD sequences are classified into three types based on  
354 percentile (0-1%, 2% -98%, 99%), (2) When the mean of the third type of AOD is greater than 5 times  
355 the standard bias of the second type, it is considered an imbalanced sequence. These data, with a total  
356 amount less than 5% of the sample, are imbalanced data, and (3) Then synthetic samples are generated  
357 with the upper limit 10% of the samples.

### 358 **2.6.3 Decision Tree Regression Model**

359 The decision tree is a machine learning algorithm based on a tree-like structure used to solve  
360 classification and regression problems. We adopt the CART algorithm to construct a regression tree by  
361 analyzing the mapping relationship between object attributes (Predictors) and object values (Target). The  
362 internal nodes have binary tree structures with feature values of "yes" and "no". In addition, each leaf  
363 node represents a specific output for a feature space. The advantages of the regression tree include the  
364 ability to handle continuous features and the ease of understanding the generated tree structure (Teixeira,  
365 2004; Steinberg and Colla, 2009). Before training the tree model, the variables (Input) are normalized to  
366 improve model performance, and after prediction, the results are obtained by denormalization. The 10-  
367 fold cross-validation method is employed to improve the generalization ability of the model (Browne,  
368 2000).

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

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

$$377 \quad f(x) = \sum_{m=1}^M c_m I(x \in R_m), m = 1, 2, \dots, M \quad \text{Eq. 4}$$

378 where  $I$  is the indicator function (Eq. 5):

$$379 \quad I = \begin{cases} 1, & x \in R_m \\ 0, & x \notin R_m \end{cases} \quad \text{Eq. 5}$$

380 When the partition of the input space is determined, the square error can be used to represent the  
381 prediction error of the regression tree for the training data, and the minimizing square error is used to

382 solve the optimal output value on each domain. The optimal value ( $\widehat{c}_m$ ) on a domain is the mean of the  
 383 outputs corresponding to all input, namely:

$$384 \quad \widehat{c}_m = \text{ave}(\mathbf{y}_i | \mathbf{x}_i \in \mathbf{R}_m) \quad \text{Eq. 6}$$

385 A heuristic method is used to split the feature space in CART. After each split, all values of all features  
 386 in the current set are examined individually, and the optimal one is selected as the split point based on  
 387 the principle of minimum sum of the square errors. The specific step is described as follows: for the  
 388 training dataset D, we recursively divide each region into two sub domains and calculate the output  
 389 values of each sub domain; then, construct a binary decision tree. For example, split variable is  $x^j$  and  
 390 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  
 391 solve the loss function  $L(j, s)$  to find the optimal  $j$  and  $s$ .

$$392 \quad L(\mathbf{j}, \mathbf{s}) = \sum_{\mathbf{x}_i \in R_1(\mathbf{j}, \mathbf{s})} (\mathbf{y}_i - \mathbf{c}_1)^2 + \sum_{\mathbf{x}_i \in R_2(\mathbf{j}, \mathbf{s})} (\mathbf{y}_i - \mathbf{c}_2)^2 \quad \text{Eq. 7}$$

393 When  $L(\mathbf{j}, \mathbf{s})$  is the smallest,  $x^j$  is the optimal split variable and  $s$  is the optimal split point for the  
 394  $x^j$ .

$$395 \quad \min_{\mathbf{j}, \mathbf{s}} \left[ \min_{\mathbf{c}_1} \sum_{\mathbf{x}_i \in R_1(\mathbf{j}, \mathbf{s})} (\mathbf{y}_i - \mathbf{c}_1)^2 + \min_{\mathbf{c}_2} \sum_{\mathbf{x}_i \in R_2(\mathbf{j}, \mathbf{s})} (\mathbf{y}_i - \mathbf{c}_2)^2 \right] \quad \text{Eq. 8}$$

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

$$398 \quad \widehat{c}_1 = \text{ave}(\mathbf{y}_i | \mathbf{x}_i \in \mathbf{R}_1(\mathbf{j}, \mathbf{s})), \widehat{c}_2 = \text{ave}(\mathbf{y}_i | \mathbf{x}_i \in \mathbf{R}_2(\mathbf{j}, \mathbf{s})) \quad \text{Eq. 9}$$

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

402 Therefore, the regression tree model  $f(\mathbf{x})$  can be represented as follows:

$$403 \quad f(\mathbf{x}) = \sum_{m=1}^M \widehat{c}_m I(\mathbf{x} \in \mathbf{R}_m), m = 1, 2, \dots, M \quad \text{Eq. 10}$$

## 404 2.7 Gridding method

405 Kriging is a regression algorithm to model and predict (interpolate) random processes/fields based on the  
 406 covariance function, which is widely used in geo-statistics (Pebesma, 2004). Ordinary Kriging is the  
 407 earliest and most extensively studied form of Kriging. It is a linear estimation system applicable to any  
 408 intrinsic stationary random field that satisfies the assumption of isotropy. The two key parameters of  
 409 Ordinary Kriging are the semi-variogram function and the weight factors (Goovaerts, 2000). It has been  
 410 widely applied in fields, such as climatology, environmental science, and agriculture (Lapen and Hayhoe,  
 411 2003; Chen et al., 2010), due to high accuracy, stability, and insensitivity to data shape and distribution.  
 412 This study utilizes area-weighted ordinary kriging algorithm to estimate the unknown values of AOD at  
 413 specific locations to generate gridded AOD. The longitude range is between  $-179.5^\circ$  E and  $180^\circ$  E, the  
 414 latitude range is between  $-60^\circ$  N and  $85^\circ$  N, and the spatial resolution is  $0.5^\circ \times 0.5^\circ$ .

## 415 2.8 Evaluation metrics

416 Evaluation metrics, including Root Mean Squared Error (RMSE), Mean Absolute Error (MAE) and

417 Pearson Correlation Coefficient (R), are used to measure the performance and accuracy of the model and  
418 gridded results.

419 
$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \hat{y}_i)^2}$$
 Eq. 11

420 
$$MAE = \frac{1}{n} \sum_{i=1}^n |y_i - \hat{y}_i|$$
 Eq. 12

421 
$$R = \frac{\sum_{i=1}^n (y_i - \bar{y})(\hat{y}_i - \bar{\hat{y}})}{\text{sqrt}(\sum_{i=1}^n (y_i - \bar{y})^2 \sum_{i=1}^n (\hat{y}_i - \bar{\hat{y}})^2)}$$
 Eq. 13

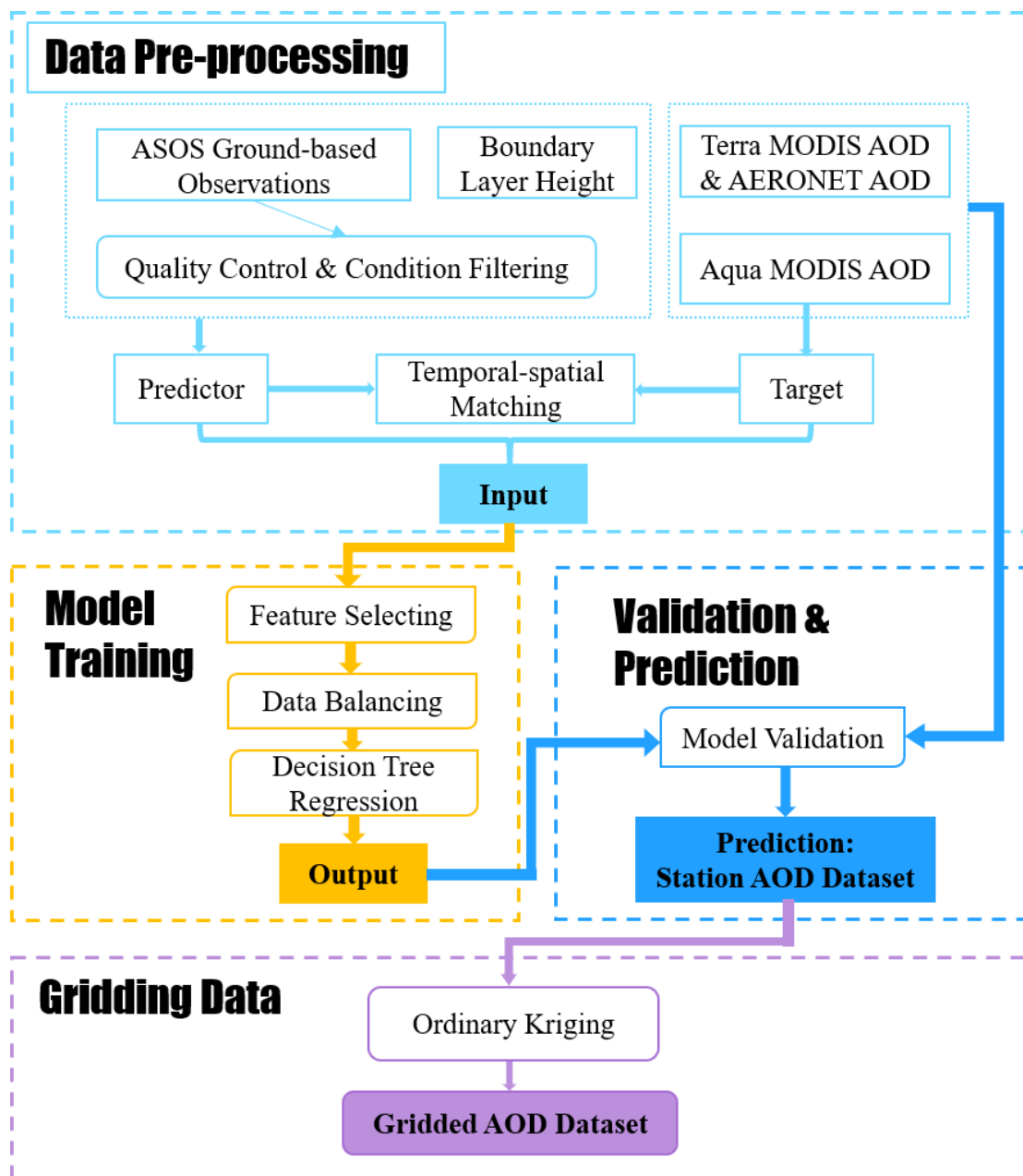
422 where  $y_i$  and  $\bar{y}$  are the predicted value and the average of the predicted values.  $\hat{y}_i$  and  $\bar{\hat{y}}$  are  
423 the target and the average of the target.  $i = 1, 2, \dots, n$ .  $n$  is the length of sample.

424 The expected error (EE) is used to evaluate the AOD derived from visibility.

425 
$$EE = \pm(0.05 + 0.15 * \tau_{target})$$
 Eq. 14

426 where  $\tau_{target}$  is AERONET AOD or Terra MODIS AOD at 550nm.

427



428

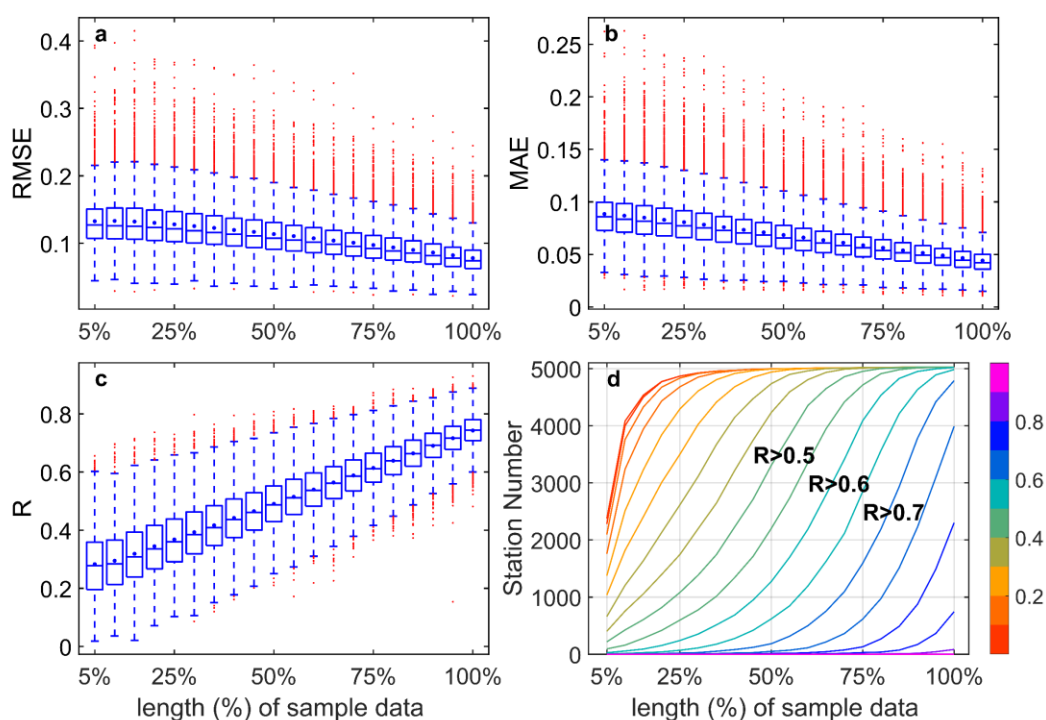
429 **Figure 2** Flowchart for deriving aerosol optical depth (AOD).

430 **2.9 Workflow**

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

434 **3 Results and discussion**

435 **3.1 Dependence of model performance on training data length**



436

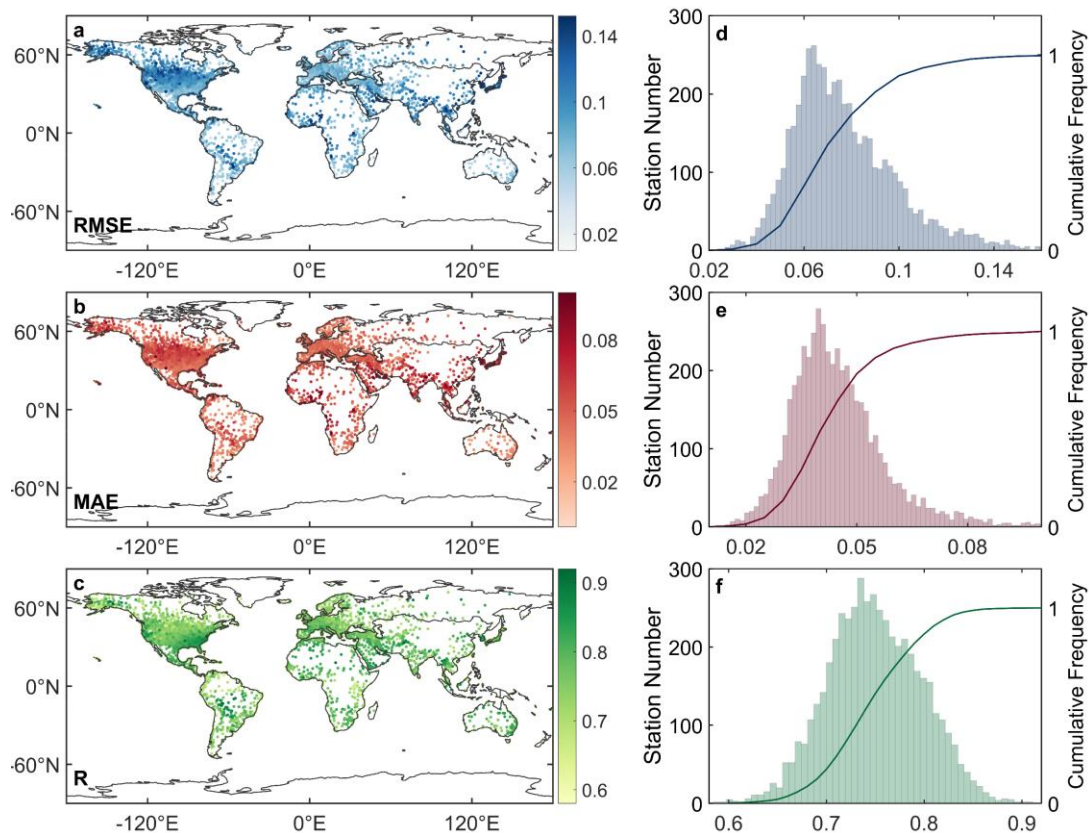
437 **Figure 3** Boxplots of root mean squared error (RMSE) (a), mean absolute error (MAE) (b), and  
 438 correlation coefficient (R) (c) between predicted values and target using different lengths of sample  
 439 data (5% interval) as the training dataset, and the correlation coefficient curve (d)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

441 We build the models using different lengths of sample data (5% to 100%, with a 5% interval) by random  
 442 allocation without overlap and evaluate the predictive performance of each model. Figure 3 depicts  
 443 RMSE(a), MAE(b), and R (c) between the predicted values and target based on the training data of 5%  
 444 to 100% sample data at a station. As the volume of the training data increases, the RMSE and MAE  
 445 decrease, and the correlation coefficient increases. Compared to 5% of the sample data, the result of 100%  
 446 sample data shows a decrease in RMSE by 41.1%, a decrease in MAE by 50.1%, and an increase in R  
 447 by 162.3%. The relationship between the length of sample data and the model's performance is positive  
 448 for each station. Figure 3 (d) shows that R of approximately 70% stations is greater than 0.5 at 50% of  
 449 the sample data, while at 75%, the R of approximately 80% of stations is greater than 0.6. When 100%  
 450 of the sample data is used as sample data, the R of approximately 80% of stations is greater than 0.75,  
 451 and the R of about 97% is greater than 0.7. This finding indicates that the predictive capability and  
 452 robustness of the model increase as the amount of training data increases. It may be attributed to the  
 453 model's ability to capture more complex patterns and relationships among the input by multi-year data.

### 454 3.2 Evaluation of model training

455 Figure 4 shows the spatial distribution (a-c) and frequency and cumulative frequency (d-e) of RMSE,  
 456 MAE, and R of all stations. The mean values of RMSE, MAE, and R are 0.078, 0.044, and 0.750,  
 457 respectively. The RMSE of 93% stations is less than 0.11, the MAE of 91% is less than 0.06, and the R  
 458 of 88% is greater than 0.7. The R values in Africa, Asia, Europe, North America, Oceania, and South  
 459 America are 0.763, 0.758, 0.736, 0.750, 0.759, and 0.738, respectively. Although the RMSE and MAE

460 of a few stations are high in America and Asia, the R is still high (>0.6). Therefore, the results of the  
 461 model's errors demonstrate that the model performs well on almost all stations.



462

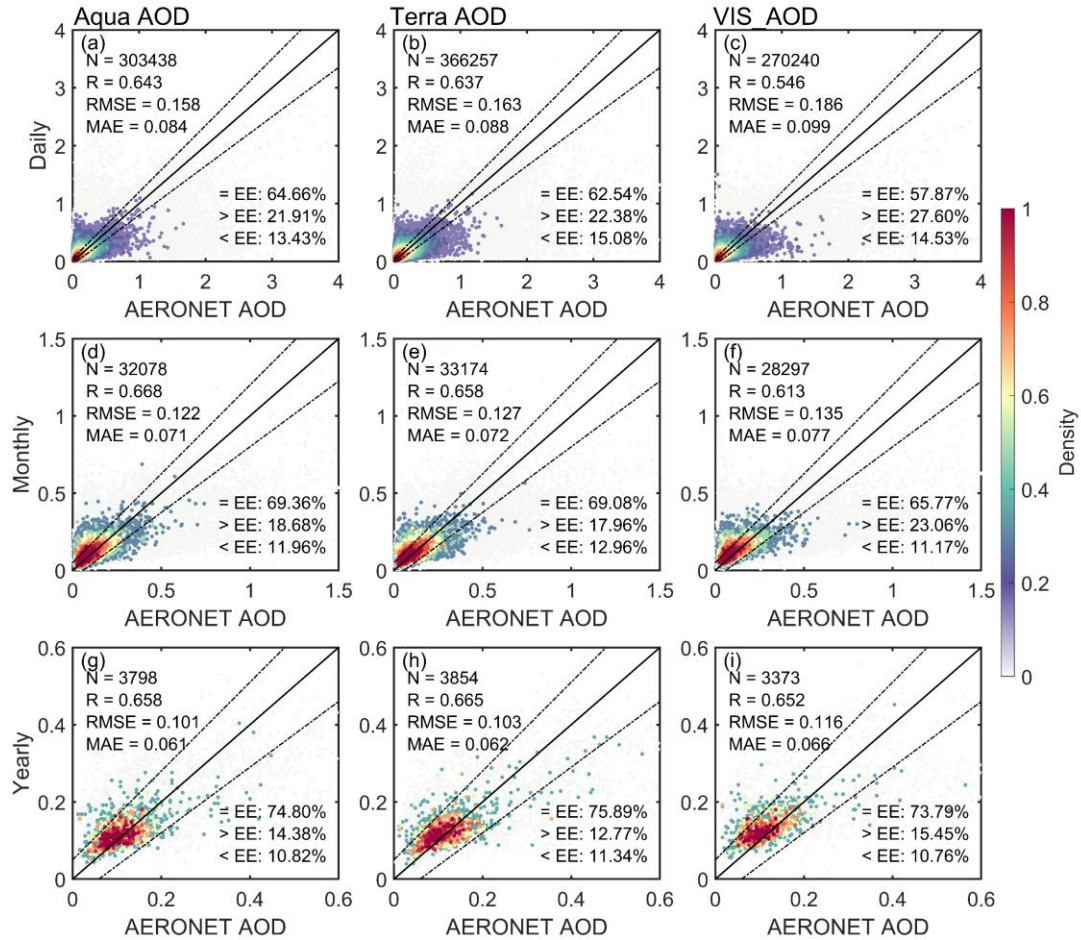
463 **Figure 4** Spatial distribution (a-c) of root mean squared error (RMSE), mean absolute error (MAE),  
 464 and correlation coefficient(R) between the model's result and target with 100% sample data. Station  
 465 number (bar) and cumulative frequency (curve) (d-e) of RMSE, MAE, and R.

### 466 3.3 Validation and comparison with MODIS and AERONET AOD

#### 467 3.3.1 Validation over global land

468 To validate the model's predictive ability, the visibility-derived AOD (for short, VIS\_AOD) is compared  
 469 with Aqua, Terra and AERONET AOD at 550nm for the global scale. Among them, Aqua AOD has been  
 470 used as training data, which is not independent. Terra AOD and AERONET AOD have not been used as  
 471 training data and can be regarded as independent data.

472 First, the relationship among daily MODIS and AERONET AOD is evaluated. Figure 5 shows the scatter  
 473 density plots between AERONET AOD and Aqua AOD (a, d, g) and Terra AOD (b, e, h). The R values  
 474 with Aqua AOD and Terra AOD are 0.643 and 0.637 on the daily scale, and 0.668 and 0.658 on the  
 475 monthly scale, 0.658 and 0.665 on the yearly scale. The RMSE with Aqua AOD and Terra AOD are 0.158  
 476 and 0.163 on the daily scale, and 0.122 and 0.127 on the monthly scale, 0.101 and 0.103 on the yearly  
 477 scale. The MAE values with Aqua AOD and Terra AOD are 0.084 and 0.088 on the daily scale, and 0.071  
 478 and 0.072 on the monthly scale, 0.061 and 0.062 on the yearly scale. The percentages of sample point  
 479 falling within the EE envelopes are 64.66% and 62.54% on the daily scale, and 69.36% and 69.08% on  
 480 the monthly scale, 74.80% and 75.89% on the yearly scale.



481

482 **Figure 5** Scatter density plots between AERONET AOD (550nm) and Aqua MODIS AOD, Terra MODIS  
 483 AOD and VIS\_AOD at the daily (a-c), monthly (d-f) and yearly (g-i) scale. The solid black line represents  
 484 the 1:1 line and the dashed lines represents expected error (EE) envelopes. The sample size (N),  
 485 correlation coefficient (R), mean absolute error (MAE), and root mean square error (RMSE) are given.  
 486 ‘=EE’, ‘>EE’, and ‘< EE’ represent the percentages (%) of retrievals falling within, above, and below  
 487 the EE, respectively. The matching time for Aqua AOD and VIS\_AOD with AERONET AOD is 13.30  
 488 ( $\pm 30$  minutes) at local time, and the matching time between Terra AOD and AERONET AOD is 10.30  
 489 ( $\pm 30$  minutes) at local time.

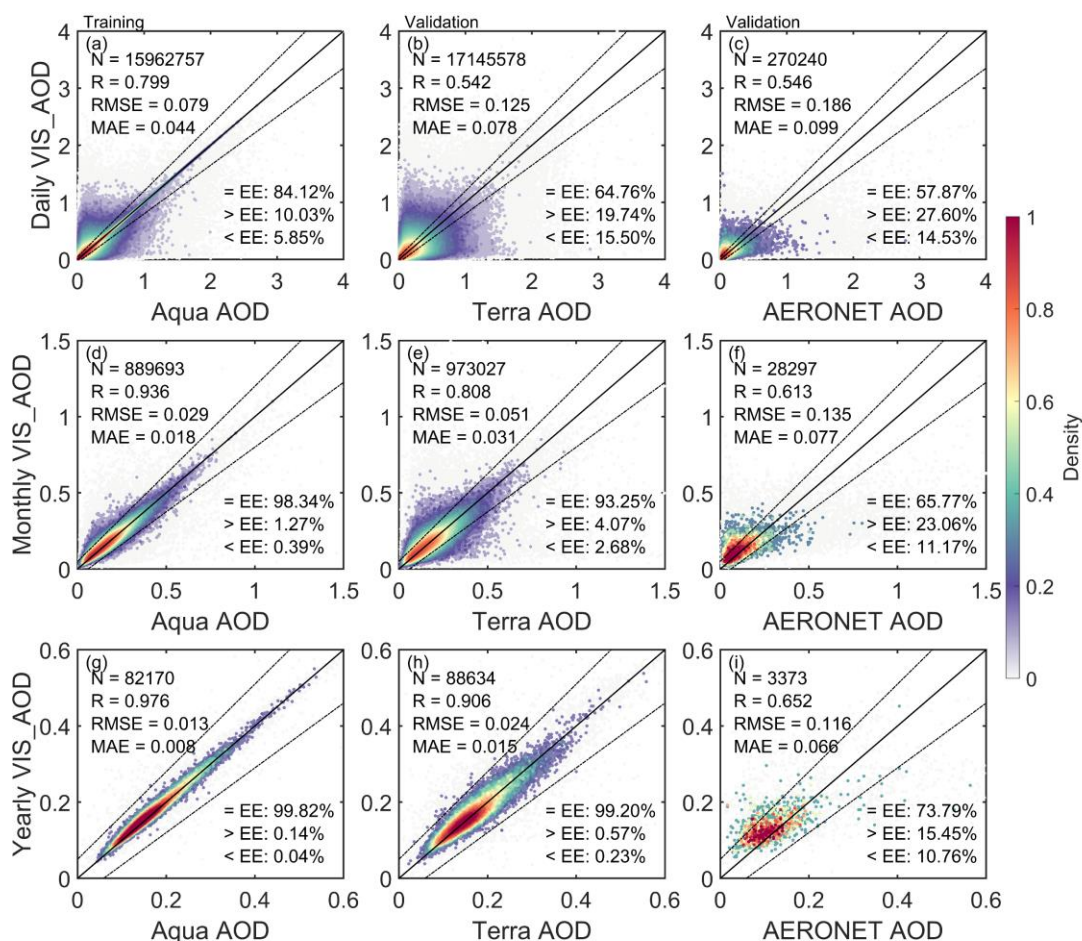
490 Figure 6 shows the scatter density plots and the EEs between VIS\_AOD and Aqua AOD, Terra AOD,  
 491 and AERONET AOD. Aqua AOD is not an independent validation, and Terra and AERONET AOD are  
 492 independent validation. For the daily scale, the R, RMSE and MAE of between VIS\_AOD and Aqua  
 493 AOD (15,962,757 pairs data) is 0.799, 0.079 and 0.044, respectively. The percentage of sample point  
 494 falling within the EE envelopes is 84.12% on the global scale (Figure 6 a). The R between VIS\_AOD  
 495 and Terra AOD (17,145,578 pairs data) is 0.542, with a RMSE of 0.125 and MAE of 0.078. The  
 496 percentage falling within the EE envelopes is 64.76% (Figure 6 b). The R between VIS\_AOD and  
 497 AERONET AOD (270,240 pairs data) at 397 sites is 0.546, with a RMSE of 0.186 and MAE of 0.099.  
 498 The percentage falling within the EE envelopes is 57.87% (Figure 6 c).

499 For the monthly and annual scales, RMSE and MAE show a significant decrease between VIS\_AOD and  
 500 Aqua, Terra, and AERONET AOD, and R and percentages falling within EE show a significant increase



501 in Figure 6 (d-i)错误!未找到引用源。 . The monthly RMSEs are 0.029, 0.051, and 0.135, the monthly  
 502 MAEs are 0.018, 0.031, and 0.077, and the R values are 0.936, 0.808, and 0.613, respectively. The  
 503 percentages falling within the EE envelopes are 98.34%, 93.25%, and 65.77%. The RMSEs at the annual  
 504 scale are 0.013, 0.024, and 0.116, the MAEs are 0.008, 0.015, and 0.066, and the R values are 0.976,  
 505 0.906, and 0.652, respectively. The percentages falling within the EE envelopes are 99.82%, 99.20%,  
 506 and 73.79%. The percentage falling within the EE envelopes against AERONET is smaller than that  
 507 against Terra, which may be related to the elevation of AERONET sites, the distance between AERONET  
 508 and meteorological stations, and observed time. The results highlighted above demonstrate a clear  
 509 improvement in performance on the monthly and annual scales compared to the daily scale (Schutgens  
 510 et al., 2017), which provided a foundation for the gridded dataset.

511 On the daily, monthly, and annual scales, compared with AERONET AOD, the correlation coefficients,  
 512 RMSE, MAE, and percentages falling within the expected error of VIS\_AOD and MODIS AOD are very  
 513 close. Since the time of AERONET AOD and VIS\_AOD overlaps before 2000, it indicates that  
 514 VIS\_AOD also has the same accuracy.



515  
 516 **Figure 6** Scatter density plots between predicted AOD (VIS\_AOD) and Aqua MODIS AOD, Terra  
 517 MODIS AOD and AERONET AOD at the daily (a-c), monthly (d-f) and yearly (g-i) scale. The solid  
 518 black line represents the 1:1 line and the dashed lines represents expected error (EE) envelopes. The  
 519 sample size (N), correlation coefficient (R), mean absolute error (MAE), and root mean square error  
 520 (RMSE) are given. ‘=EE’, ‘>EE’, and ‘< EE’ represent the percentages (%) of retrievals falling  
 521 above, and below the EE, respectively. Note Aqua AOD is not an independent validation for predicted

522 results, while Terra and AERONET are independent validation.

### 523 **3.3.2 Validation over regions**

524 Aerosol loading exhibits spatial variability. Evaluation metrics for the relationships between  
525 visibility-derived AOD and AERONET AOD and Terra AOD for each region are listed in Table 1.  
526 Over Europe and North America, the results are similar to those of Terra and AERONET, with a  
527 large number of data pairs, greater than  $10^5$  (AERONET) and greater than  $10^7$  except for Eastern  
528 Europe (Terra) on the daily scale. Approximately 63% -70% fall within the EE envelopes. The  
529 RMSE is approximately 0.1100, except for western North America, the MAE is approximately  
530 0.0700, with a correlation coefficient between 0.44 and 0.54.

531 Over Central South America, South Africa, and Australia, data pairs are about  $10^{3-4}$  (AERONET)  
532 and  $10^6$  (Terra) on the daily scale. 52-60% fall within the EE envelopes compared to AERONET,  
533 and 58-67% compared to Terra. The RMSE is 0.03-0.05 compared to Terra, and 0.11-0.17 compared  
534 to AERONET. The correlation coefficient ranges from 0.4 to 0.74, with the highest correlation  
535 coefficient in South America at 0.740.

536 In Asia, India, and West Africa, the data pairs are only approximately  $10^4$  (AERONET). 32% to 50%  
537 fall within the EE envelopes compared to AERONET, the RMSE ranges from 0.2 to 0.5, and the  
538 MAE ranges from 0.11 to 0.36. 51 to 58%, compared to Terra, fall within the EE envelopes, the  
539 RMSE is around 0.16, and the MAE is around 0.11. Compared to AERONET, in these high aerosol  
540 loading regions, RMSE and MAE increase, and the percentages falling within the EE envelopes  
541 decrease, but the correlation coefficients do not significantly decrease.

542 Compared to Terra AOD, 55% -67% of data falls within the EE envelopes on the daily scale, 87% -  
543 96% on the monthly scale, and over 97% on the yearly scale. Compared to AERONET AOD, 32-  
544 68% of data falls within the EE envelopes, 24% -84% on the monthly scale, and 15% -97% on the  
545 yearly scale. On both monthly and yearly scales, all metrics have shown a significant increase in  
546 performance when compared to Terra. However, compared to AERONET, not all metrics increase  
547 in some regions due to limited data pairs, such as West Africa, Northeast Asia, and India, which may  
548 be due to the spatial differences between AERONET sites and meteorological stations.

549 Overall, the AOD from visibility is more effective in regions such as Europe and North America,  
550 which may also be related to the better performance of the MODIS DT algorithm in vegetation-  
551 covered regions. In high aerosol load areas affected by deserts, such as Africa and Asia, the AOD of  
552 visibility inversion needs to be improved.

### 553 **3.3.3 Validation at a site scale**

554 Sites, especially AERONET, are not completely uniform across the world or in any region, and  
555 different stations have different sample sizes, which may lead to a certain uncertainty. Therefore,  
556 further analysis was conducted on the spatial distribution of different evaluation metrics. Figure 7  
557 shows the validation and comparison of daily VIS\_AOD against Terra and AERONET AOD at a  
558 site scale.

559 Compared to Terra daily AOD, the R of 67% stations is greater than 0.4, the mean bias of 83% is

**Table 1** Evaluation metrics for the relationships between visibility-derived AOD and AERONET AOD and Terra AOD for each region.

Region		N			R			RMSE			MAE			Within EE (%)		
		daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly
Eastern Europe	AERONET	21724	2317	271	0.463	0.493	0.653	0.1069	0.0647	0.0326	0.0714	0.0442	0.0263	65.69	83.77	97.42
	TERRA	661630	36435	3278	0.464	0.665	0.790	0.1095	0.0471	0.0214	0.0726	0.0286	0.0122	66.07	94.71	99.18
Western Europe	AERONET	53043	6033	697	0.445	0.487	0.344	0.1089	0.0716	0.0513	0.0711	0.0474	0.0347	64.40	79.21	89.10
	TERRA	1778013	104620	9166	0.467	0.763	0.811	0.1096	0.0391	0.0210	0.0712	0.0268	0.0124	66.99	95.42	99.40
Western North America	AERONET	33859	2948	334	0.503	0.484	0.509	0.1465	0.0949	0.0566	0.0747	0.0597	0.0419	63.58	67.37	81.14
	TERRA	1725226	82734	7201	0.542	0.765	0.906	0.1144	0.0465	0.0180	0.0671	0.0267	0.0125	69.48	94.42	99.61
Eastern North America	AERONET	47407	5359	608	0.527	0.526	0.559	0.1135	0.0824	0.0436	0.0657	0.0472	0.0331	67.52	77.78	87.50
	TERRA	6280277	359520	31343	0.515	0.799	0.847	0.1159	0.0435	0.0165	0.0726	0.0275	0.0111	66.70	94.94	99.80
Central South America	AERONET	10911	1176	149	0.740	0.811	0.866	0.1735	0.1272	0.1060	0.1021	0.0904	0.0688	52.40	47.96	67.79
	TERRA	444780	26362	2410	0.545	0.820	0.776	0.1447	0.0591	0.0369	0.0909	0.0396	0.0219	58.48	89.29	97.39
Southern Africa	AERONET	4255	309	38	0.423	0.480	0.630	0.1553	0.1128	0.0705	0.1033	0.0805	0.0525	52.08	59.55	78.95
	TERRA	216239	11304	1118	0.518	0.821	0.870	0.1258	0.0511	0.0296	0.0836	0.0340	0.0191	60.64	91.70	98.21
Australia	AERONET	6426	516	63	0.488	0.654	0.363	0.1094	0.0827	0.0725	0.0711	0.0620	0.0563	59.96	59.88	71.43
	TERRA	284693	14588	1286	0.398	0.784	0.831	0.1091	0.0363	0.0188	0.0666	0.0261	0.0143	67.01	94.65	99.38
Western Africa	AERONET	2205	205	34	0.553	0.594	0.762	0.3180	0.2873	0.3357	0.2082	0.2029	0.2587	37.96	40.00	23.53
	TERRA	156392	10468	1028	0.501	0.769	0.849	0.1769	0.0706	0.0412	0.1198	0.0482	0.0242	51.83	88.01	97.57
Southeast Asia	AERONET	4134	504	74	0.405	0.542	0.488	0.2037	0.1447	0.1198	0.1274	0.0988	0.0821	50.17	56.15	60.81
	TERRA	402465	27058	2500	0.470	0.753	0.872	0.1730	0.0729	0.0342	0.109	0.0455	0.0198	57.25	87.01	97.96
Eastern China	AERONET	7396	927	118	0.513	0.551	0.356	0.3571	0.2355	0.1933	0.2038	0.1392	0.1382	40.10	49.84	50.00
	TERRA	241185	17324	1518	0.523	0.811	0.895	0.1646	0.0638	0.0302	0.1073	0.0435	0.0225	55.77	88.07	98.88
Northeast Asia	AERONET	9979	1178	142	0.569	0.593	0.367	0.4941	0.3249	0.2604	0.2924	0.2425	0.2202	35.17	29.54	21.13
	TERRA	78823	5485	467	0.553	0.872	0.965	0.1973	0.0636	0.0263	0.1201	0.0440	0.0198	56.48	87.77	98.29

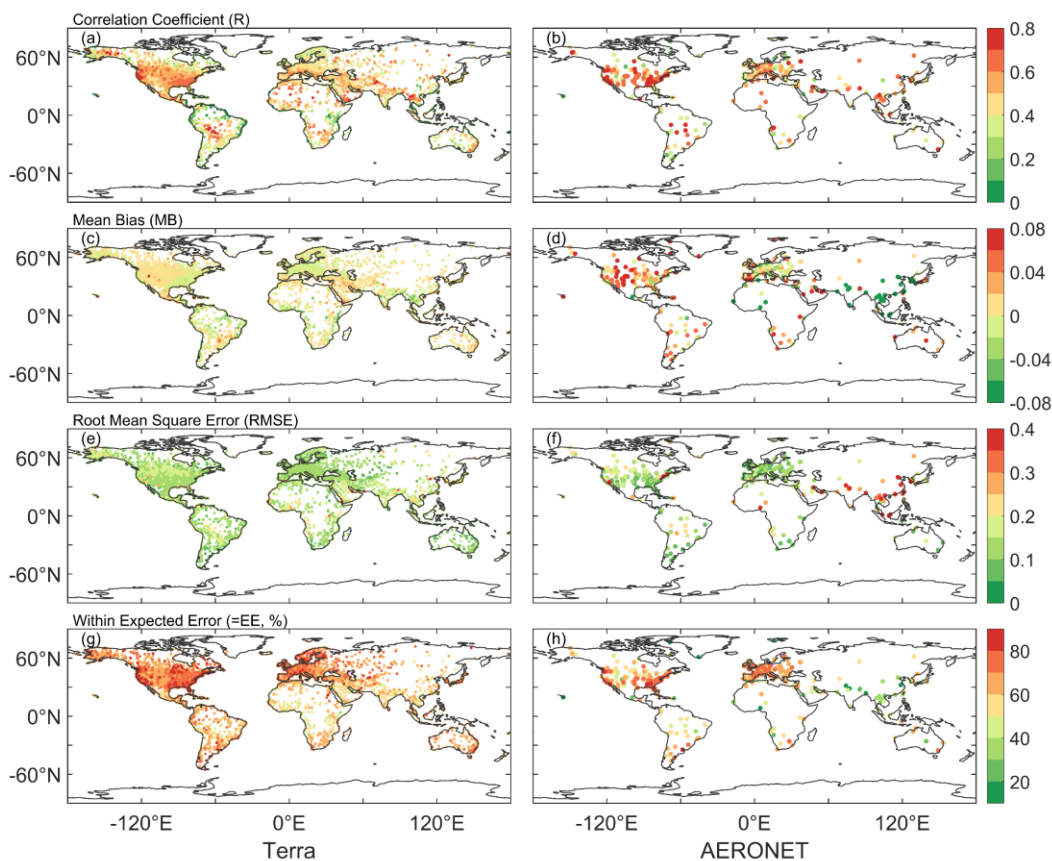
<i>India</i>	<i>AERONET</i>	2208	203	32	0.521	0.462	0.534	0.2957	0.3015	0.3588	0.2049	0.2283	0.2862	32.11	24.63	15.63
	<i>TERRA</i>	179928	9564	862	0.526	0.815	0.915	0.1564	0.0599	0.0352	0.1089	0.042	0.0238	55.16	90.43	98.14

561

562 less than 0.01, the RMSE of 85% is less than 0.15, and the percentage falling within the EE of 67%  
 563 is greater than 60%. More than 85% of stations fall within the EE is greater than 60% in Europe,  
 564 North America, and Oceania, while 40-60% in South America, Africa, and Asia. The percentage of  
 565 expected error is low in South and East Asia, and Central Africa, with some underestimation. Above  
 566 60% in Africa, Asia, North America, and Europe have a correlation coefficient greater than 0.4. The  
 567 regions with lower correlation are the coastal regions of South America, eastern Africa, western  
 568 Australia, northeastern North America, and northern Europe. Above 90% of the RMSE in Europe,  
 569 North America, and Oceania have a correlation coefficient smaller than 0.15. High RMSE regions  
 570 are in western North America, Asia, central South America, and central Africa.

571 Compared to AERONT daily AOD, the R of 74% stations is greater than 0.4, and the spatial  
 572 distribution is similar to Terra's. The mean bias of 44% is less than 0.01, the RMSE of 68% is less  
 573 than 0.15, and the percentage falling within the EE of 53% is greater than 60%. More than 70% of  
 574 sites have a correlation coefficient greater than 0.4 in Africa, Asia, Europe, and North America.  
 575 More than 57% of sites have an expected error percentage of over 60% in Europe, North America,  
 576 and Oceania. Except for Asia. Over 72% of sites have a RMSE less than 0.15. Except for Oceania  
 577 and South America, over 71% of sites in other regions have MAE less than 0.01. Almost all sites in  
 578 Asia show a negative bias, significantly underestimating. However, there is a significant  
 579 overestimation in western North America and western Australia. Most sites in Asia falling within  
 580 the expected error are less than 50%. High RMSE region are in Asia, India, and central Africa.

581 The validation and comparison on the site scale show a limitation similar to the MODIS DT  
 582 algorithm. In areas with high vegetation coverage, the AOD from visibility are better than those in  
 583 bright areas such as deserts.



584

585 **Figure 7** Validation of VIS\_AOD against Terra and AERONET AODs at each site: (a–b) correlation  
586 (R), (c–d) mean bias (MB), (e–f) root mean square error (RMSE), (g–h) percentage (%) of VIS\_AOD  
587 within the expected error envelopes.

### 588 **3.3.4 Discussion and uncertainty analysis**

589 The atmospheric visibility is a horizontal physical quantity, while AOD is a column-integrated  
590 physical quantity. We have linked the two variables together using machine learning methods, which  
591 partially compensates for the scarcity of AOD data. However, we have to face some limitations.  
592 Although the boundary layer height is considered, it is not sufficient. Pollutants such as smoke from  
593 biomass burning, dust, volcanic ash, and gas-aerosol conversion of sulfur dioxide to sulfate aerosols  
594 in the upper and lower troposphere can undergo long-range aerosol transport under the influence of  
595 circulation. The pollution transport and aerosol conversion processes above the boundary layer are  
596 still significant and cannot be ignored (Eck et al., 2023). Compared to surface visibility, bias occurs  
597 when the aerosol layer rises and affects AERONET measurements and MODIS retrievals. Therefore,  
598 it should be considered when using this data. If there were sufficient historical vertical aerosol  
599 measurements with high temporal and spatial resolution, the results of this data would be greatly  
600 improved. Although some studies use aerosol profiles from pollution transport models or assumed  
601 profiles as substitutes for observed profiles (Li et al., 2020; Zhang et al., 2020), the biases introduced  
602 by these non-observed profiles are still significant.

603 In machine learning, we used MODIS Aqua AOD as the target value for the model because the  
604 validation results for MODIS C6.1 products have a correlation coefficient of 0.9 or higher with  
605 AERONET AOD at the daily scale (Wei et al., 2019; Wei et al., 2020). Compared to AERONET,  
606 MODIS AOD provides more sample data with a high global coverage. However, apart from  
607 modeling errors, the systematic biases and uncertainties of MODIS Aqua AOD cannot be ignored  
608 (Levy et al., 2013; Levy et al., 2018; Wei et al., 2019). Averaging over time scale significantly  
609 reduces systematic errors but cannot diminish errors caused by emission sources and terrain.  
610 Therefore, the strong correlation at monthly and annual scales indicates a substantial reduction in  
611 errors (Schutgens et al., 2017). This is also one of the reasons why this dataset shows stronger  
612 correlation with Terra AOD and weaker correlation with AERONET in validation.

613 The spatial matching between meteorological stations and AERONET sites may cause some biases.  
614 AERONET sites are usually not co-located with meteorological stations in terms of elevation and  
615 horizontal distance, this is another reason for the weak correlation between VIS\_AOD and  
616 AERONET AOD. The meteorological stations are located at the airport. Different horizontal  
617 distances may result in meteorological stations and AERONET sites being located on different  
618 surfaces (such as urban, forest, mountainous). Differences in site elevation significantly impact the  
619 relationship between AOD and measured visibility. When the AERONET site is at a higher elevation  
620 than the meteorological station, there may be fewer measurements of aerosols over the sea at the  
621 AERONET site.

622 Different pollution levels and station elevation affect the AOD derived from visibility. The elevation  
623 difference and distance between meteorological stations and AERONET sites also have an impact  
624 on the validation results. Therefore, the error and performance of different AERONET AOD values,  
625 station elevation, and distance were analyzed.

#### 626 **3.3.4.1 Uncertainty with pollution level**

627 As the AOD increases, the variability of bias also increases in Figure 8 (a). Almost all mean bias  
628 values are within the envelope of EE, except for 1.1-1.2 and 1.5-1.6. The average bias is 0.015  
629 (AOD <0.1), with 83% of data within the EE envelopes. The mean bias is -0.0011 (AOD,0.1-0.2),  
630 with 54% within the EE envelopes. The mean bias is negative (AOD, 0.3-1.0), with 20%-40%  
631 falling within the EE envelopes. There is a positive bias (AOD, 1.1, 1.4 and >1.6), and there is a  
632 negative bias at 1.2-1.3 and 1.5-1.6. The results indicate that as pollution level increases, the  
633 negative mean bias becomes significant and the underestimation increases.

#### 634 **3.3.4.2 Uncertainty with elevation of AERONET site**

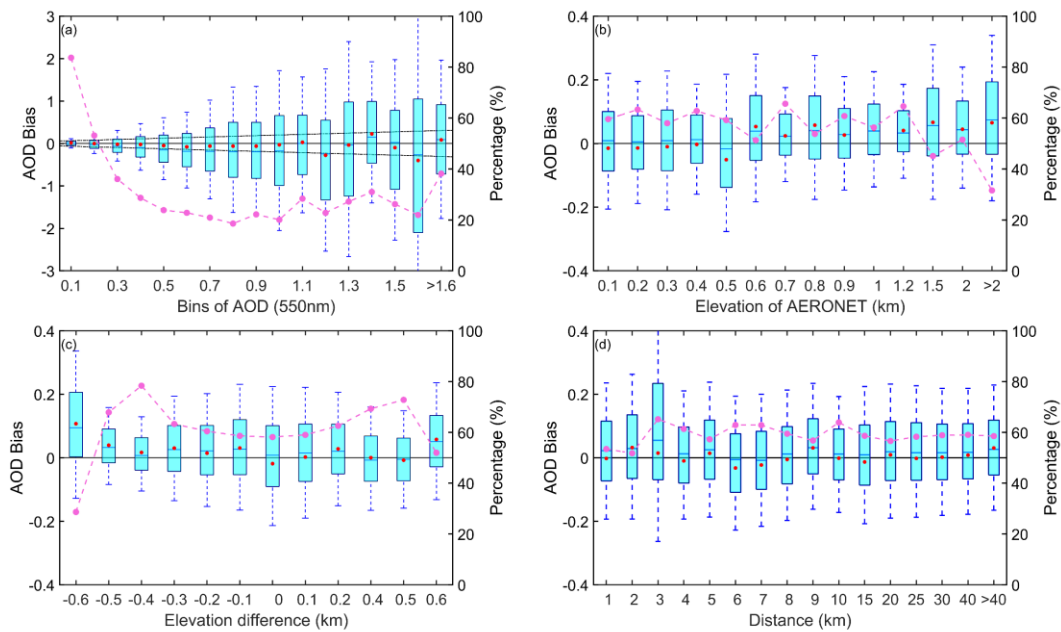
635 The contribution of particulate matter near the ground to the column aerosol loading is significant.  
636 The elevation of the site affects the measurement of column aerosol loading in Figure 8 (b). There  
637 is a negative bias in the low elevation ( $\leq 0.5$ km) with a percentage of 60%-64% falling within the  
638 EE envelopes and a positive bias in high elevation (0.5-1.2km) with a percentage of 50%-65%  
639 falling within the EE envelopes. The percentage significantly decreases ( $>1.2$ km), and the average  
640 bias increases. Therefore, the elevation of AERONET's site will cause bias in validation, and the  
641 uncertainty greatly increases in high elevation.

#### 642 **3.3.4.3 Uncertainty with elevation of meteorological station**

643 Due to the elevation difference between the meteorological station and AERONET site in the  
644 vertical direction, the uncertainty caused by elevation differences of site was analyzed in Figure 8  
645 (c). When the elevation difference is negative (the elevation of the meteorological station is lower  
646 than that of the AERONET station), there is a significant positive bias. When the difference is  
647 positive, the mean bias approaches 0 or is positive. The percentage is greater than 60% (-0.5 km-  
648 0.5km). The positive mean bias is greater than the negative mean bias, and the uncertainty greatly  
649 increases when the elevation of meteorological stations is lower than that of AERONET sites. It  
650 indicates that the contribution of the near surface aerosol to the column aerosol loading is significant  
651 and cannot be ignored.

#### 652 **3.3.4.4 Uncertainty with distance between meteorological station and AERONET site**

653 The spatial variability of aerosols is significant. Meteorological stations and AERONET sites are  
654 not collocated, resulting in a certain distance in spatial matching. In this study, the upper limit of  
655 distance is 0.5 degree. Figure 8 (d) shows the error of the distance between stations, where the  
656 degree is converted to the distance at WGS84 coordinates. The bias does not change significantly  
657 with increasing distance. The average bias is around 0, with the maximum positive mean bias  
658 (0.0322) at a distance of 2km and the maximum negative mean deviation (-0.0323) at 6km. The  
659 median is almost positive, except at 5km and 6km. The percentage falling within the EE envelopes  
660 is over 50%, with the maximum percentage (66%) at 3km and the minimum (62%) at 2km.



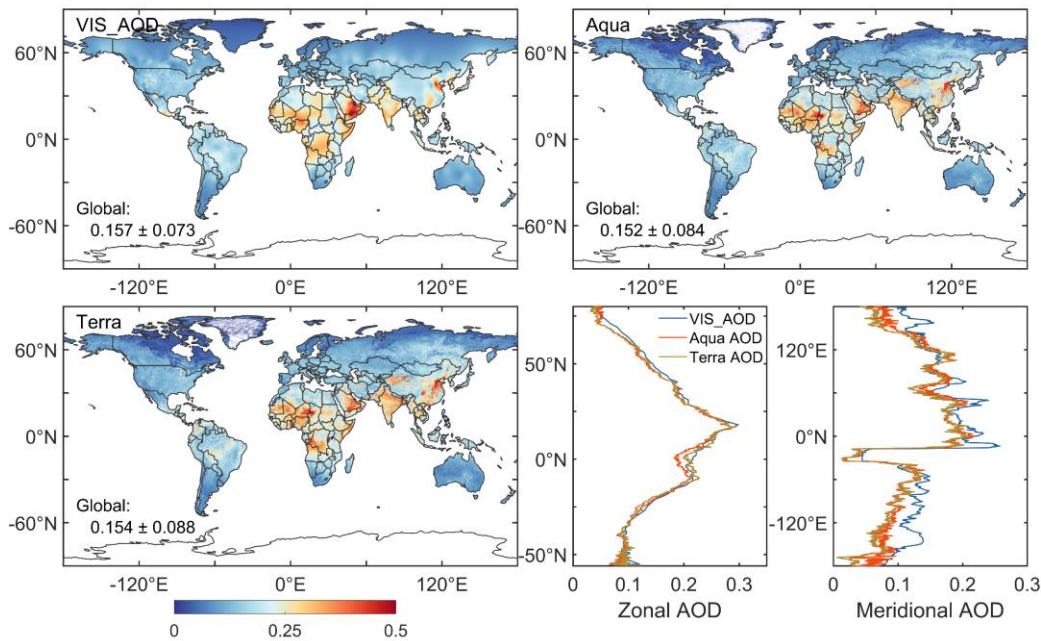
661

662 **Figure 8** Box plots of AOD bias and the percentage falling within the EE envelopes (curves): (a)  
 663 AERONET AOD levels, (b) elevation of AERONET sites, (c) elevation difference between  
 664 meteorological stations and AERONET sites, (d) distance (km) between meteorological stations and  
 665 AERONET sites. The black horizontal line represents the zero bias. For each box, the upper, lower,  
 666 and middle horizontal lines, and whiskers represent the AOD bias 75th and 25th percentiles, median,  
 667 and 1.5 times the interquartile difference, respectively. The black solid lines represent the EE  
 668 envelopes ( $\pm(0.05+0.15*AOD_{AERONET})$ ). No site with a difference of +0.3km (x-axis label without  
 669 0.3) in (c).

### 670 3.4 Gridded visibility-derived AOD

671 Figure 9 shows the gridded AOD based on ordinary kriging interpolation with the area-weighted  
 672 method and compares the multi-year spatial, zonal, and meridional distributions of AOD with Aqua  
 673 and Terra AOD over land from 2003 to 2021. The VIS\_AOD is  $0.157\pm 0.073$  over land, which is  
 674 almost equal to the Aqua ( $0.152\pm 0.084$ ) and Terra ( $0.154\pm 0.088$ ) AOD values with relative biases  
 675 of 3.3%, and 1.9%, respectively. In order to compare the spatial correlation, Aqua and Terra MODIS  
 676 AOD are averaged to the 0.5-degree resolution. In the heatmap (Figure 10), the R of VIS\_AOD and  
 677 Aqua AOD is 0.798, the RMSE is 0.049 with a bias of 32% compared to the mean, and the MAE is  
 678 0.008, with a bias of 5% compared to the mean. Compared to Terra AOD, the R is 0.787, and the  
 679 RMSE is 0.051, with a bias of 33% compared to the mean, and the MAE is 0.005, with a bias of 3%  
 680 compared to the mean. The R between Aqua and Terra AOD is 0.980. The R values between  
 681 VIS\_AOD and Aqua and Terra AOD are 0.995 and 0.990 for the zonal distribution and 0.986 and  
 682 0.897 for the meridional distribution, respectively. In the low aerosol loading region, VIS\_AOD  
 683 exhibits a little overestimation. Whether in meridional or zonal distribution, the peak and valley  
 684 regions are basically consistent (Tian et al., 2023). Due to the limitations of satellite inversion  
 685 algorithms, a bias appears on the bright surface, especially in northern North America with extensive  
 686 snow cover (Levy et al., 2013). All above results suggest that the gridded AOD is consistent with  
 687 satellite observations in spatial distribution.

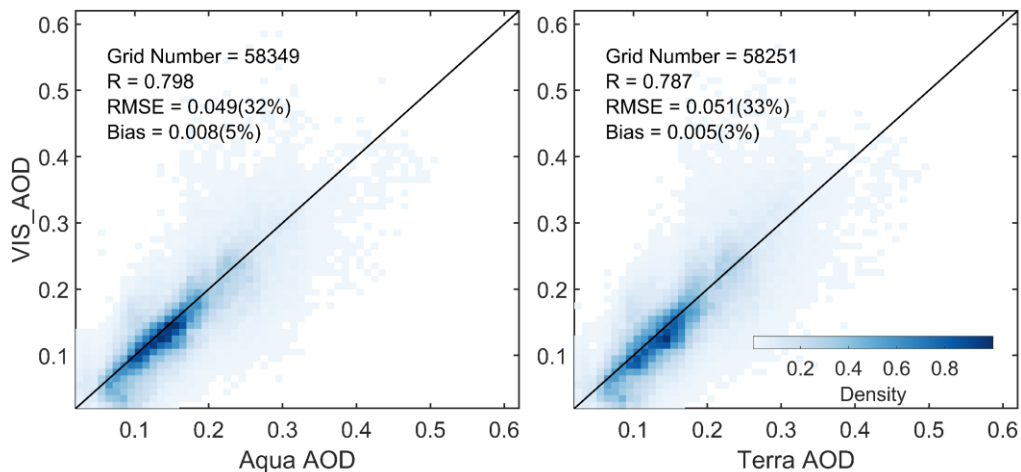




689

690 **Figure 9** The spatial, zonal and meridional distributions of the multi-year mean VIS\_AOD, Aqua  
 691 AOD, and Terra AOD over land from 2003 to 2021.

692



693

694 **Figure 10** Heatmap of multi-year mean gridded VIS\_AOD and Aqua AOD and Terra AOD during  
 695 2003-2021. Terra and Aqua AOD are averaged onto a grid of 0.5°.

### 696 3.5 Interannual variability and trend of visibility-derived AOD over global land

697 The spatial distribution of multi-year average AOD from 1980 to 2021 over land is shown in Figure  
 698 11 (a). The mean AOD of land (-60-85°N), Northern Hemisphere (NH, 0-85°N), and the Southern  
 699 Hemispheres (SH, -60-0°N) is  $0.161 \pm 0.074$ ,  $0.158 \pm 0.076$ , and  $0.173 \pm 0.059$ , respectively. The  
 700 AOD values of Africa, Asia, Europe, North America, Oceania, and South America are 0.241, 0.222,  
 701 0.110, 0.111, 0.129 and 0.117, respectively.

702 Due to the influence of geography, atmospheric circulation, population, and emissions, the AOD  
703 varies in different latitudes. Figure 12 illustrates the multi-year average AOD in different latitude  
704 ranges for land, the NH, and the SH from 1980 to 2021. Within  $[-20, 20^{\circ}\text{N}]$ , the global average AOD  
705 reaches its maximum (0.234), and the maximum AOD NH is 0.256 in  $[0, 20^{\circ}\text{N}]$ . The highest AOD  
706 in SH is 0.217 in  $[-15, 0^{\circ}\text{N}]$ . The average AOD in SH rapidly decreases from  $-15^{\circ}\text{N}$  to  $-35^{\circ}\text{N}$ . In  
707 NH, AOD is generally greater than in SH from  $5^{\circ}\text{N}$  to  $65^{\circ}\text{N}$ . When, the latitude is greater than  $70^{\circ}\text{N}$ ,  
708 the NH's AOD is smaller than the SH's.

709 There are many regions of high AOD values occur in NH, with the distribution of population density.  
710 Approximately 7/8 of the global population resides in the NH, with 50% concentrated at  $20^{\circ}\text{N}$ - $40^{\circ}\text{N}$   
711 (Kummu et al., 2016), indicating a significant impact of human activities on aerosols. The highest  
712 AOD values are observed near  $17^{\circ}\text{N}$ , including the Sahara Desert, Arabian Peninsula, and  
713 southeastern India, suggesting that in addition to anthropogenic sources, deserts also play a crucial  
714 role in aerosol emissions. Lower AOD regions of the SH are from  $25^{\circ}\text{S}$  to  $60^{\circ}\text{S}$ , encompassing  
715 Australia, southern Africa, and southern South America, indicating lower aerosol burdens in these  
716 areas. Additionally, North America also exhibits low aerosol loading. Chin et al. (2014) analyzed  
717 the AOD over land from 1980 to 2009 with the Goddard Chemistry Aerosol Radiation and Transport  
718 model, which is similar to the visibility-derived AOD. The spatial distribution is consistent with the  
719 satellite results (Remer et al., 2008; Hsu et al., 2012; Hsu et al., 2017; Tian et al., 2023). The AOD  
720 and extinction coefficient retrieved from visibility show a similar distribution at global scale, with  
721 a correlation coefficient of nearly 0.6 (Mahowald et al., 2007). Similar global (Husar et al., 2000;  
722 Wang et al., 2009) and regional (Koelemeijer et al., 2006; Wu et al., 2014; Boers et al., 2015; Zhang  
723 et al., 2017; Zhang et al., 2020) spatial distributions have been reported.

724 AOD loadings exhibit significant seasonal variations worldwide, particularly over land. In this study,  
725 a year is divided into four parts: December-January-February (DJF), March-April-May (MAM),  
726 June-July-August (JJA), and September-October-November (SON), corresponding to winter  
727 (summer), spring (autumn), summer (winter), and autumn (spring) in NH (SH), respectively. Figure  
728 11 (b-e) also depicts the spatial distribution of seasonal average AOD over land from 1980 to 2021.  
729 The global AOD in DJF, MAM, JJA, and SON is  $0.158\pm 0.062$ ,  $0.162\pm 0.081$ ,  $0.175\pm 0.093$ , and  
730  $0.153\pm 0.070$ , respectively. The standard bias of AOD in JJA and MAM are greater than those in  
731 DJF and SON. AOD exhibits seasonal changes, with the highest in JJA, followed by MAM, DJF,  
732 and SON. From 1980 to 2021, the seasonal AOD in NH is  $0.152\pm 0.064$  (DJF),  $0.161\pm 0.088$  (MAM),  
733  $0.176\pm 0.090$  (JJA), and  $0.144\pm 0.060$  (SON), and in SH is  $0.184\pm 0.041$  (DJF),  $0.166\pm 0.044$  (MAM),  
734  $0.169\pm 0.072$  (JJA), and  $0.19\pm 0.060$  (SON).

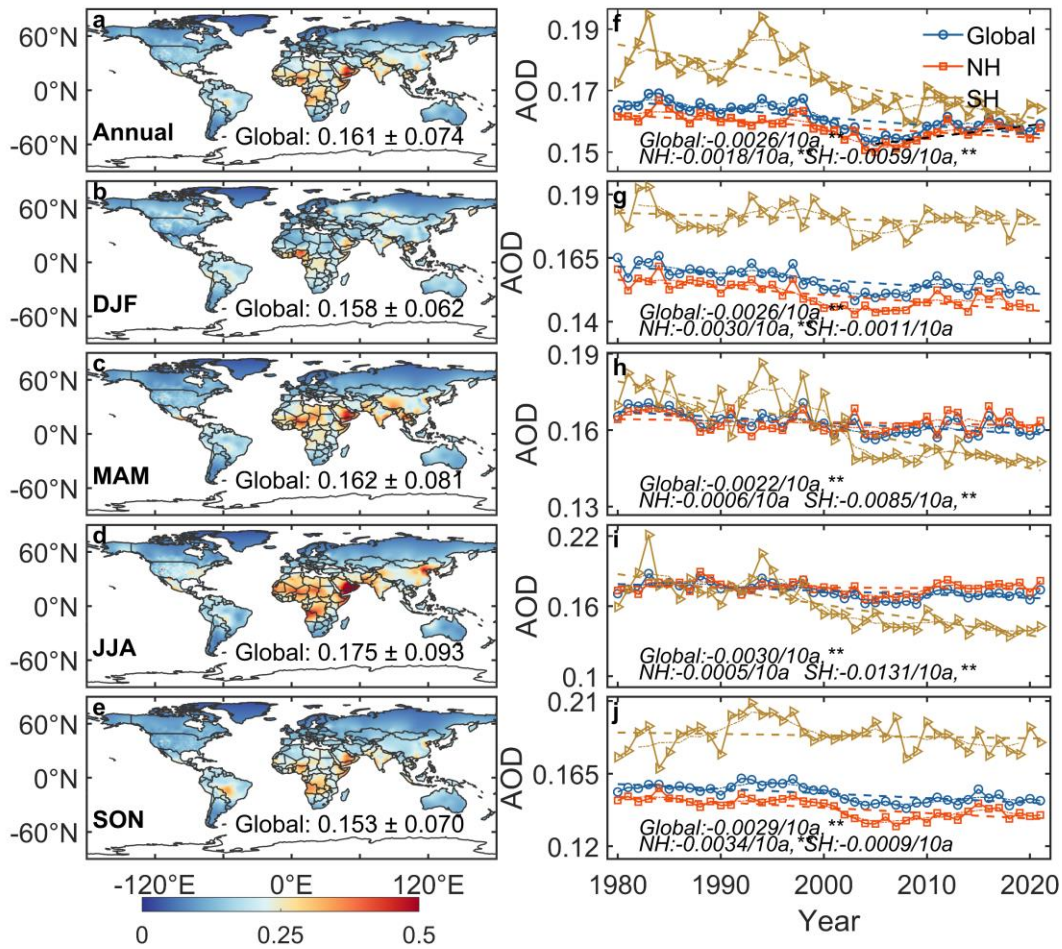
735 In NH, the AOD ranking from high to low in season is summer > spring > winter > autumn. In SH,  
736 the AOD ranking from high to low in season is spring > summer > winter > autumn. The highest  
737 AOD is observed during JJA in NH, while in SH, the peak occurs during SON. The occurrence of  
738 high AOD values is highly associated with the growth of hygroscopic particle and the photochemical  
739 reaction of aerosol precursors under higher relative humidity in Asia (JJA) (Remer et al., 2008) and  
740 Europe such as Russia (JJA), and biomass burning in South America (SON), Southern Africa (SON),  
741 and Indonesia (SON) (Ivanova et al., 2010; Krylov et al., 2014). On the other hand, the lowest global  
742 AOD values are observed during autumn, which may be attributed to the weakening of monsoon  
743 systems (Li et al., 2016; Zhao et al., 2019).

744 In addition to the spatial characteristics of AOD, the temporal variations in AOD have also been of  
745 great interest due to the significant relationship between aerosols and climate change. Figure 11 (f)  
746 shows the temporal trends of annual average AOD (\*\* represents passing the significance test,  
747  $p < 0.01$ ) over the global land, the SH and the NH during 1980-2021. The global land, NH, and SH  
748 trends demonstrate decreasing trends of AOD with values of  $-0.0026/10a$ ,  $-0.0018/10a$ , and  $-$   
749  $0.0059/10a$ , respectively, with all passing the significance test with a confidence level of 95%.  
750 Notably, the declining trend is much greater in the SH than in the NH. It may be related to the  
751 decrease in the frequency of sandstorms and wildfires and the increase in precipitation, such as in  
752 Australia. Two AOD peaks in 1983 and 1994 and two AOD valleys in 1980 and 1990 are observed  
753 before 2000. The two AOD peaks may be attributed to large volcanic eruptions, which has been  
754 confirmed by previous studies. The volcanic eruptions and their associated fires of the El Chichón  
755 volcano in Mexico in 1982 (Hirono and Shibata, 1983) and Mount Pinatubo in the Philippines in  
756 1991 (Tupper et al., 2005) resulted in elevating global AOD levels in the following years. The AOD  
757 recovery to the previous low levels after volcanic eruptions takes approximately 10 years (Chazette  
758 et al., 1995; Sun et al., 2019). This further indicates the efficiency of our data capturing the volcanic  
759 eruption emission features.

760 Due to the influence of geography, atmospheric circulation, population, and emissions, the trend of  
761 global aerosols varies in different latitude Figure 12 illustrates the multi-year average AOD in  
762 different latitude ranges for land, the NH, and the SH from 1980 to 2021. Within  $[-20, 20^\circ N]$ , the  
763 global average AOD reaches its maximum (0.234), and the maximum AOD NH is 0.256 in  $[0, 20^\circ N]$ .  
764 The highest AOD in SH is 0.217 in  $[-15, 0^\circ N]$ . The average AOD in SH rapidly decreases from  $-$   
765  $15^\circ N$  to  $-35^\circ N$ . In NH, AOD is generally greater than in SH from  $5^\circ N$  to  $65^\circ N$ . When, the latitude  
766 is greater than  $70^\circ N$ , the NH's AOD is smaller than the SH's, which may be related to low emission  
767 intensity and low population density in high latitude areas.

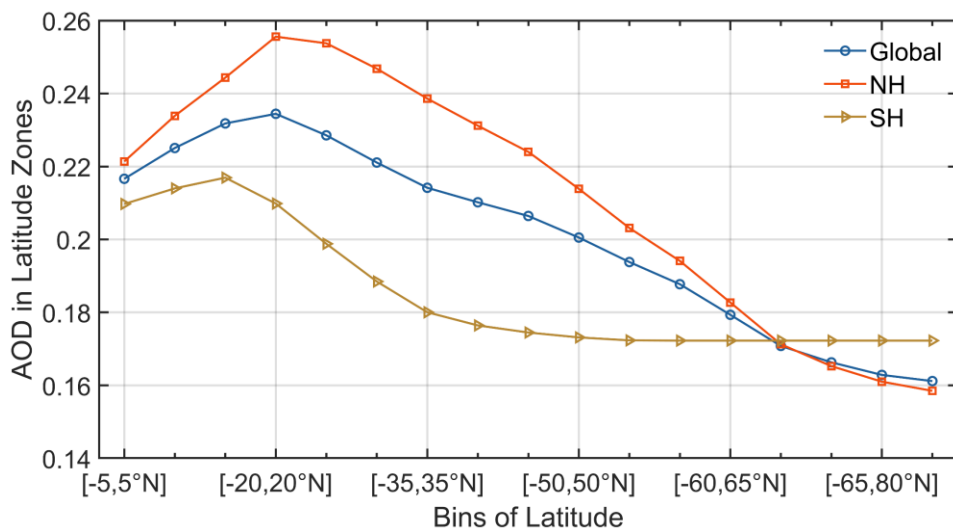
768 The seasonal trends of AOD during 1980-2021 at the global and hemispheric scales are shown in  
769 Figure 11 (g-j). The global AOD shows a decreasing trend in all seasons ( $-0.002 \sim -0.003/10a$ ). The  
770 large declining trends are observed in JJA and SON, with decreasing trend values of  $-0.003/10a$  and  
771  $-0.0029/10a$ , respectively. DJF and MAM follow with decreasing trend values of  $-0.0026/10a$  and  $-$   
772  $0.0022/10a$ , respectively, all passing the significance test ( $p < 0.01$ ). For the NH, the AOD trends in  
773 different seasons are  $-0.0030/10a$  (DJF),  $-0.0006/10a$  (MAM),  $-0.0005/10a$  (JJA), and  $-0.0034/10a$   
774 (SON). DJF and SON pass the significance test ( $p < 0.01$ ), while MAM and JJA do not. In the SH,  
775 the trends are as follows:  $-0.0011/10a$  (DJF),  $-0.0085/10a$  (MAM),  $-0.0131/10a$  (JJA), and  $-$   
776  $0.0009/10a$  (SON). Interestingly, in contrast to the NH, MAM and JJA pass the significance test,  
777 while DJF and SON do not. The largest declining season in the NH is winter, while in the SH, it is  
778 summer. The decreasing trend in the SH is more than four times greater than that in the NH,  
779 particularly before the year 2000. While both the global and SH AOD exhibit a decreasing trend  
780 since 2005, the NH has shown a significant increase in winter AOD, leading to an overall increasing  
781 trend. Moreover, the NH shows an increasing trend of  $0.004/10a$  from 2005 to 2021.

782 Annual  $SO_2$  emissions increased from 9.4 to 15.3 TgS from 2000 to 2005, which ultimately ended  
783 up as sulfate aerosols, leading to a significant increase in sulfate aerosols (Hofmann et al., 2009).  
784 More relevantly, the frequent volcanic eruptions in tropical regions from 2002 to 2006, combined  
785 with seasonal circulation patterns during winter, led to the transport of aerosol particles to higher  
786 latitudes (Hofmann et al., 2009; Vernier et al., 2011; Sawamura et al., 2012; Andersson et al., 2015).



788

789 **Figure 11** The multi-year averages of VIS\_AOD from 1980 to 2021. Global land (circle), northern  
 790 hemisphere (NH,0-85°N) (triangle) and southern hemisphere (SH,0-60°S) (square) annual and  
 791 seasonal AOD. The symbol, \*\*, represents that the test passed at a significance level of 0.01. DJF  
 792 represents December and next January and February. MAM represents March, April, and May. JJA  
 793 represents June, July, and August. SON represents September, October, and November.



794

795 Figure 12 The global land (blue), northern hemisphere's (red) and southern hemisphere's (yellow)  
796 multi-year average VIS\_AOD from 1980 to 2021 in different latitude zones. The latitude range is  
797 from -60 to 85°N, with a bin of 5°.

### 798 **3.6 Interannual variability and trend of visibility-derived AOD over regions**

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

804 To analyze the spatiotemporal characteristics and trends of AOD in different regions, we selected  
805 12 representative regions that are influenced by various aerosol sources(Wang et al., 2009; Hsu et  
806 al., 2012; Chin et al., 2014), such as desert, industry, anthropogenic emissions, and biomass burning  
807 emissions, which nearly cover the most land and are densely populated regions (Kummu et al.,  
808 2016). These representative regions are Eastern Europe, Western Europe, Western North America,  
809 Eastern North America, Central South America, Western Africa, Southern Africa, Australia,  
810 Southeast Asia, Northeast Asia, Eastern China, and India, as shown in Figure 1. We use multi-year  
811 average and seasonal average AOD to evaluate aerosol loadings (Figure 13), the annual average of  
812 monthly anomalies to analyze interannual trends (Figure 14), and the seasonal average to analyze  
813 seasonal trends (Figure 15) in 12 regions from 1980 to 2021.

814 We can see some differences between VIS\_AOD and MODIS AOD. In addition to model errors,  
815 the spatial matching between meteorological stations and MODIS, terrain, surface coverage, and  
816 station altitude will also bring errors. When particle transport and photochemical reactions occur  
817 above the boundary layer, visibility cannot capture the feature, which will also increase the  
818 uncertainty. However, bias is inevitable and can only be kept as small as possible. From the trend,  
819 they have similar changing characteristics, especially on monthly and yearly scales.

820 Figure 13 shows the regions with high AOD level from 1980 to 2021 (multi-year average AOD >  
821 0.2) are in West Africa, Northeast Asia, Eastern China, and India. The AOD values in Eastern North  
822 America, Central South America, South Africa, and Southeast Asia range from 0.15 to 0.2. The  
823 AOD values in Eastern Europe, Western Europe, Western North America, and Australia are less than  
824 0.15.

825 Europe is an industrial region with a low aerosol loading region, and the multi-year average AOD  
826 in Eastern Europe ( $0.144 \pm 0.007$ ) is higher than that in Western Europe ( $0.139 \pm 0.003$ ) during 1980-  
827 2021. Eastern Europe shows a greater downward trend in AOD ( $-0.0041/10a$ ) compared to Western  
828 Europe ( $-0.0021/10a$ ). The highest AOD is observed in JJA, the dry period when solar irradiation  
829 and boundary layer height increase, with Eastern Europe at 0.161 and Western Europe at 0.162,  
830 which could be due to increases in secondary aerosols, biomass burning, and dust transport from  
831 the Sahara (Mehta et al., 2016). However, there are seasonal variations. In Eastern Europe, the  
832 seasonal AOD ranking from high to low is JJA (0.161) > DJF (0.147) > MAM (0.138) > SON  
833 (0.131), while in Western Europe, it is JJA (0.162) > MAM (0.140) > SON (0.136) > DJF (0.117).  
834 The differences among seasons are larger in Western Europe. AOD in Eastern Europe shows  
835 declining trends in all seasons, while it does not pass the significance test in MAM. Among four

836 seasons, SON has the largest decline trend of AOD (-0.0058/10a). In Western Europe, DJF, JJA, and  
837 SON exhibit declining trends of AOD that pass the significance test, while the MAM shows a  
838 significant increase trend of AOD (0.0022/10a), which may be due to eruptions of the  
839 Eyjafjallajökull volcano in Iceland in spring 2010 (Karbowska and Zembrzuski, 2016). Both  
840 Western and Eastern Europe experienced increasing trends in AOD during the period of 1995-2005,  
841 with Western Europe showing a greater increase. However, after 2000, the decline rate accelerated  
842 in both regions. The downward trend in Europe is attributed to the reduction of biomass burning,  
843 anthropogenic aerosols, and aerosol precursors (such as sulfur dioxide)(Wang et al., 2009; Chin et  
844 al., 2014; Mortier et al., 2020).

845 North America is also an industrial region with a low aerosol loading. The average AOD values for  
846 Eastern and Western North America during 1980-2021 are  $0.153\pm 0.004$  and  $0.131\pm 0.005$ ,  
847 respectively, with the Eastern region being higher than the Western region by 0.022. From 1980 to  
848 2021, both Eastern (-0.0021/10a) and Western North America (-0.0009/10a) show a downward trend;  
849 however, the decline in the Western region is not statistically significant. And the trend is -  
850  $0.0172/10a$  from 1995 to 2005 and  $0.0096/10a$  from 2005 to 2021. The average AOD values in DJF,  
851 MAM, JJA, and SON in Western North America are 0.1367, 0.1286, 0.1457, and 0.114, respectively,  
852 compared to 0.137, 0.145, 0.1913, and 0.138 in Eastern North America. The lowest AOD values of  
853 12 regions during DJF and SON are observed in Western North America (Remer et al., 2008).  
854 Specifically, in the Western region, there is a consistent increasing trend during MAM (0.004/10a)  
855 from 1980 to 2021, while JJA and SON also show an increase after 2000, except for DJF (-  
856  $0.0032/10a$ ). In contrast, the AOD trends in the Eastern region remain unchanged during the period  
857 1980-2021, except for MAM, which shows a stable increasing trend (0.0033/10a), while DJF, JJA,  
858 and SON exhibit decreasing trends (-0.0023/10a, -0.0040/10a, -0.0053/10a, respectively). In the  
859 Western region, the annual mean AOD started to increase after 2005, while in the Eastern region,  
860 the increase was not significant. The upward trend may be due to low rainfall and increased wildfire  
861 activities (Yoon et al., 2014). The decrease in AOD in Eastern North America is related to the  
862 reduction of sulfate and organic aerosols, as well as the decrease in anthropogenic emissions caused  
863 by environmental regulations (Mehta et al., 2016).

864 Central South America is a relatively high aerosol loading region, sourced from biomass burning,  
865 especially in SON (Remer et al., 2008; Mehta et al., 2016), with a multi-year average AOD of  
866  $0.192\pm 0.017$ . There is a clear downward trend (-0.0100/10a) from 1980 to 2021, which is slightly  
867 greater than the trend (-0.0090/10a) from 1998 to 2010 (Hsu et al., 2012) and AOD decreased from  
868 1980 to 2006 (Streets et al., 2009) and from 2001 to 2014 (Mehta et al., 2016). Although DJF (0.199)  
869 and SON (0.226) have higher values compared to MAM (0.180) and JJA (0.163), the large declining  
870 trends are observed in MAM (-0.0126/10a) and JJA (-0.0167/10a). It indicates that although AOD  
871 has decreased overall, the aerosol loading caused by seasonal deforestation and biomass combustion  
872 is still large (Mehta et al., 2016).

873 Africa is also one of the regions with a high aerosol loading worldwide. In West Africa, the average  
874 AOD is  $0.275\pm 0.012$  during 1980-2021, and the annual AOD shows a downward trend (-0.0008/10a,  
875  $p>0.05$ ). The world's largest desert (Sahara Desert) is in West Africa, with much dust aerosol  
876 discharged. AOD values in all seasons are above 0.25, with JJA (0.301) and MAM (0.300) reaching  
877 0.3, and DJF and SON being 0.252 and 0.250, respectively. The AOD in DJF (-0.0135/10a,  $p<0.01$ )  
878 and SON (-0.0026/10,  $p>0.05$ ) exhibit decreasing trends, while JJA (0.0088/10a,  $p<0.01$ ) and MAM

879 (0.0037/10a,  $p > 0.05$ ) show an opposite trend. The multi-year average AOD in South Africa is  
880  $0.177 \pm 0.020$ , lower than that of West Africa. The annual mean AOD in South Africa shows a  
881 significant decrease ( $-0.0096/10a$ ). The AOD values range from 0.12 to 0.2 during 2000-2009,  
882 dominated by fine particle matter from industrial pollution from biomass and fossil fuel combustion  
883 (Hersey et al., 2015). The average AOD values in DJF, MAM, JJA, and SON are 0.189, 0.162, 0.147,  
884 and 0.210, respectively. JJA ( $-0.0268/10a$ ,  $p < 0.01$ ), MAM ( $-0.0126/10a$ ,  $p < 0.01$ ) and SON ( $-$   
885  $0.0001/10a$ ,  $p > 0.05$ ) exhibit a downward AOD trend, while DJF ( $0.0006/10a$ ,  $p > 0.05$ ) shows an  
886 upward trend. AERONET and simulation results also show a decreasing trend of AOD (Chin et al.,  
887 2014).

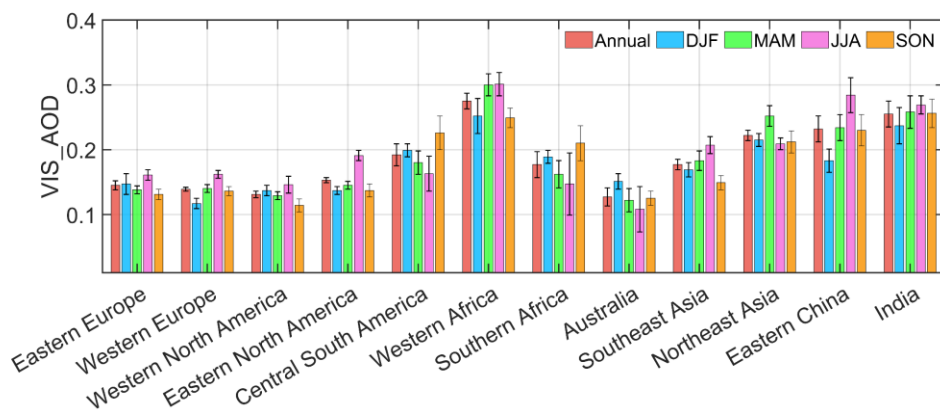
888 Australia is a region with a low aerosol loading. The multi-year mean AOD is  $0.127 \pm 0.014$  during  
889 1980-2021. The AOD ranges from 0.05 to 0.15 from AERONET during 2000-2021, and dust and  
890 biomass burning are important contributors to the aerosol loading (Yang et al., 2021a). There is a  
891 downward trend of AOD ( $-0.0081/10a$ ,  $p < 0.01$ ), which may be related to a decrease in dust and  
892 biomass burning (Yoon et al., 2016; Yang et al., 2021a). In addition, research has shown that the  
893 forest area in Australia has increased sharply since 2000 (Giglio et al., 2013), surpassing the forest  
894 fire area of the past 14 years. The seasonal average of AOD in MAM, JJA, SON, and DJF are 0.122,  
895 0.108, 0.125, and 0.151. The AOD in JJA is the lowest among all seasons and regions. The highest  
896 AOD is in DJF with an increasing trend ( $0.0056/10a$ ,  $p < 0.01$ ), while the trends during MAM, JJA  
897 and SON are  $-0.0096/10a$  ( $p < 0.01$ ),  $-0.0231/10a$  ( $p < 0.01$ ) and  $-0.0042/10a$  ( $p < 0.01$ ), respectively.  
898 Ground-based and satellite observations indicate that wildfires, biomass burning and sandstorms  
899 lead to high AOD in DJF and SON. The low AOD of MAM and JJA is due to a decrease in the  
900 frequency of sandstorms and wildfires and an increase in precipitation (Gras et al., 1999; Yang et  
901 al., 2021a; Yang et al., 2021b).

902 Asia is also a high aerosol loading area with various sources. In Southeast Asia, the multi-year  
903 average AOD is 0.177 during 1980-2021 with a downward trend of AOD ( $-0.0003/10a$ ,  $p > 0.05$ ). It  
904 is also a biomass-burning area. The seasonal average AOD ranking from high to low is JJA (0.207) >  
905 MAM (0.183) > DJF (0.169) > SON (0.149). The trends in DJF ( $-0.0035/10a$ ,  $p < 0.05$ ), JJA ( $-$   
906  $0.0007/10a$ ,  $p > 0.05$ ) and SON ( $-0.0021/10a$ ,  $p > 0.05$ ) are opposite to MAM ( $0.0050/10a$ ,  $p < 0.01$ ).  
907 Southeast Asia has no clear long-term trend in estimated AOD or observed surface solar radiation  
908 (Streets et al., 2009). In Northeast Asia, the multi-year average AOD is 0.222 during 1980-2021,  
909 with no significant temporal trend. The seasonal AOD values are 0.252 in MAM, 0.215 in DJF,  
910 0.212 in SON and 0.209 in JJA. AOD in MAM is significantly higher than other seasons, which  
911 may be related to sandstorms in East Asia, and the reason for the high AOD in winter may be related  
912 to the transportation. The trends of AOD in DJF ( $-0.0025/10a$ ,  $p > 0.05$ ), MAM ( $0.0031/10a$ ,  $p > 0.05$ ),  
913 JJA (0) and SON ( $-0.0006/10a$ ,  $p > 0.05$ ) are not significant. In Eastern China, the multi-year average  
914 AOD is 0.233, with an increasing trend ( $0.0071/10a$ ,  $p < 0.01$ ). The trend is  $0.0151/10a$  from 1980 to  
915 2006 and  $-0.0469/10a$  from 2006 to 2021. The seasonal average AOD ranking from high to low is  
916 JJA (0.284), MAM (0.234), SON (0.230) and DJF (0.183). The AOD trends in DJF ( $0.0093/10a$ ,  
917  $p < 0.01$ ), MAM ( $0.0092/10a$ ,  $p < 0.01$ ), JJA ( $0.0038/10a$ ,  $p > 0.05$ ) and SON ( $0.0065/10a$ ,  $p < 0.05$ ) are  
918 all positive but the trend in JJA does not pass the significance test. We can see that there are three  
919 stages of changes in AOD: 1980-2005, 2006-2013 and 2014-2021. In the first stage, AOD increased  
920 steadily. In the second stage, AOD maintained a larger positive anomaly accompanied by  
921 oscillations. The third stage experienced a rapid decline, reaching the level of the 1980s by 2021.

922 The increasing trend of AOD before 2006 may be due to the significant increase in industrial activity,  
 923 and after 2013, the significant decrease is closely related to the implementation of air quality-related  
 924 laws and regulations, along with adjustments in the energy structure (Hu et al., 2018; Cherian and  
 925 Quaas, 2020).

926 India is a high aerosol loading area. The multi-year average AOD is 0.255, with an upward trend  
 927 (0.0096/10a,  $p < 0.01$ ) from 1980 to 2021. Dust and biomass burning has an influence on AOD level.  
 928 There are three stages: 1980-1997 (0.0032/10a,  $p < 0.01$ ), 1997-2005 (-0.0420/10a,  $p < 0.01$ ), 2005-  
 929 2021 (0.0481/10a,  $p < 0.01$ ). Although the trend is downward in the second stage, the larger positive  
 930 trend is in the third stage. The seasonal average AOD values are 0.237 in DJF, 0.258 in MAM, 0.269  
 931 in JJA, and 0.256 in SON. The largest AOD is in JJA. In winter and autumn, it affected by biomass  
 932 burning, and in spring and summer, it is also affected by dust, transported from the Sahara under  
 933 during the monsoon period (Remer et al., 2008). The trends in DJF (0.0152/10a,  $p < 0.01$ ), MAM  
 934 (0.0091/10a,  $p < 0.01$ ), JJA (0.0025/10a,  $p > 0.05$ ), and SON (0.0107/10a,  $p < 0.05$ ) are positive. There  
 935 largest trend is in winter.

936 To summarize, there are significant differences in the spatial distribution, annual trends, and  
 937 seasonal trends of AOD across different regions from 1980 to 2021. The high aerosol loadings from  
 938 1980 to 2021 are in West Africa, India and Asia, and low aerosol loading regions are in Europe,  
 939 Western North America, and Australia. Eastern China and India show an increasing trend, Southeast  
 940 Asia and Northeast Asia show no significant trend, and the other regions show downward trends.  
 941 However, not all regional seasonal trends are consistent with their annual trends. The results in this  
 942 study have supplemented the long-term trend and distribution of AOD over land.

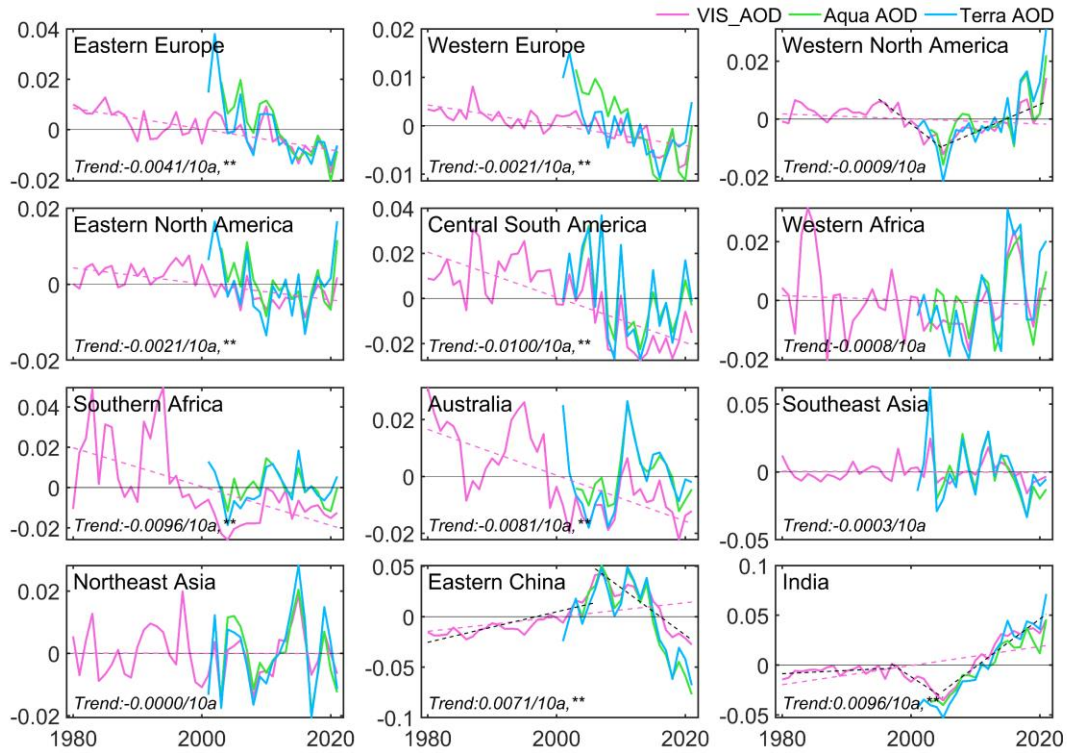


943

944 **Figure 13** Annual and seasonal averages of AOD in 12 regions during 1980-2021.

945

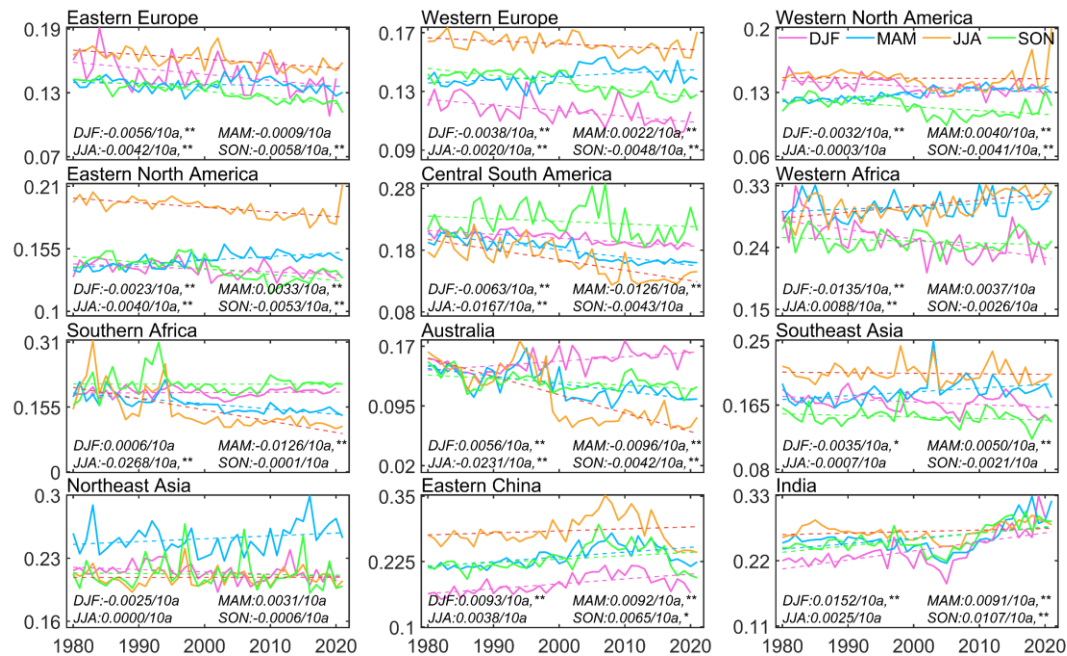




946

947 **Figure 14** Annual averages of monthly anomaly gridded VIS\_AOD (pink line), Aqua (green line),  
 948 and Terra (blue line) MODIS AOD in 12 regions. The dotted line is the trend line.

949



950

951 **Figure 15** Seasonal averages of gridded VIS\_AOD during 1980 to 2021 in 12 regions (Eastern  
 952 Europe, Western Europe, Western North America, Eastern North America, Central South America,  
 953 Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and

954 India). The dotted line is the trend line.

## 955 **4 Data availability**

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

## 959 **5 Conclusions**

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

971 **1. Modeling and gridding evaluation.** The mean RMSE, MAE, and R of all stations are 0.078,  
972 0.044, and 0.750, respectively. The RMSE of 93% stations is less than 0.11, the MAE of 91% is less  
973 than 0.06, and the R of 88% is greater than 0.7, respectively. Compared to Aqua and Terra, the  
974 average biases of gridded AOD are 3.3% and 1.9%, and the spatial correlation coefficients are 0.80  
975 and 0.79, with the zonal correlation coefficients of 0.99 and 0.99 and the meridional correlation  
976 coefficients of 0.99 and 0.90.

977 **2. Model validation.** For the daily scale, the R, RMSE and MAE of between VIS\_AOD and Aqua  
978 AOD is 0.799, 0.079 and 0.044, respectively. The percentage of sample point falling within the EE  
979 envelopes is 84.12%. The R between VIS\_AOD and Terra AOD is 0.542, with a RMSE of 0.125  
980 and MAE of 0.078. The percentage falling within the EE envelopes is 64.76%. The R between  
981 VIS\_AOD and AERONET AOD is 0.546, with a RMSE of 0.186 and MAE of 0.099. The percentage  
982 falling within the EE envelopes is 57.87%. For the monthly and annual scales, RMSE and MAE  
983 show a significant decrease between VIS\_AOD and Aqua, Terra, and AERONET AOD, and R and  
984 percentages falling within EE show a significant increase 错误!未找到引用源。 .

985 **3. Error analysis.** The average bias is 0.015 (AOD < 0.1), with 83% of data within the EE envelopes.  
986 As pollution level increases, the negative mean bias becomes significant and the underestimation  
987 increases. There is a negative bias in the low elevation ( $\leq 0.5$ km) with a percentage of 60%-64%  
988 falling within the EE envelopes and a positive bias in high elevation (0.5-1.2km) with a percentage  
989 of 50%-65% falling within the EE envelopes. The elevation of AERONET's site caused a bias in  
990 high elevation. When the elevation difference is negative (the elevation of the meteorological station  
991 is lower than that of the AERONET site), there is a significant positive bias. When the difference is

992 positive, the mean bias approaches 0 or is positive. The bias does not change significantly with  
993 increasing distance between the meteorological station and AERONET site.

994 **4. Global land AOD.** The global, NH, and SH AOD values from 1980 to 2021 are  $0.161 \pm 0.074$ ,  
995  $0.158 \pm 0.076$ , and  $0.173 \pm 0.059$ , respectively. Trends in AOD for the global, NH, and SH  
996 demonstrate a decreasing trend of  $-0.0026/10a$ ,  $-0.0018/10a$ , and  $-0.0059/10a$ , respectively ( $p < 0.01$ ).  
997 The seasonal AOD ranking from high to low is JJA > MAM > DJF > SON over the global land and in  
998 the NH, while in the SH, it is DJF > JJA > MAM > SON. The largest declining trends are observed in  
999 NH summer and SH winter.

1000 **4. Regional AOD.** From 1980 to 2021, regions with high aerosol loadings ( $AOD > 0.2$ ) were found  
1001 in West Africa, Northeast Asia, Eastern China, and India. Regions with moderate aerosol loadings  
1002 ( $AOD$  between 0.15 and 0.2) are Eastern North America, Central South America, South Africa, and  
1003 Southeast Asia. Eastern Europe, Western Europe, Western North America, and Australia are regions  
1004 with low aerosol loadings ( $AOD < 0.15$ ). The trends are  $-0.0041/10a$ ,  $-0.0021/10a$ ,  $-0.0009/10a$ ,  
1005  $-0.0021/10a$ ,  $-0.0100/10a$ ,  $-0.0008/10a$ ,  $-0.0096/10a$ ,  $-0.0081/10a$ ,  $-0.0003/10a$ ,  $-0.0000/10a$ ,  
1006  $0.0071/10a$ , and  $0.0096/10a$  in Eastern Europe, Western Europe, Western North America, Eastern  
1007 North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia,  
1008 Northeast Asia, Eastern China, and India, respectively.

## 1009 **Competing interests**

1010 The contact author has declared that none of the authors has any competing interests.

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1014 visibility data were download from <https://mesonet.agron.iastate.edu/ASOS>. The Aerosol  
1015 Robotic Network (AERONET) 15-minute aerosol optical depth (AOD) data were download from  
1016 which can be downloaded from <https://aeronet.gsfc.nasa.gov>. The MODIS AOD data were  
1017 download from <https://ladsweb.modaps.eosdis.nasa.gov>.

1018

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