

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

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12 Abstract

13 Long-term and high spatial resolution aerosol optical depth (AOD) data are ~~essential~~necessary for
14 climate change detection and attribution. Global ground-based AOD observation stations are
15 ~~sparse~~ly distributed, and satellite AOD observations have a low ~~temporal~~ time-frequency, as well
16 low accuracy before 2000 over land. In this study, AOD ~~was~~is derived from hourly visibility
17 observations collected at more than 5000 meteorological stations ~~of the Automated Surface~~
18 ~~Observing System (ASOS)~~ over global land from 1980 to 2021. The AOD retrievals of the Moderate
19 Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua Earth observation satellite ~~were~~
20 are used to train the machine learning ~~method~~model, and the ERA5 reanalysis boundary layer height
21 ~~was~~is used to convert the surface visibility to AOD. Comparisons with independent dataset show
22 that as input. The predicted result AOD has correlation coefficients of 0.54 and 0.55~~1~~
23 MODIS satellite retrievals and AERONET ground observations at daily time scale. The correlation
24 coefficients are higher at monthly and annual scales, which are 0.8~~108~~ and 0.613 for the monthly
25 and 0.9~~106~~ and 0.652 for the annual, compared with Terra MODIS and AERONET AOD,
26 respectively. The visibility-derived AOD at ~~ASOS~~ stations ~~scale is~~was gridded into a 0.5° ~~degree~~
27 resolution grid by ~~area-weighted~~ ordinary kriging interpolation. ~~Analysis of visibility-derived~~
28 ~~AOD indicates that for the global scale,~~ The mean visibility-derived AOD of over the global land
29 (-60°N-85°N), the Northern Hemisphere, and the Southern Hemisphere are 0.161, 0.158, and 0.173
30 from 1980 to 2021, with a trend of -0.0026/10a, -0.0018/10a, and -0.0059/10a from 1980 to 2021,
31 respectively. For the regional scale, the mean ~~AOD~~(trends) of AOD from 1980 to 2021 are 0.145 (-
32 0.0041/10a), 0.139 (-0.0021/10a), 0.131 (-0.0009/10a), 0.153 (-0.0021/10a), 0.192 (-0.0100/10a),
33 0.275 (-0.0008/10a), 0.177 (-0.0096/10a), 0.127 (-0.0081/10a), 0.177 (-0.0003/10a), 0.222 (-
34 0.0000/10a), 0.232 (0.0071/10a), and 0.255 (0.0096/10a) in Eastern Europe, Western Europe,
35 Western North America, Eastern North America, Central South America, Western Africa, Southern
36 Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India, ~~respectively~~. The
37 visibility-derived AOD at station and grid scales over global land from 1980 to 2021 are available

38 at National Tibetan Plateau / Third Pole Environment Data Center
39 (<https://doi.org/10.11888/Atmos.tpsc.300822>) (Hao et al., 2023).

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41 depth over global land (1980-2021). National Tibetan Plateau / Third Pole Environment Data
42 Center. <https://doi.org/10.11888/Atmos.tpsc.300822>.

43 1 Introduction

44 Atmospheric aerosols are composed of solid and liquid particles suspended in the atmosphere.
45 ~~Aerosol particles are directly emitted into the atmosphere or formed through gas-particle~~
46 ~~transformation. Aerosol particles are primarily discharged from the Earth's surface broadly classified~~
47 ~~into natural and anthropogenic sources~~ (Calvo et al., 2013). ~~They possess, with~~ diverse shapes and
48 sizes (Fan et al., 2021), optical properties, and various components (Liao et al., 2015; Zhang et al.,
49 2020; Li et al., 2022), ~~such as inorganic salts, organic matter, metal elements and elemental carbon.~~
50 Most atmospheric aerosols are concentrated in the troposphere, especially in the boundary layer
51 (Liu et al., 2022), with a high concentration near emission sources (Kulmala et al., 2004) , and a
52 small portion are distributed in the stratosphere. ~~with a sharp increase during large volcanic~~
53 ~~eruptions. Some aerosols from wildfires, volcanoes and sandstorms, play an important role in~~
54 ~~tropospheric aerosols. Studies have showed that 75% of volcanic eruptions inject volcanic aerosols~~
55 ~~and sulfur-containing gases into the troposphere (Halmer et al., 2002), wildfire aerosols contribute~~
56 ~~up to approximately 35% of the fine particles in Europe (Barnaba et al., 2011), and dust aerosols are~~
57 ~~mainly concentrated in the middle and low troposphere (Filonchik et al., 2018).~~ Atmospheric
58 aerosols severely impact the atmospheric environment and human health. They deteriorate air
59 quality, reduce visibility, and cause other environmental issues (Wang et al., 2012; Boers et al.,
60 2015). They ~~impair~~ affect human health or other organisms' conditions by increasing cardiovascular
61 and respiratory disease incidence and mortality rates (Chafe et al., 2014; Yang et al., 2022). The
62 Global Burden of Disease shows that global exposure to ambient PM_{2.5} resulted in 0.37 million
63 deaths and 9.9 million disability-adjusted life years (Chafe et al., 2014).

64

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

73 Tropospheric aerosols are considered as the second largest forcing factor for global climate change
74 (Li et al., 2022), and they reduce the warming due to greenhouse gases by -0.5°C (Ipcc, 2021).
75 However, aerosols are also regarded as the largest contributor to quantifying the uncertainty of
76 present-day climate change (Ipcc, 2021). ~~The uncertainties are caused by the deficiencies of the~~
77 ~~global descriptions of aerosol optical properties (such as scattering and absorption) and~~

78 ~~microphysical properties (such as size and component), and the impact on cloud and precipitation,~~
79 ~~further affecting the estimation of aerosol radiative forcing~~The deficiency of the global descriptions
80 ~~of aerosol optical and microphysical properties is the primary reason for the uncertainty and the~~
81 ~~uncertainty also exists in climate models~~ (Lee et al., 2016; Ipcc, 2021). Therefore, sufficient aerosol
82 observations are crucial. In aerosol measurements, aerosol optical depth (AOD) is often used to
83 describe its column properties, which represents the vertical integration of aerosol extinction
84 coefficients. AOD is an important physical quantity for estimating the content, atmospheric
85 pollution and climatology of aerosols (Zhang et al., 2020).

86 ~~AOD data usually from ground-based and satellite-borne remote sensing observation. They have~~
87 ~~both advantages and disadvantages. The measurements of aerosols are usually divided into in-situ~~
88 ~~and remote sensing observations. In situ observations accurately measure the mass, number~~
89 ~~concentration, shapes, compositions and scattering and absorption of aerosols by directly sampling~~
90 ~~the air (Herich et al., 2008; Laj et al., 2020). Observations from airplanes and balloons can provide~~
91 ~~vertical structure (Ziemba et al., 2013). Because of its accuracy, in-situ observation is often used as~~
92 ~~the benchmark for models and satellites, but its spatial representativeness is limited. Another method~~
93 ~~is g~~Ground-based lidar observation, ~~which~~ is an active remote sensing technology. Lidar generally
94 emits laser and receives backscattered signals to invert the extinction coefficient of aerosols at
95 different heights (Klett, 1985). By using the depolarization ratio, the type of aerosol, such as fine
96 particles or dust, can ~~also~~ be distinguished (Bescond et al., 2013). The AOD within a certain height
97 can be calculated by integrating the extinction coefficients; however, scattering signals are usually
98 not received near the ground, leading to blind spots (Singh et al., 2019). At present, there are many
99 ground-based lidar worldwide and regional networks, which provides important support ~~in the study~~
100 of vertical changes in aerosols, such as the NASA Micro-Pulse Lidar Network (MPLNET) in the
101 early 1990s (Welton et al., 2002), the European Aerosol Research Lidar Network (EARLINET)
102 since 2000 (Bösenberg and Matthias, 2003), the Latin American Lidar Network (LALINET) since
103 2013 (Guerrero-Rascado et al., 2016).

104
105 ~~The other two passive remote sensing observations of aerosol properties are ground-based and~~
106 ~~satellite-borne remote sensing observations.~~Ground-based remote sensing observations supply
107 aerosol loading data (such as AOD), by measuring the attenuation of radiation from the top of the
108 atmosphere to the surface (Holben et al., 1998). This type of observations mainly uses weather-
109 resistant automatic sun and sky scanning spectral radiometers to retrieve optical and microphysical
110 aerosol properties (Che et al., 2014). The Aerosol Robotic Network (AERONET) is a popular global
111 network composed of NASA and multiple international partners that provides high-quality and high-
112 frequency aerosol optical and microphysical properties under various geographical and
113 environmental conditions (Holben et al., 1998; Dubovik et al., 2000). The AERONET observations
114 are extensively used to validate ~~of~~ satellite remote sensing observations and model simulations, as
115 well as climatology study (Dubovik et al., 2002b). There are many regional networks of sun
116 photometers, such as the Maritime Aerosol Network (MAN), which use a handheld sun photometer
117 to collect data ~~overon~~ the ocean and is merged into AERONET (Smirnov et al., 2009), the China
118 Aerosol Robot Sun Photometer Network (CARSPNET) (Che et al., 2009), the Canadian sub-network
119 of AERONET (AEROCAN) (Bokoye et al., 2001), Aerosol characterization via Sun photometry:
120 Australian Network (AeroSpan) (Mukkavilli et al., 2019), and the sky radiometer network

121 (SKYNET) in Asia and Europe (Kim et al., 2004; Nakajima et al., 2020). Another very valuable
122 global network is the NOAA/ESRL Federated Aerosol Network (FAN), which uses integrated
123 nephelometers distinct from sun photometers, mainly located in ~~remote~~ areas ~~with less human~~
124 ~~activity impact~~, providing ~~background regionally representative~~ aerosol properties over 30 sites
125 (Andrews et al., 2019).

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

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

154 To overcome these limitations and enrich aerosol data, alternative observation data could be utilized
155 to derive AOD. ~~For example, some studies used solar radiation data to infer AOD and analyze the~~
156 ~~characteristics of AOD in different regions (King et al., 1978; Vasilyev et al., 1995; Marengo et al.,~~
157 ~~1997; Qiu, 1997). There are also some studies deriving AOD based on empirical relationship~~
158 ~~between particle concentration and AOD (Xie et al., 2015; Li, 2020). These methods partially~~
159 ~~mitigate the scarcity of AOD data in spatial coverage, but it is also important to acknowledge the~~
160 ~~inherent limitation of long temporal coverage. Another more suitable alternative is a~~Atmospheric
161 horizontal visibility is a suitable alternative (Wang et al., 2009; Zhang et al., 2020), because it has
162 the advantages of the long-term records with a large number of stations worldwide.

163

164 Atmospheric visibility is a physical quantity that describes the transparency of the atmosphere
165 through manual and automatic observations, ~~and. The automatic observations of visibility usually~~
166 ~~measure atmospheric extinction (scattering coefficient and transmissivity), including particulate matter,~~
167 ~~water vapor, and gas molecules, which makes it a favorable choice for inferring AOD.~~ Koschmieder
168 (1924) first proposed the relationship between the meteorological optical range and the total optical
169 depth. Elterman (1970) further established a formula between AOD and visibility by assuming an
170 exponential decrease in aerosol concentration with altitude, considering the extinction of molecules
171 and ozone to analyze air pollution, which called the Elterman model. Qiu and Lin (2001) corrected
172 the Elterman model by considering the influence of water vapor and used two water vapor pressure
173 correction coefficients to retrieve AOD of 16 stations in China in 1990. Wang et al. (2009) analyzed
174 the trend of AOD using visibility-based retrievals from 1973 to 2007 over land. Lin et al. (2014)
175 retrieved the AOD in eastern China in 2006 using visibility and aerosol vertical profiles provided
176 by GEOS-Chem. Wu et al. (2014) and Zhang et al. (2017) parameterized the constants in the
177 Elterman model and use satellite retrieved AOD to solve the parameters in the models at different
178 stations, to retrieve the long-term AOD in China.

179 Zhang et al. (2020) reviewed the methods of visibility retrieval of AOD, indicating that visibility-
180 based retrieval of AOD can compensate for the shortcomings of long-term aerosol observation data.
181 Simultaneously, various parameters, such as station altitude, consistency of visibility data, water
182 vapor and aerosol vertical profiles (scale height), were discussed with modified suggestions
183 proposed. These studies have enriched AOD data regionally. ~~Due to the similar spatial distribution~~
184 ~~of the extinction coefficient and AOD, and the proportional relationship between the reciprocal of~~
185 ~~visibility and the extinction coefficient, Wang et al. (2009) analyzed the trend of AOD using~~
186 ~~visibility-based retrievals from 1973 to 2007 over land.~~ These studies have enriched aerosol data in
187 some extent. At present, there are very few studies on global visibility-retrieved AOD and to analyze
188 climatology of aerosols.

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

effectively solve complex relationships among variables. And —previous studies are mostly conducted at the regional or national scale, and few studies at the global scale. Thus, it is feasible to derive AOD from atmospheric visibility over global land by using the machine learning method.

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

2 Data and method

2.1 Study area

The study area is global land. A total of 5032 meteorological stations ~~of the Automated Surface Observing System (ASOS), which is a joint surface weather observing network of the National Weather Service (NWS), the Federal Aviation Administration (FAA), and the Department of Defense (DOD)~~ (Noaa et al., 1998). ~~A total of and 573 stations of~~ 395 AERONET sites are selected in this study, ~~and~~ shown in Figure 1~~Figure 1 (a)~~. Twelve ~~12~~ regions are selected for special analysis, including Eastern Europe, Western Europe, Western North America, Eastern North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and Middle East~~India~~. The time range ~~in of~~ the study is from 1980 to 2021, during which the records of meteorological stations are sufficient with a uniform spatial distribution. As shown in Figure 1~~Figure 1 (b)~~, the daily records have exceeded 1500 stations, and monthly and annual records have exceeded 2000 during 1980-1990. After 2000, monthly records have reached 3000, which is the foundation of gridding AOD.

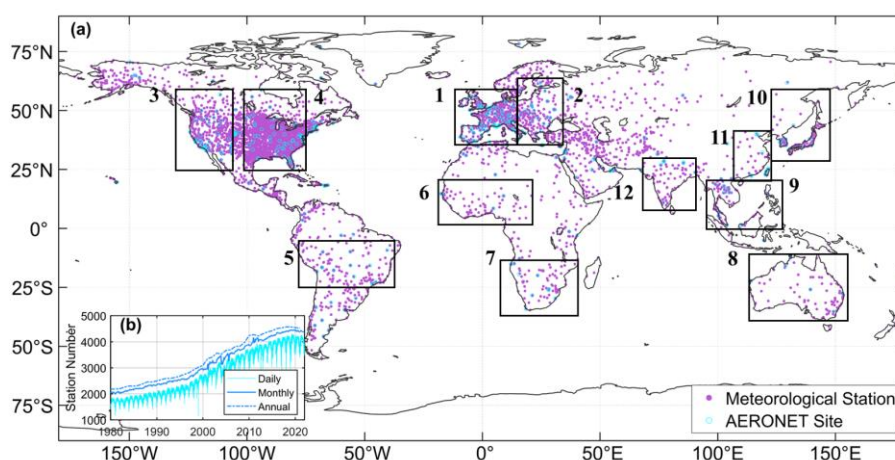


Figure 1 Study area (a) and the meteorological station number (b) with daily, monthly, and annual

234 records. The number of meteorological stations (filled circles) is 5032. The number of AERONET
235 sites (empty circles) is 3795. The box regions of labelled with number 1-12 are Eastern Europe,
236 Western Europe, Western North America, Eastern North America, Central South America, Western
237 Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India.

238 **2.2 Meteorological data**

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

250 The daily average visibility is calculated using harmonic mean. Experiments have found that
251 harmonic average visibility can better detect the weather phenomena than arithmetic average
252 visibility (Noaa et al., 1998). The visibility is calculated using the extinction coefficient, which is
253 directly proportional to the reciprocal of visibility (Wang et al., 2009). Harmonious average
254 visibility can capture the process of visibility decline more quickly. Therefore, daily visibility will
255 have greater representativeness:

256 ~~The hourly meteorological data from 1980 to 2021 are collected at 5032 globally distributed stations~~
257 ~~(Figure 1) from the Automated Surface Observing System (ASOS), which is a joint effort of the~~
258 ~~National Weather Service, the Federal Aviation Administration, and the Department of Defense,~~
259 ~~downloaded at [https://www.ncei.noaa.gov/products/land-based-station/automated-surface-weather-](https://www.ncei.noaa.gov/products/land-based-station/automated-surface-weather-observing-systems)~~
260 ~~observing-systems . From the 1960s to the 1970s, the Automated Meteorological Observing System~~
261 ~~and Remote Automated Weather Observing System only reported objective elements, such as~~
262 ~~temperature, dew point temperature, wind (speed and direction), and pressure. With technological~~
263 ~~advancements, the ASOS was deployed and utilized in the 1980s. The automatic surface~~
264 ~~observations reduced errors associated with human involvement in data acquisition, processing, and~~
265 ~~transmission. Effective quality control methods are employed to ensure the quality of ASOS~~
266 ~~products. ASOS provided hourly and even minutely ground automatic observations, primarily for~~
267 ~~airports (Noaa et al., 1998; Dover et al., 2002). Atmospheric visibility of ASOS is measured by the~~
268 ~~forward-scatter visibility sensor at 550 nm. The scattering angle of the sensor ranges from 0 to 45~~
269 ~~degree, the sampling volume is 0.75 cubic feet and the response time is 20 seconds. The sensor~~
270 ~~provides 1-minute average visibility with the day or night indication. Hourly visibility is calculated~~
271 ~~based on the harmonic average of minutely visibility. Experiments have found that harmonic~~
272 ~~average visibility can better detect the development of some weather phenomena than arithmetic~~
273 ~~average visibility (Noaa et al., 1998). The sensor measured visibility has a strong agreement with~~
274 ~~the human observed during haze and homogeneous weather over a large area, even during periods~~
275 ~~when weather conditions are quite variable (Noaa et al., 1998). The same algorithm is used to~~

276 ~~calculate the daily, monthly, seasonally and yearly average visibility.~~

$$277 \quad V = n / \left(\frac{1}{V_1} + \frac{1}{V_2} + \dots + \frac{1}{V_n} \right) \quad \text{Eq. 1}$$

278 where V is the harmonic mean visibility, n = 24 for the daily ~~mean~~visibility, and V_1, V_2, \dots, V_n are
279 the individual hourly ~~values~~visibility.

280 ~~Visibility in METAR is reported in statute miles (SM). The reportable increments are: M1/4SM,~~
281 ~~1/4SM, 1/2SM, 3/4SM, 1SM, 1 1/4SM, 1 1/2SM, 1 3/4SM, 2SM, 2 1/2SM, 3SM, 4SM, 5SM, 6SM,~~
282 ~~7SM, 8SM, 9SM and 10SM. It is noted that visibility between zero and 1/4 statute mile is reported~~
283 ~~as M1/4SM8. Visibility values of exactly halfway between reportable values are rounded down.~~
284 ~~Visibility values of 10 miles or greater are reported as 10SM (Noaa et al., 1998).—~~

285 In addition to hourly visibility (VIS), other variables closely related to aerosol properties are selected,
286 including relative humidity (RH), dew point temperature (DT), temperature (TMP), wind speed
287 (WS) and sea-level pressure (SLP). Temperature affects atmospheric stability and the rate of
288 secondary particle formation, and humidity influences the size and hygroscopic growth, and wind
289 speed and pressure significantly impact the transport and deposition.~~In addition to hourly visibility~~
290 ~~(VIS), we also selected other automatically observed variables closely related to aerosol properties~~
291 ~~in this study. Because relative humidity influences the size and hygroscopic growth rate of particle~~
292 ~~matter, and wind speed and pressure significantly impact the transport and deposition of aerosols,~~
293 ~~relative humidity (RH), dew point temperature (DT), temperature (TMP), wind speed (WS) and sea-~~
294 ~~level pressure (SLP) are adopted. Additionally, s~~Sky conditions (cloud amount) and hourly
295 precipitation are also selected to remove the ~~influence~~ records of extensive cloud cover and
296 precipitation ~~when deriving AOD.~~

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

$$306 \quad VISD = VIS / (0.26 + 0.4285 * \log(100 - RH)) \quad \text{Eq. 2}$$

307 where VISD is the dry visibility.

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

310 **2.3 Boundary layer height**

311 The hourly boundary layer height (BLH) from 1980 to 2021 is available from the Fifth Generation
312 reanalysis of the European Medium-Range Weather Forecast Center (ERA5) with a resolution of
313 $0.25^\circ \times 0.25^\circ$ (<https://cds.climate.copernicus.eu>), which is the successor of ERA-Interim and has

314 undergone various improvements (Hersbach et al., 2020). The atmospheric boundary layer is the
315 layer closest to the Earth's surface and exhibits complex turbulence activities, and its height
316 undergoes significant diurnal variation. The effects of the boundary layer on aerosols are mainly
317 manifested in vertical distribution, concentration changes, transport, and deposition (Ackerman et
318 al., 1995). The characteristics and variations in the boundary layer play a crucial role in regulating
319 and adjusting the distribution of atmospheric aerosols. The boundary layer height serves as an
320 approximate measure of the scale height for aerosols (Zhang et al., 2020).

321 Compared to observations of 300 stations over world from 2012 to 2019, the BLH of ERA5 was
322 underestimated by 131.96m. Compared with the underestimated MERRA-2 (166.35m), JRA-55
323 (351.49m), and NECP-2 (420.86m), the BLH of ERA5 was closest to the observations ~~The BLH of~~
324 ~~ERA5 is considered to be the more promising dataset compared to the MERRA-2, JRA-55, and~~
325 ~~NCEP-2 datasets~~ (Guo et al., 2021). The BLH hourly data is temporally and spatially matched with
326 the meteorological ASOS stations data before calculating the daily average.

327 ~~Because the inverse of visibility is proportional to the extinction coefficient and positively related~~
328 ~~to AOD (Wang et al., 2009), we calculated the reciprocal of visibility (VISI) and the reciprocal of~~
329 ~~dry visibility (VISDI). Due to the influence of boundary layer height on the vertical distribution of~~
330 ~~particles and the atmospheric aerosols are largely distributed in the boundary layer~~ (Zhang et al.,
331 2020), we calculated the product (VISDIB) of the reciprocal of dry visibility and BLH three variables
332 (VISI, VISDI, VISDIB) are increased, shown in Eq. 3: Therefore, the Predictor (Figure 2) is
333 composed of 11 variables (TMP, Td, dT, RH, SLP, WS, VIS, BLH, VISI, VISDI, and VISDIB).

334 2.4 MODIS AOD Products

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

343 MODIS, carried on the Terra and Aqua satellites is a crucial instrument in the NASA Earth
344 Observing System program, which is designed to observe global biophysical processes
345 (Salomonson et al., 1987). The 2,330 km-wide swath of the orbit scan can cover the entire globe
346 every one to two days. MODIS has 36 channels and more spectral channels than previous satellite
347 sensors (such as AVHRR). The spectral range from 0.41 to 15- μm representing three spatial
348 resolutions: 250 m (2 channels), 500 m (5 channels), and 1 km (29 channels). The aerosol retrieval
349 algorithms uses seven of these channels (0.47–2.13 μm) to retrieve aerosol characteristics and uses
350 additional wavelengths in other parts of the spectrum to identify clouds and river sediments.
351 Therefore, it has the ability to characterize the spatial and temporal characteristics of the global
352 aerosol field.

353 The MODIS aerosol product actually takes use of different algorithms for deriving aerosols over
354 land and ocean. The Dark Target (DT) algorithm is applied to densely vegetated areas because the
355 surface reflectance over dark-target areas was lower in the visible channels and had nearly fixed
356 ratios with the surface reflectance in the shortwave and infrared channels (Levy et al., 2007; Levy

357 et al., 2013). The Deep Blue (DB) algorithm was originally applied to bright land surfaces (such as
358 deserts), and later extended to cover all cloud-free and snow-free land surfaces (Hsu et al., 2006;
359 Hsu et al., 2013). MODIS Collection 6.1 aerosol product was released in 2017, incorporating
360 significant improvements in radiometric calibration and aerosol retrieval algorithms.

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

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

371 2.5 Ground-based AOD

372 Ground-based daily-15-minute AOD data are available from the Aerosol Robotic Network
373 (AERONET) Version 3.0 Level 2.0 product at 573-395 stations (Figure 1), which can be downloaded
374 from <https://aeronet.gsfc.nasa.gov>. The AERONET program is a federation of ground-based remote
375 sensing aerosol networks established by NASA and PHOTONS, including many subnetworks (such
376 as AeroSpan, AEROCAN, NEON, and CARSNET). The sun photometer (CE-318) measures
377 spectral sun and sky irradiance in the 340-1020 nm spectral range. When the aerosol loading is low,
378 the error is significant. When the AOD at 440 nm wavelength is less than 0.2, the error is 0.01,
379 which is equivalent to the error of the absorption band in the total optical depth (Dubovik et al.,
380 2002a). The total uncertainty in AOD under cloud-free conditions is less than ± 0.01 for wavelength
381 more than 440 nm, and ± 0.02 for wavelength less than 440 nm (Holben et al., 1998). AERONET
382 has three levels of AOD products: Level 1.0 (unscreened), Level 1.5 (cloud screened), and Level
383 2.0 (cloud screened and quality assured). Compared to Version 2, the Version 3 Level 2.0 database
384 has undergone further cloud screening and quality assurance, which is generated based on Level 1.5
385 data with pre- and post-calibration and temperature adjustment and is recommended for formal
386 scientific research (Giles et al., 2019). AERONET provides AOD products at wavelengths of 440,
387 675, 870, and 1020 nm. The AOD at 440nm and the Ångström index at 440-675nm are used for
388 AOD at 550 nm not provided by AERONET, as shown in Eq. 3. AERONET AOD, as the ‘true’
389 value, is the average of at least two times within 1 hour (± 30 minutes) of Aqua transit time (Wei et
390 al., 2019a).

$$391 \tau_{550} = \tau_{440} \left(\frac{550}{440}\right)^{-\alpha} \quad \text{Eq. 3}$$

392 where τ_{440} and τ_{550} are the AOD at a wavelength of 440nm and 550 nm, and α is the Ångström
393 index.

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

396 2.6 Decision Tree Regression

397 2.6.1 Feature selection

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

410 2.6.2 Data balance

411 ~~When it is clear, the AOD value is small, the variability of AOD is small ($AOD < 0.5$), and the data is~~
412 ~~concentrated near the mean value. When heavy pollution, the AOD value is large ($AOD > 0.5$). Compared~~
413 ~~to clear sky, the AOD sequence will show "abnormal" large values with low frequency, which is the~~
414 ~~imbalance of AOD data. Under good weather conditions (such as clear weather), the observed AOD~~
415 ~~values are concentrated around the average value. Under bad weather conditions (such as heavy haze,~~
416 ~~wildfires, sandstorms), the value values will vary significantly compared to the good weather conditions,~~
417 ~~and the frequency of large AOD value is low. When the AOD time series is observed under both good~~
418 ~~and bad weather conditions, the minority class is large AOD value. This is a phenomenon of data~~
419 ~~imbalance.~~ When dealing with imbalanced datasets, because of the tendency of machine learning
420 algorithms to perform better on the majority class and overlook the minority class, the model can be
421 underfit (Chuang and Huang, 2023). Data augmentation techniques are commonly employed to address
422 the issue in imbalance data, which applies a series of transformations or expansions to generate new
423 training data, thereby increasing the diversity and quantity of the training data.

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

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

436 2.6.3 Decision Tree Regression Model

437 The decision tree is a machine learning algorithm based on a tree-like structure used to solve

438 classification and regression problems. We adopt the CART algorithm to construct a regression tree by
 439 analyzing the mapping relationship between object attributes (Predictors) and object values (Target). The
 440 internal nodes have binary tree structures with feature values of "yes" and "no". In addition, each leaf
 441 node represents a specific output for a feature space. The advantages of the regression tree include the
 442 ability to handle continuous features and the ease of understanding the generated tree structure (Teixeira,
 443 2004; Steinberg and Colla, 2009). Before training the tree model, the variables (Input) are normalized to
 444 improve model performance, and after prediction, the results are obtained by denormalization. The 10-
 445 fold cross-validation method is employed to improve the generalization ability of the model (Browne,
 446 2000).

447 The core problems of the regression tree need to be solved are to find the optimal split variable and
 448 optimal split point. The optimal split point of Predictors is determined by the minimum MSE, which in
 449 turn determines the optimal tree structure. We set $Y = [y_1, y_2, \dots, y_N]$ as the Target. We set $X =$
 450 $[x_1, x_2, \dots, x_N]$ 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
 451 N is the length of sample. We set a training dataset as $D = [(x_1, y_1), (x_2, y_2), \dots, (x_N, y_N)]$.

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

$$455 \quad f(x) = \sum_{m=1}^M c_m I(x \in R_m), m = 1, 2, \dots, M \quad \text{Eq. 4}$$

456 where I is the indicator function (Eq. 8.5):

$$457 \quad I = \begin{cases} 1, & x \in R_m \\ 0, & x \notin R_m \end{cases} \quad \text{Eq. 5}$$

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

$$462 \quad \widehat{c}_m = \text{ave}(y_i | x_i \in R_m) \quad \text{Eq. 6}$$

463 A heuristic method is used to split the feature space in CART. After each split, all values of all features
 464 in the current set are examined individually, and the optimal one is selected as the split point based on
 465 the principle of minimum sum of the square errors. The specific step is described as follows: for the
 466 training dataset D , we recursively divide each region into two sub domains and calculate the output
 467 values of each sub domain; then, construct a binary decision tree. For example, split variable is x^j and
 468 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
 469 solve the loss function $L(j, s)$ to find the optimal j and s .

$$470 \quad L(j, s) = \sum_{x_i \in R_1(j, s)} (y_i - c_1)^2 + \sum_{x_i \in R_2(j, s)} (y_i - c_2)^2 \quad \text{Eq. 7}$$

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

$$473 \quad \min_{j, s} \left[\min_{c_1} \sum_{x_i \in R_1(j, s)} (y_i - c_1)^2 + \min_{c_2} \sum_{x_i \in R_2(j, s)} (y_i - c_2)^2 \right] \quad \text{Eq. 8}$$

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

$$476 \quad \widehat{c}_1 = \text{ave}(y_i | x_i \in R_1(j, s)), \widehat{c}_2 = \text{ave}(y_i | x_i \in R_2(j, s)) \quad \text{Eq. 9}$$

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

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

$$481 \quad f(x) = \sum_{m=1}^M \widehat{c}_m I(x \in R_m), m = 1, 2, \dots, M \quad \text{Eq. 10}$$

482 2.7 Gridding method

483 Kriging is a regression algorithm to model and predict (interpolate) random processes/fields based on the
 484 covariance function, which is widely used in geo-statistics (Pebesma, 2004). Ordinary Kriging is the
 485 earliest and most extensively studied form of Kriging. It is a linear estimation system applicable to any
 486 intrinsic stationary random field that satisfies the assumption of isotropy. The two key parameters of
 487 Ordinary Kriging are the semi-variogram function and the weight factors (Goovaerts, 2000). It has been
 488 widely applied in fields, such as climatology, environmental science, and agriculture (Lapen and Hayhoe,
 489 2003; Chen et al., 2010), due to high accuracy, stability, and insensitivity to data shape and distribution.
 490 This study utilizes area-weighted ordinary kriging algorithm to estimate the unknown values of AOD at
 491 specific locations to generate gridded AOD. The longitude range is between ~~-179.5~~180° E and 180° E,
 492 the latitude range is between -60° N and 85° N, and the spatial resolution is 0.5°*0.5°.

493 2.8 Evaluation metrics

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

$$497 \quad RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (y_i - \widehat{y}_i)^2} \quad \text{Eq. 11}$$

$$498 \quad MAE = \frac{1}{n} \sum_{i=1}^n |y_i - \widehat{y}_i| \quad \text{Eq. 12}$$

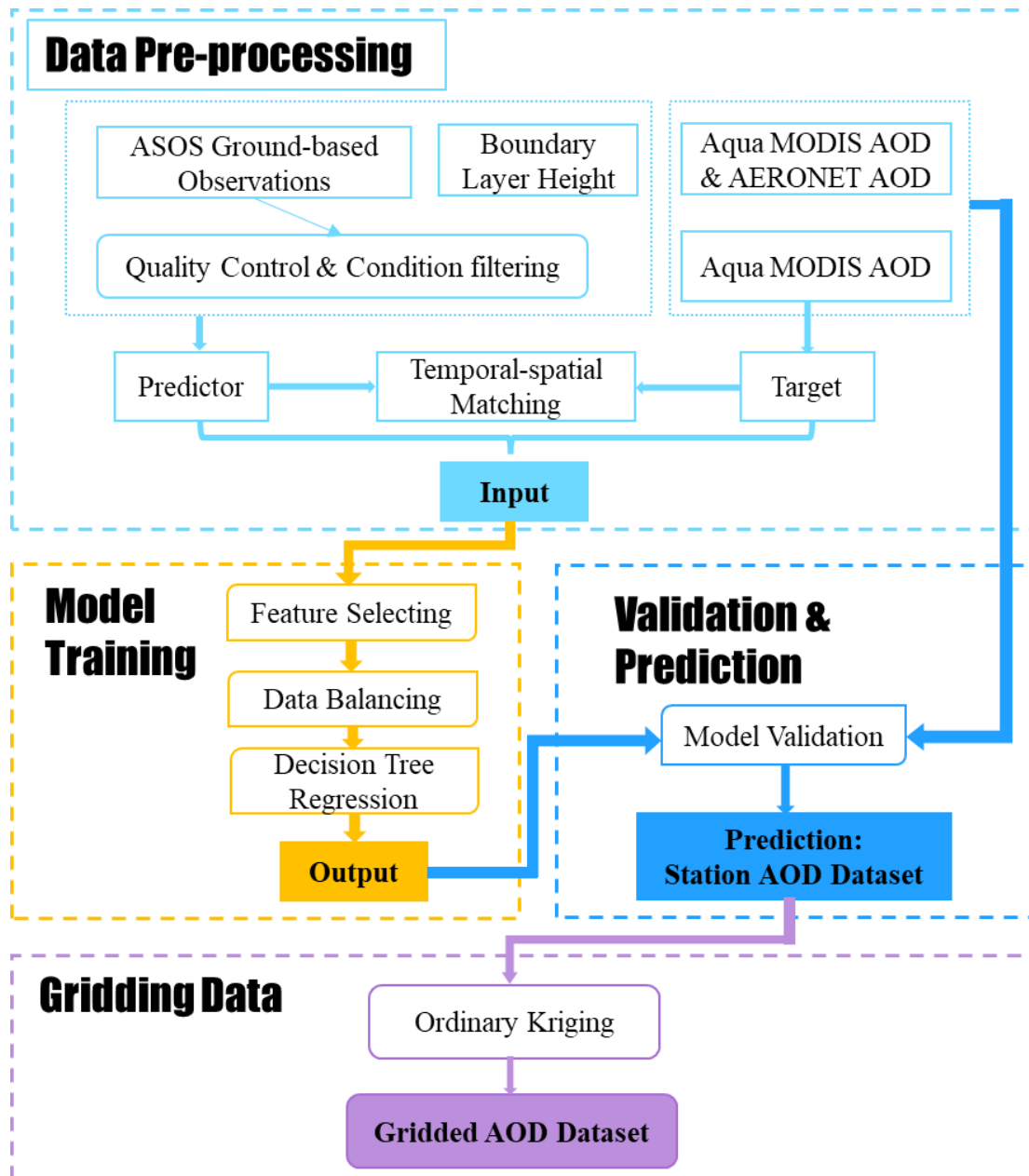
$$499 \quad R = \frac{\sum_{i=1}^n (y_i - \bar{y})(\widehat{y}_i - \bar{\widehat{y}})}{\text{sqrt}(\sum_{i=1}^n (y_i - \bar{y})^2 \sum_{i=1}^n (\widehat{y}_i - \bar{\widehat{y}})^2)} \quad \text{Eq. 13}$$

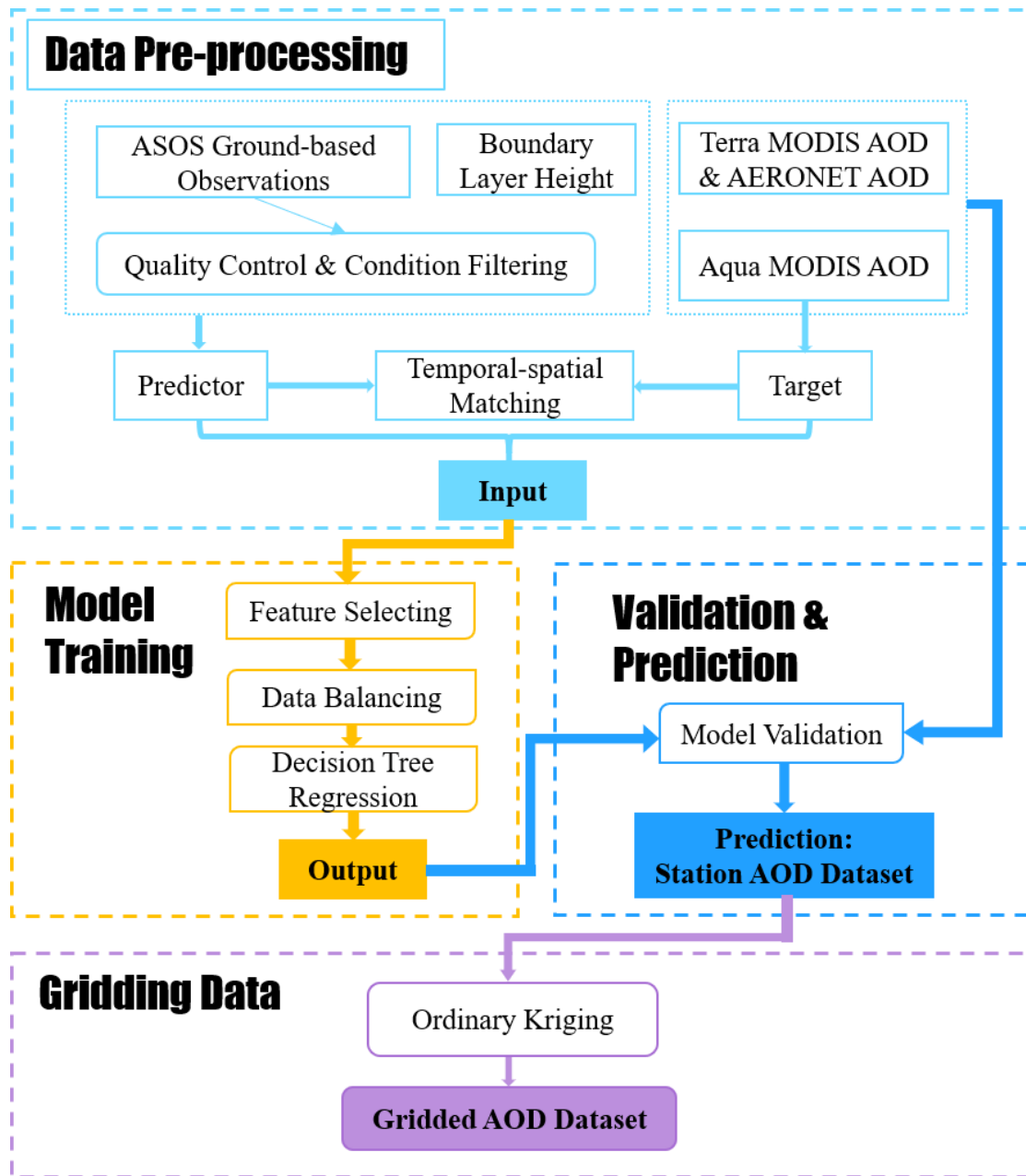
500 where y_i and \bar{y} are the predicted value and the average of the predicted values. \widehat{y}_i and $\bar{\widehat{y}}$ are
 501 the target and the average of the target. $i = 1, 2, \dots, n$. n is the length of sample.

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

$$503 \quad EE = \pm(0.05 + 0.15 * \tau_{target}) \quad \text{Eq. 14}$$

504 where τ_{target} is AERONET AOD or Terra MODIS AOD at 550nm.





506

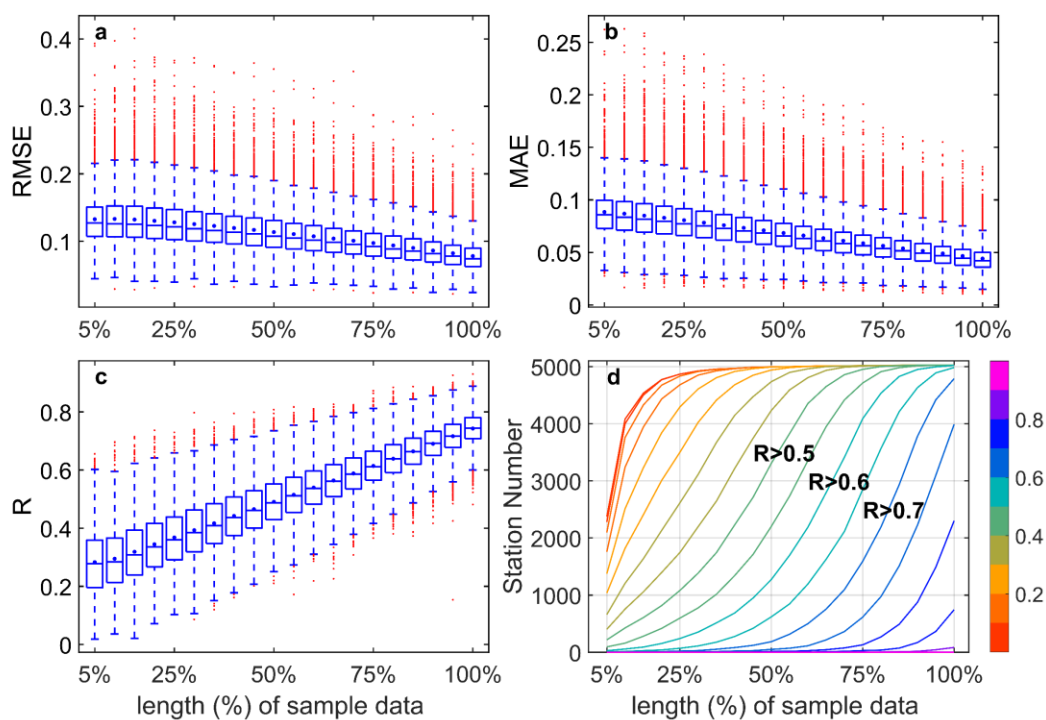
507 **Figure 2** Flowchart for deriving aerosol optical depth (AOD).

508 2.9 Workflow

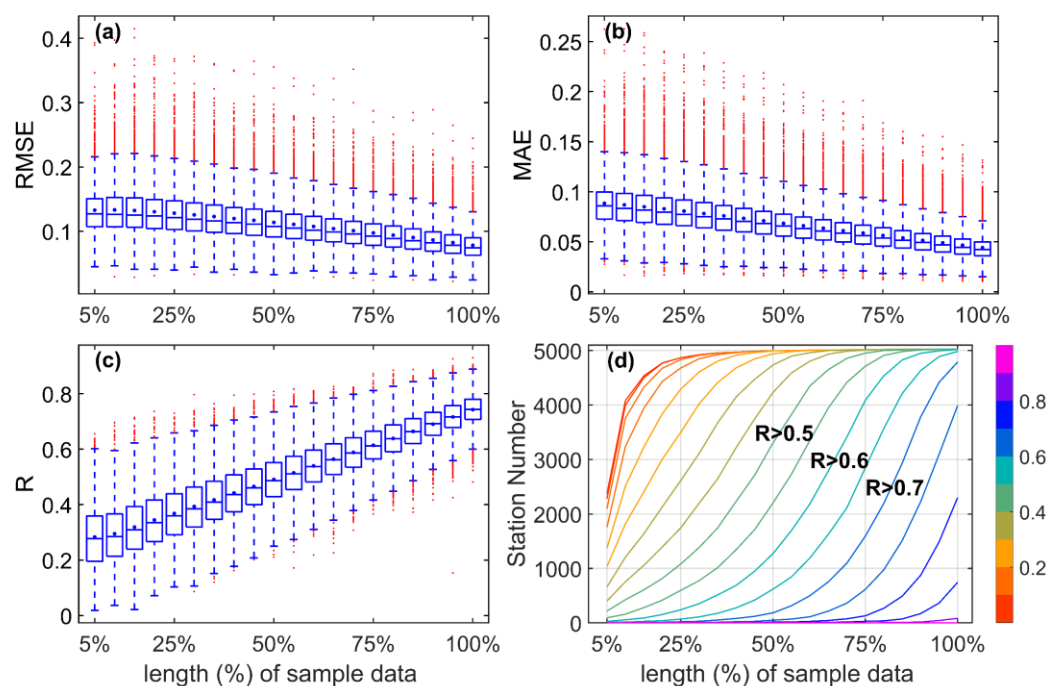
509 **Figure 2** is the summarized flowchart and provides an overview of the structure of this
 510 study, which involves four main parts: (1) data preprocessing, (2) model training, (3) validation and
 511 prediction, and (4) data gridding.

512 3 Results and discussion

513 **3.1 Dependence of model performance on training data length**
 514 **Examination of the model performance**



515



516

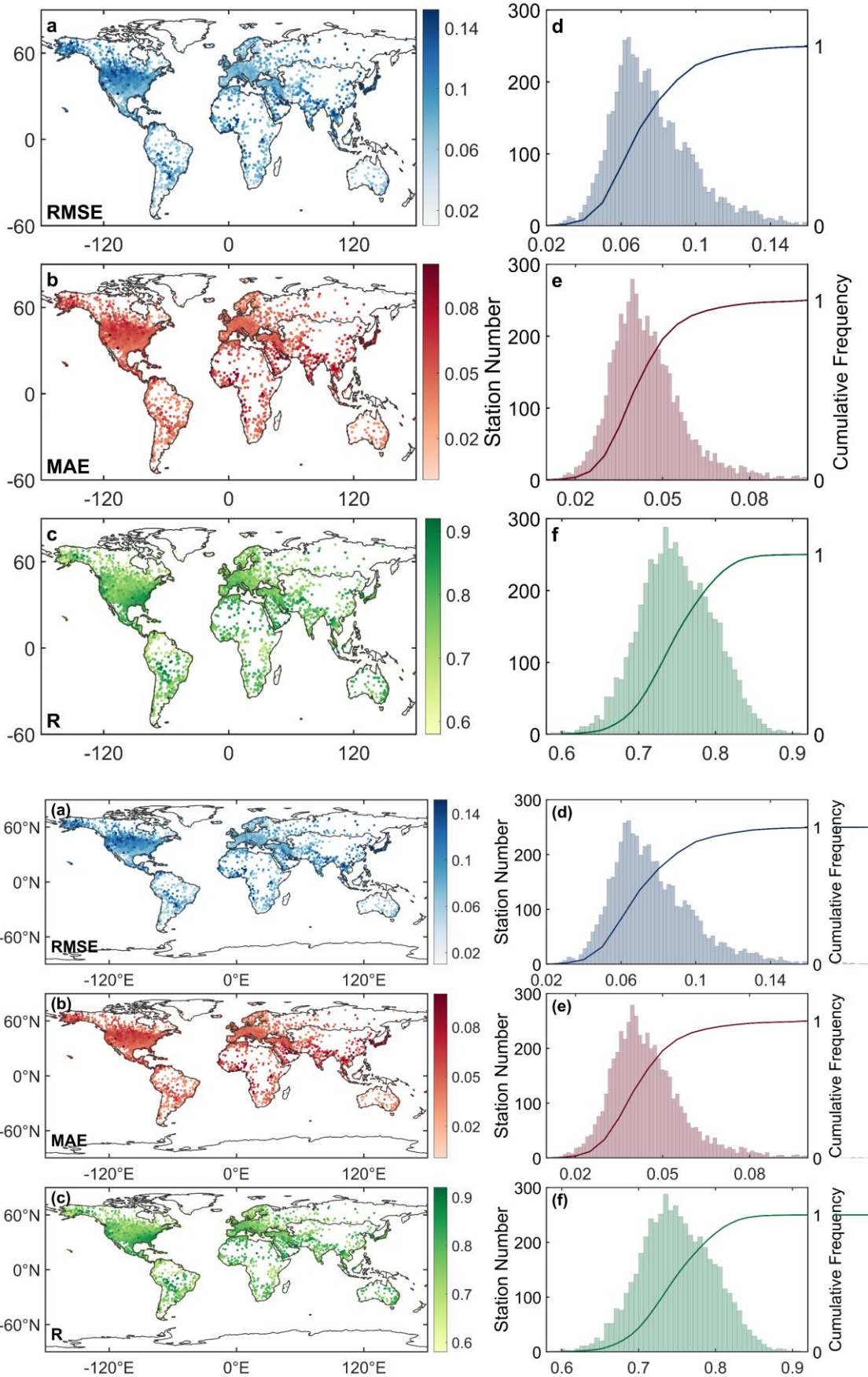
517 **Figure 3** Boxplots of root mean squared error (RMSE) (a), mean absolute error (MAE) (b),
 518 and correlation coefficient (R) (c) between predicted values and target using different lengths of
 519 sample data (5% interval) as the training dataset, and the correlation coefficient curve (d) of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

521 We build the models using different lengths of sample data (5% to 100%, with a 5% interval) by random
 522 allocation without overlap and evaluate the predictive performance of each model. **Figure 3**

523 depicts RMSE(a), MAE(b), and R (c) between the predicted values and target based on the training data
524 of 5% to 100% sample data at a station. As the volume of the training data increases, the RMSE and
525 MAE decrease, and the correlation coefficient increases. Compared to 5% of the sample data, the result
526 of 100% sample data shows a decrease in RMSE by 41.1%, a decrease in MAE by 50.1%, and an increase
527 in R by 162.3%. The relationship between the length of sample data and the model's performance is
528 positive for each station. ~~Figure 3~~ Figure 3 (d) shows that R of approximately 70% stations is greater
529 than 0.5 at 50% of the sample data, while at 75%, the R of approximately 80% of stations is greater than
530 0.6. When 100% of the sample data is used as sample data, the R of approximately 80% of stations is
531 greater than 0.75, and the R of about 97% is greater than 0.7. This finding indicates that the predictive
532 capability and robustness of the model increase as the amount of training data increases. It may be
533 attributed to the model's ability to capture more complex patterns and relationships among the input by
534 multi-year data.

535 **3.2 Evaluation of model ~~error~~training**

536 ~~The more sample data input, the better the model performs. Therefore, 100% of the sample data were~~
537 ~~used as training data.~~ Figure 4 Figure 4 shows the spatial distribution (a-c) and frequency and cumulative
538 frequency (d-e) of RMSE, MAE, and R of all stations. The mean values of RMSE, MAE, and R are 0.078,
539 0.044, and 0.750, respectively. The RMSE of 93% stations is less than 0.11, the MAE of 91% is less than
540 0.06, and the R of 88% is greater than 0.7. The R values in Africa, Asia, Europe, North America, Oceania,
541 and South America are 0.763, 0.758, 0.736, 0.750, 0.759, and 0.738, respectively. Although the RMSE
542 and MAE of a few stations are high in America and Asia, the R is still high (>0.6). Therefore, the results
543 of the model's errors demonstrate that the model performs well on almost all stations.



544

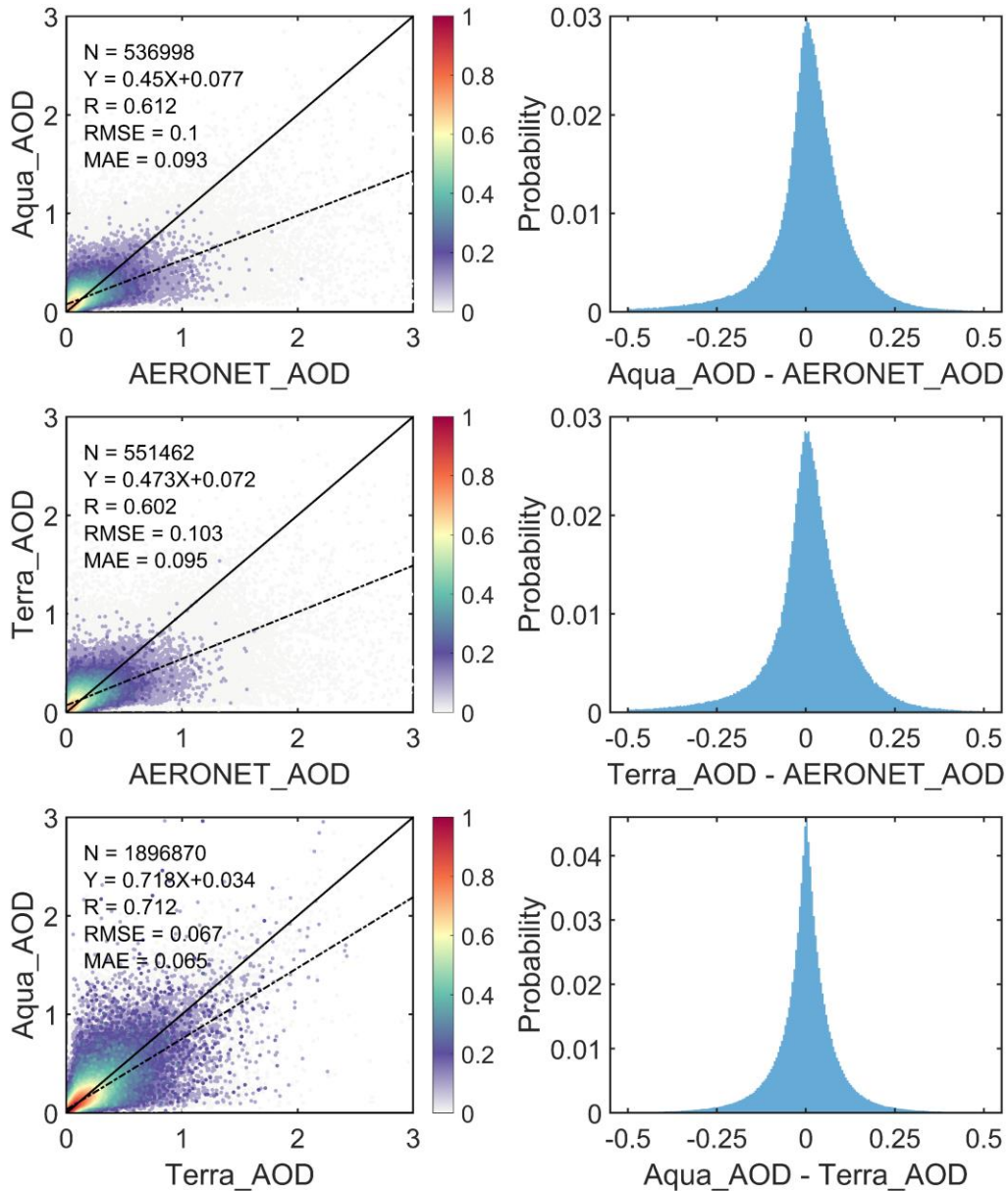
545

546 **Figure 4** Figure 4 Spatial distribution (a-c) of root mean squared error (RMSE), mean absolute

547 error (MAE), and correlation coefficient(R) between the model's result and target with 100% sample
548 data. Station number (bar) and cumulative frequency (curve) (d-e) of RMSE, MAE, and R.

549 **3.3 Validation and comparison with MODIS and AERONET AOD**
550 **Validation of derived AOD against MODIS and AERONET AOD**

551 ~~First, the relationship among daily MODIS and AERONET AOD is evaluated. Figure 5 presents the~~
552 ~~scatter density plots (the left column) and bias probability distribution (the right column) among daily~~
553 ~~Aqua, Terra and AERONET AOD. The R, RMSE, and MAE of 536,998 data couples between Aqua AOD~~
554 ~~and AERONET AOD are 0.612, 0.1, and 0.093, respectively. Then, 86.8% of the data have a bias within~~
555 ~~± 0.093 . The R, RMSE, and MAE of 551,462 data couples between Terra AOD and AERONET AOD are~~
556 ~~0.602, 0.103, and 0.095, respectively. Then 86% of the data have a bias within ± 0.095 . The R, RMSE,~~
557 ~~and MAE of 1,896,870 data couples between Aqua AOD and Terra AOD are 0.712, 0.067, and 0.065,~~
558 ~~respectively, and the bias is within ± 0.065 for 92% of the data. On the global scale, the AOD retrieved~~
559 ~~by satellites may be overestimated at low AOD levels and underestimated at high AOD levels compared~~
560 ~~to AERONET AOD. Approximately 86% of the bias values are less than the MAEs. Terra and Aqua have~~
561 ~~good consistency, although one is for morning transit and the other is for afternoon transit. Finally, 92%~~
562 ~~of the data bias are less than the MAEs. Thus, there is good consistency among them on the daily scale.~~



563

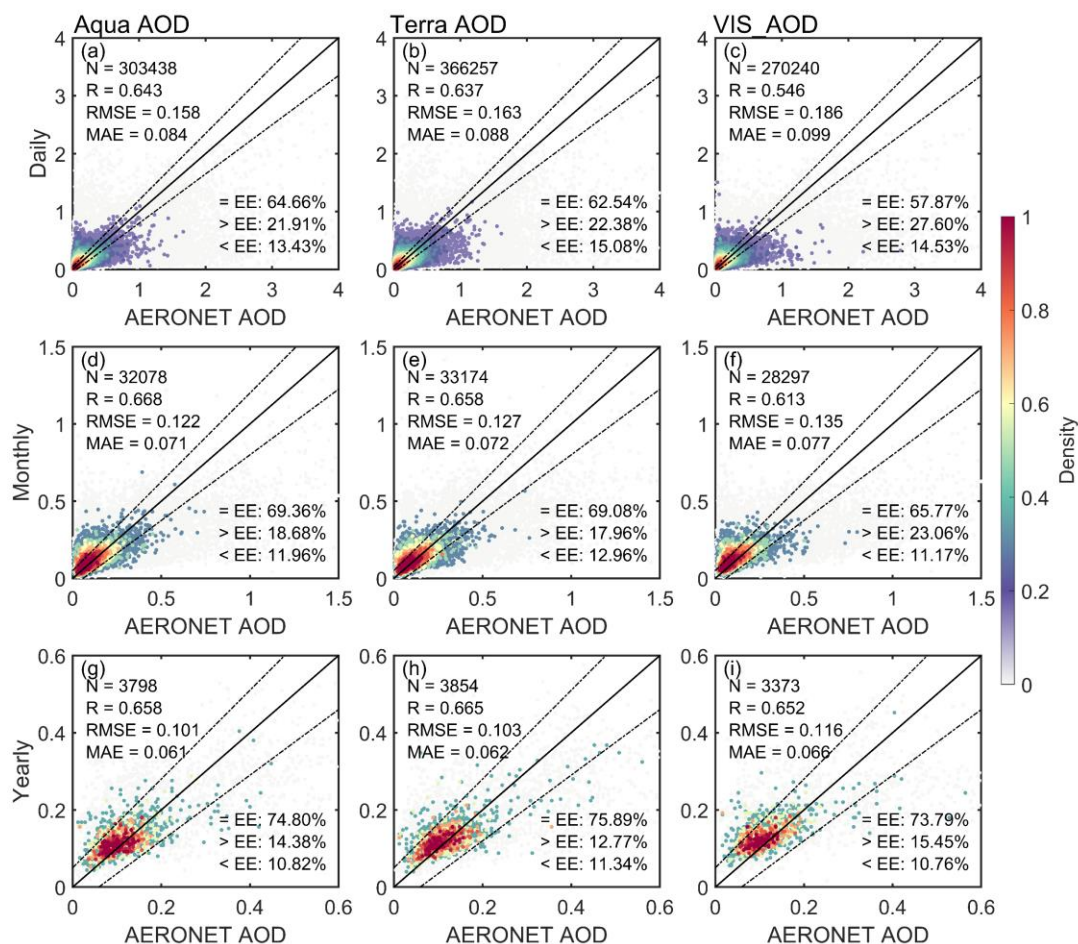
564 **Figure 5** Scatter density plots and bias probability between Aqua AOD, Terra AOD and AERONET
 565 AOD at a daily scale. The solid black line represents the 1:1 line and the dashed black line is the
 566 linear regression line.

567 **3.3.1 Validation over global land**

568 To validate the model's predictive ability, the visibility-derived AOD (for short, VIS_AOD) is compared
 569 with Aqua, Terra and AERONET AOD at 550nm for the global scale ~~other observed data for daily,~~
 570 ~~monthly, and yearly scales of Aqua, Terra and AERONET AOD.~~ Among them, Aqua AOD has been used
 571 as training data, which is not independent. Terra AOD and AERONET AOD have not been used as
 572 training data and can be regarded as independent data.

573 First, the relationship among daily MODIS and AERONET AOD is evaluated. Figure 5 shows the scatter
 574 density plots between AERONET AOD and Aqua AOD (a, d, g) and Terra AOD (b, e, h). The R values

575 with Aqua AOD and Terra AOD are 0.643 and 0.637 on the daily scale, and 0.668 and 0.658 on the
 576 monthly scale, 0.658 and 0.665 on the yearly scale. The RMSE with Aqua AOD and Terra AOD are 0.158
 577 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
 578 scale. The MAE values with Aqua AOD and Terra AOD are 0.084 and 0.088 on the daily scale, and 0.071
 579 and 0.072 on the monthly scale, 0.061 and 0.062 on the yearly scale. The percentages of sample point
 580 falling within the EE envelopes are 64.66% and 62.54% on the daily scale, and 69.36% and 69.08% on
 581 the monthly scale, 74.80% and 75.89% on the yearly scale.



582

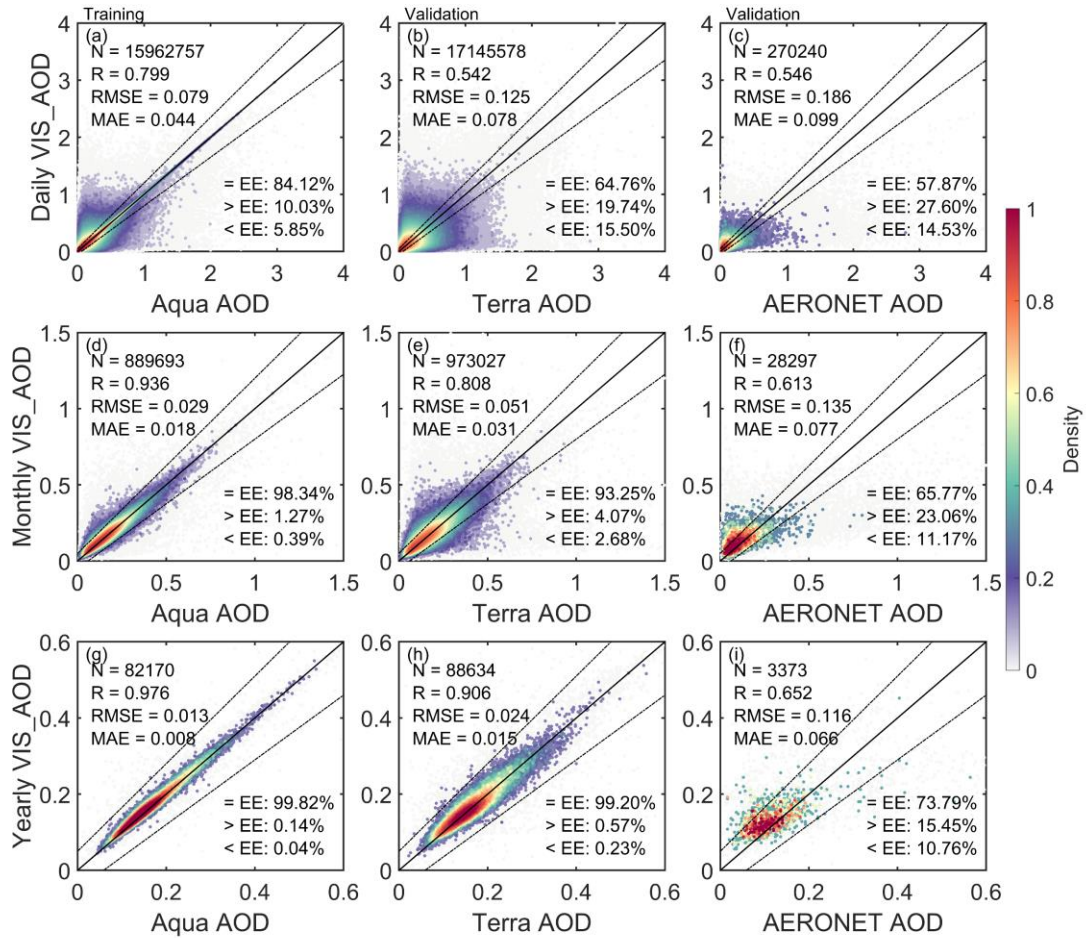
583 **Figure 5** Scatter density plots between AERONET AOD (550nm) and Aqua MODIS AOD, Terra MODIS
 584 AOD and VIS AOD at the daily (a-c), monthly (d-f) and yearly (g-i) scale. The solid black line represents
 585 the 1:1 line and the dashed lines represents expected error (EE) envelopes. The sample size (N),
 586 correlation coefficient (R), mean absolute error (MAE), and root mean square error (RMSE) are given.
 587 ‘=EE’, ‘>EE’, and ‘<EE’ represent the percentages (%) of retrievals falling within, above, and below
 588 the EE, respectively. The matching time for Aqua AOD and VIS_AOD with AERONET AOD is 13.30
 589 (± 30 minutes) at local time, and the matching time between Terra AOD and AERONET AOD is 10.30
 590 (± 30 minutes) at local time.

591 Figure 6 shows the scatter density plots and the EEs between VIS_AOD and Aqua AOD, Terra AOD,
 592 and AERONET AOD. Aqua AOD is not an independent validation, and Terra and AERONET AOD are
 593 independent validation. For the daily scale, the R, RMSE and MAE of between VIS_AOD and Aqua
 594 AOD (15,962,757 pairs data) is 0.799, 0.079 and 0.044, respectively. The percentage of sample point

595 falling within the EE envelopes is 84.12% on the global scale (Figure 6 a). The R between VIS_AOD
596 and Terra AOD (17,145,578 pairs data) is 0.542, with a RMSE of 0.125 and MAE of 0.078. The
597 percentage falling within the EE envelopes is 64.76% (Figure 6 b). The R between VIS_AOD and
598 AERONET AOD (270,240 pairs data) at 397 sites is 0.546, with a RMSE of 0.186 and MAE of 0.099.
599 The percentage falling within the EE envelopes is 57.87% (Figure 6 c).

600 For the monthly and annual scales, RMSE and MAE show a significant decrease between VIS_AOD and
601 Aqua, Terra, and AERONET AOD, and R and percentages falling within EE show a significant increase
602 in Figure 6 (d-i). The monthly RMSEs are 0.029, 0.051, and 0.135, the monthly MAEs are 0.018, 0.031,
603 and 0.077, and the R values are 0.936, 0.808, and 0.613, respectively. The percentages falling within the
604 EE envelopes are 98.34%, 93.25%, and 65.77%. The RMSEs at the annual scale are 0.013, 0.024, and
605 0.116, the MAEs are 0.008, 0.015, and 0.066, and the R values are 0.976, 0.906, and 0.652, respectively.
606 The percentages falling within the EE envelopes are 99.82%, 99.20%, and 73.79%. The percentage
607 falling within the EE envelopes against AERONET is smaller than that against Terra, which may be
608 related to the elevation of AERONET sites, the distance between AERONET and meteorological stations,
609 and observed time. The results highlighted above demonstrate a clear improvement in performance on
610 the monthly and annual scales compared to the daily scale (Schutgens et al., 2017), which provided a
611 foundation for the gridded dataset.

612 On the daily, monthly, and annual scales, compared with AERONET AOD, the correlation coefficients,
613 RMSE, MAE, and percentages falling within the expected error of VIS_AOD and MODIS AOD are very
614 close. Since the time of AERONET AOD and VIS_AOD overlaps before 2000, it indicates that
615 VIS_AOD also has the same accuracy.



616

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

624 3.3.2 Validation over regions

625 Aerosol loading exhibits spatial variability. Evaluation metrics for the relationships between
 626 visibility-derived AOD and AERONET AOD and Terra AOD for each region are listed in Table 1.
 627 Over Europe and North America, the results are similar to those of Terra and AERONET, with a
 628 large number of data pairs, greater than 10^5 (AERONET) and greater than 10^7 except for Eastern
 629 Europe (Terra) on the daily scale. Approximately 63% -70% fall within the EE envelopes. The
 630 RMSE is approximately 0.1100, except for western North America, the MAE is approximately
 631 0.0700, with a correlation coefficient between 0.44 and 0.54.

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

636 coefficient in South America at 0.740.

637 In Asia, India, and West Africa, the data pairs are only approximately 10⁴ (AERONET). 32% to 50%
638 fall within the EE envelopes compared to AERONET, the RMSE ranges from 0.2 to 0.5, and the
639 MAE ranges from 0.11 to 0.36. 51 to 58%, compared to Terra, fall within the EE envelopes, the
640 RMSE is around 0.16, and the MAE is around 0.11. Compared to AERONET, in these high aerosol
641 loading regions, RMSE and MAE increase, and the percentages falling within the EE envelopes
642 decrease, but the correlation coefficients do not significantly decrease.

643 Compared to Terra AOD, 55% -67% of data falls within the EE envelopes on the daily scale, 87% -
644 96% on the monthly scale, and over 97% on the yearly scale. Compared to AERONET AOD, 32-
645 68% of data falls within the EE envelopes, 24% -84% on the monthly scale, and 15% -97% on the
646 yearly scale. On both monthly and yearly scales, all metrics have shown a significant increase in
647 performance when compared to Terra. However, compared to AERONET, not all metrics increase
648 in some regions due to limited data pairs, such as West Africa, Northeast Asia, and India, which may
649 be due to the spatial differences between AERONET sites and meteorological stations.

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

654 **3.3.3 Validation at a site scale**

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

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

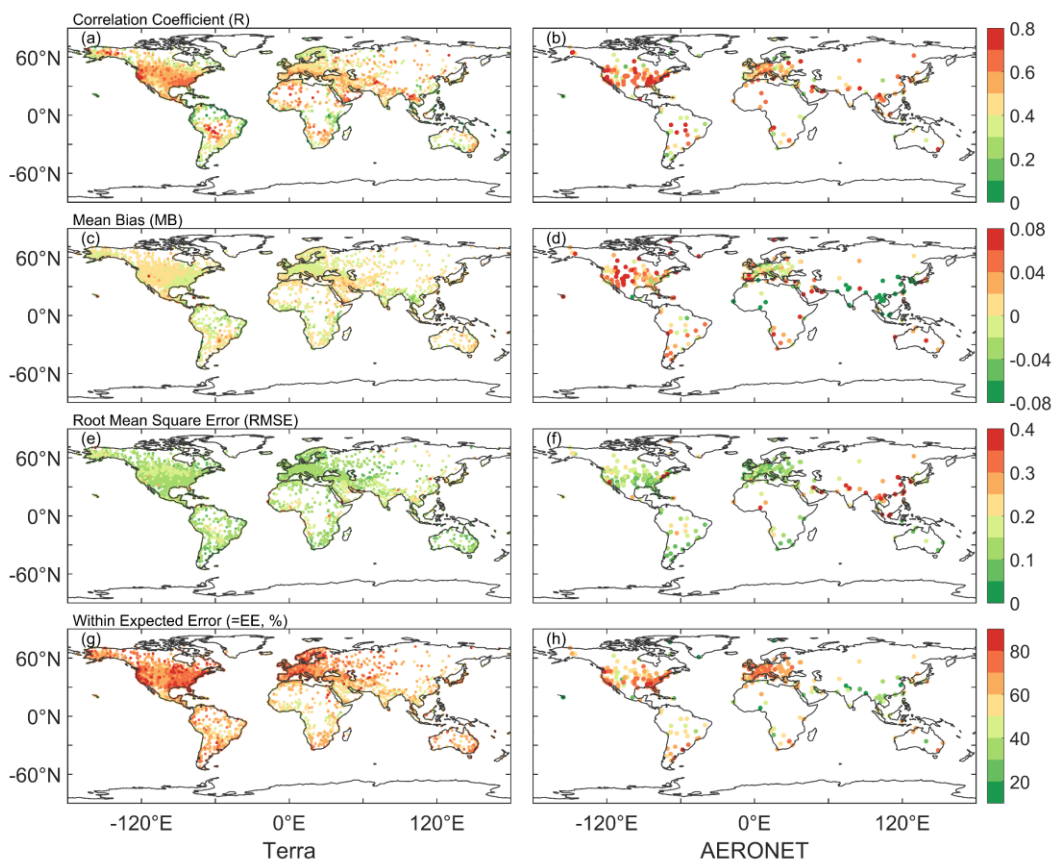
Table 1 Evaluation metrics for the relationships between visibility-derived AOD and AERONET AOD and Terra AOD for each region.

Region	—	<i>N</i>			<i>R</i>			<i>RMSE</i>			<i>MAE</i>			<i>Within EE (%)</i>		
		daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly	daily	monthly	yearly
<i>Eastern Europe</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Western Europe</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Western North America</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Eastern North America</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Central South America</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Southern Africa</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Australia</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Western Africa</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Southeast Asia</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Eastern China</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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
<i>Northeast Asia</i>	<i>AERONET</i>	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
	<i>TERRA</i>	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

662

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

663 less than 0.01, the RMSE of 85% is less than 0.15, and the percentage falling within the EE of 67%
664 is greater than 60%. More than 85% of stations fall within the EE is greater than 60% in Europe,
665 North America, and Oceania, while 40-60% in South America, Africa, and Asia. The percentage of
666 expected error is low in South and East Asia, and Central Africa, with some underestimation. Above
667 60% in Africa, Asia, North America, and Europe have a correlation coefficient greater than 0.4. The
668 regions with lower correlation are the coastal regions of South America, eastern Africa, western
669 Australia, northeastern North America, and northern Europe. Above 90% of the RMSE in Europe,
670 North America, and Oceania have a correlation coefficient smaller than 0.15. High RMSE regions
671 are in western North America, Asia, central South America, and central Africa.
672 Compared to AERONT daily AOD, the R of 74% stations is greater than 0.4, and the spatial
673 distribution is similar to Terra's. The mean bias of 44% is less than 0.01, the RMSE of 68% is less
674 than 0.15, and the percentage falling within the EE of 53% is greater than 60%. More than 70% of
675 sites have a correlation coefficient greater than 0.4 in Africa, Asia, Europe, and North America.
676 More than 57% of sites have an expected error percentage of over 60% in Europe, North America,
677 and Oceania. Except for Asia. Over 72% of sites have a RMSE less than 0.15. Except for Oceania
678 and South America, over 71% of sites in other regions have MAE less than 0.01. Almost all sites in
679 Asia show a negative bias, significantly underestimating. However, there is a significant
680 overestimation in western North America and western Australia. Most sites in Asia falling within
681 the expected error are less than 50%. High RMSE region are in Asia, India, and central Africa.
682 The validation and comparison on the site scale show a limitation similar to the MODIS DT
683 algorithm. In areas with high vegetation coverage, the AOD from visibility are better than those in
684 bright areas such as deserts.



685

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

689 **3.3.4 Discussion and uncertainty analysis**

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

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

714 The spatial matching between meteorological stations and AERONET sites may cause some biases.
715 AERONET sites are usually not co-located with meteorological stations in terms of elevation and
716 horizontal distance, this is another reason for the weak correlation between VIS_AOD and
717 AERONET AOD. The meteorological stations are located at the airport. Different horizontal
718 distances may result in meteorological stations and AERONET sites being located on different
719 surfaces (such as urban, forest, mountainous). Differences in site elevation significantly impact the
720 relationship between AOD and measured visibility. When the AERONET site is at a higher elevation
721 than the meteorological station, there may be fewer measurements of aerosols over the sea at the
722 AERONET site.

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

727 **3.3.4.1 Uncertainty with pollution level**

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

735 **3.3.4.2 Uncertainty with elevation of AERONET site**

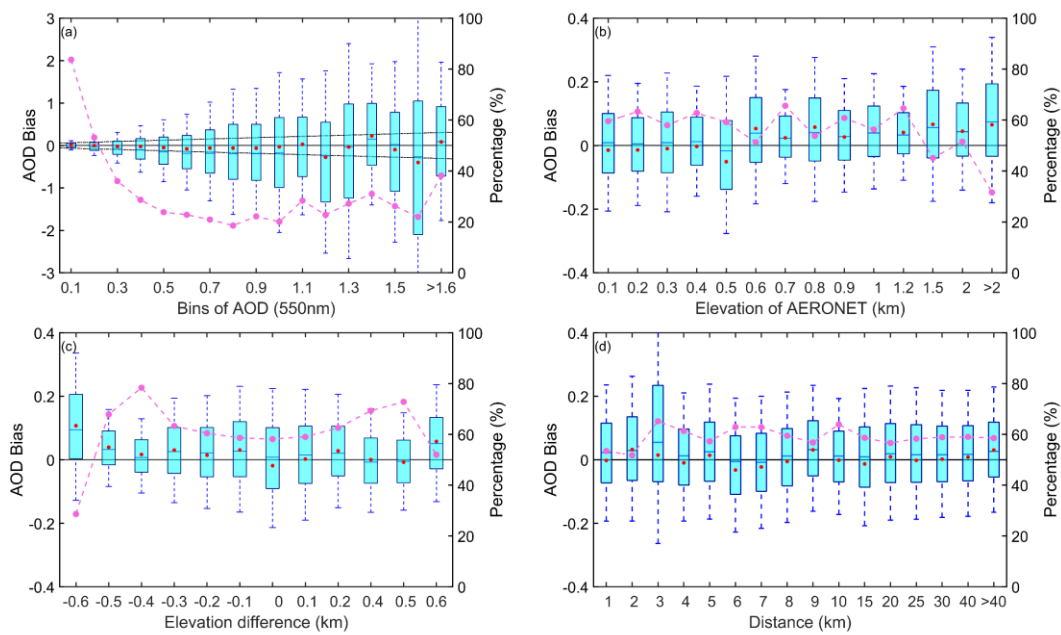
736 The contribution of particulate matter near the ground to the column aerosol loading is significant.
737 The elevation of the site affects the measurement of column aerosol loading in Figure 8 (b). There
738 is a negative bias in the low elevation (<=0.5km) with a percentage of 60%-64% falling within the
739 EE envelopes and a positive bias in high elevation (0.5-1.2km) with a percentage of 50%-65%
740 falling within the EE envelopes. The percentage significantly decreases (>1.2km), and the average
741 bias increases. Therefore, the elevation of AERONET's site will cause bias in validation, and, the
742 uncertainty greatly increases in high elevation.

743 **3.3.4.3 Uncertainty with elevation of meteorological station**

744 Due to the elevation difference between the meteorological station and AERONET site in the
745 vertical direction, the uncertainty caused by elevation differences of site was analyzed in Figure 8
746 (c). When the elevation difference is negative (the elevation of the meteorological station is lower
747 than that of the AERONET station), there is a significant positive bias. When the difference is
748 positive, the mean bias approaches 0 or is positive. The percentage is greater than 60% (-0.5 km-
749 0.5km). The positive mean bias is greater than the negative mean bias, and the uncertainty greatly
750 increases when the elevation of meteorological stations is lower than that of AERONET sites. It
751 indicates that the contribution of the near surface aerosol to the column aerosol loading is significant
752 and cannot be ignored.

753 **3.3.4.4 Uncertainty with distance between meteorological station and AERONET site**

754 The spatial variability of aerosols is significant. Meteorological stations and AERONET sites are
755 not collocated, resulting in a certain distance in spatial matching. In this study, the upper limit of
756 distance is 0.5 degree. Figure 8 (d) shows the error of the distance between stations, where the
757 degree is converted to the distance at WGS84 coordinates. The bias does not change significantly
758 with increasing distance. The average bias is around 0, with the maximum positive mean bias
759 (0.0322) at a distance of 2km and the maximum negative mean deviation (-0.0323) at 6km. The
760 median is almost positive, except at 5km and 6km. The percentage falling within the EE envelopes
761 is over 50%, with the maximum percentage (66%) at 3km and the minimum (62%) at 2km.



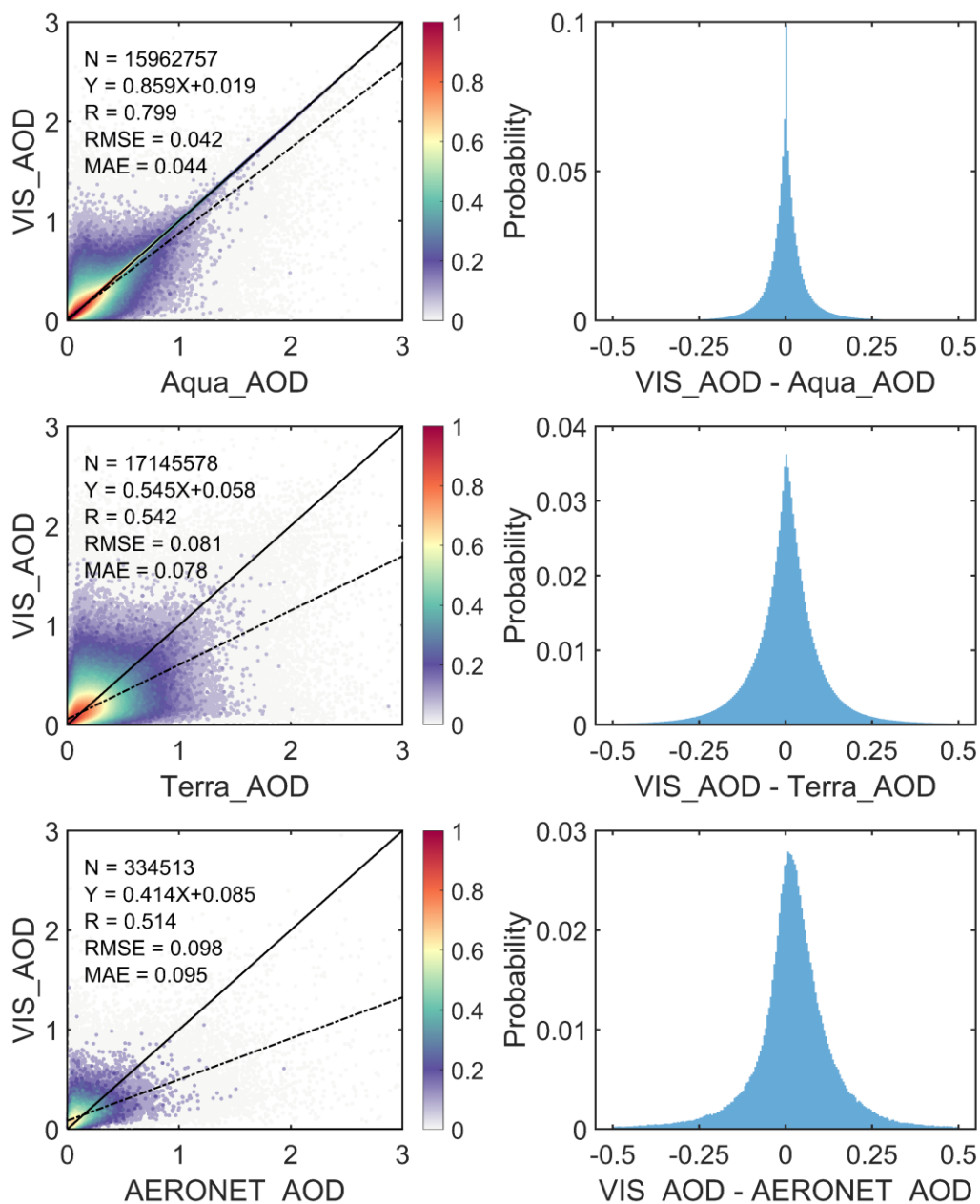
762

763 **Figure 8** Box plots of AOD bias and the percentage falling within the EE envelopes (curves): (a)
 764 AERONET AOD levels, (b) elevation of AERONET sites, (c) elevation difference between
 765 meteorological stations and AERONET sites, (d) distance (km) between meteorological stations and
 766 AERONET sites. The black horizontal line represents the zero bias. For each box, the upper, lower,
 767 and middle horizontal lines, and whiskers represent the AOD bias 75th and 25th percentiles, median,
 768 and 1.5 times the interquartile difference, respectively. The black solid lines represent the EE
 769 envelopes ($\pm(0.05+0.15*AOD_{AERONET})$). No site with a difference of +0.3km (x-axis label without
 770 0.3) in (c).

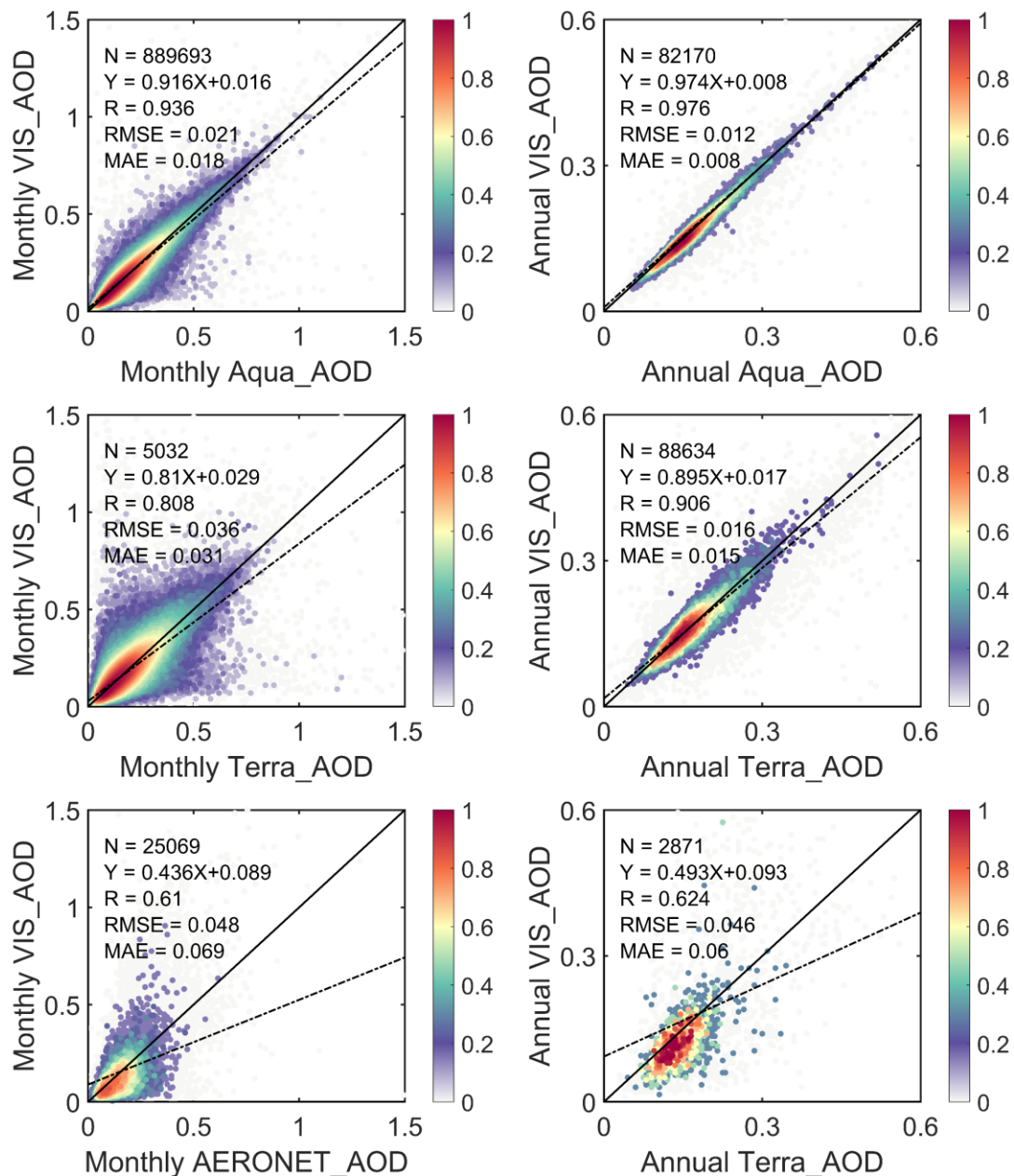
771 –Figure 6 shows the scatter density plots and probability distribution of the bias between daily VIS_AOD
 772 and Aqua AOD, Terra AOD, and AERONET AOD. The R of 15,962,757 pairs data between VIS_AOD
 773 and Aqua AOD is 0.799, higher than the R between AERONET AOD and Aqua AOD, as well as Terra
 774 AOD and Aqua AOD. The RMSE is 0.042 and the MAE is 0.044. Then, 69.7% of the data pairs have a
 775 bias within ± 0.044 , and 69.7% have a bias within ± 0.093 . The R of 17,145,578 pairs of data between
 776 VIS_AOD and Terra AOD is 0.542, the RMSE is 0.081 and the MAE is 0.078. Then, 66.8% of the data
 777 pairs have a bias within ± 0.078 , and 73.3% have a bias within ± 0.095 . The R of 334,513 data pairs
 778 between VIS_AOD and AERONET AOD is 0.514. The RMSE is 0.098 and the MAE is 0.095. Finally,
 779 78.3% of the data pairs have a bias within ± 0.095 –

780 At the monthly and annual scales, RMSE and MAE show a significant decrease between VIS_AOD and
 781 Aqua, Terra, and AERONET AOD, and R shows a significant increase in Figure 7. The monthly RMSEs
 782 are 0.021, 0.036, and 0.048, the monthly MAEs are 0.018, 0.031, and 0.069, and the R values are 0.936,
 783 0.808, and 0.61, respectively. The RMSE values at the annual scale are 0.012, 0.016, and 0.025, the MAE
 784 values are 0.008, 0.015, and 0.006, and the R values are 0.976, 0.0906, and 0.624, respectively. The
 785 monthly and annual R is slightly higher than those in previous studies (Wang et al., 2009; Wu et al., 2014;
 786 Zhang et al., 2017). In addition to the differences between models, it may also be related to the calculation
 787 method of daily average visibility and the consideration of boundary layer height–

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



794
 795 **Figure 6** Scatter density plots and bias probability between predicted AOD (VIS_AOD) and Aqua
 796 MODIS AOD, Terra MODIS AOD and AERONET ground-based observations of AOD at the daily
 797 scale. The solid black line represents the 1:1 line and the dashed black line is the linear regression
 798 line.



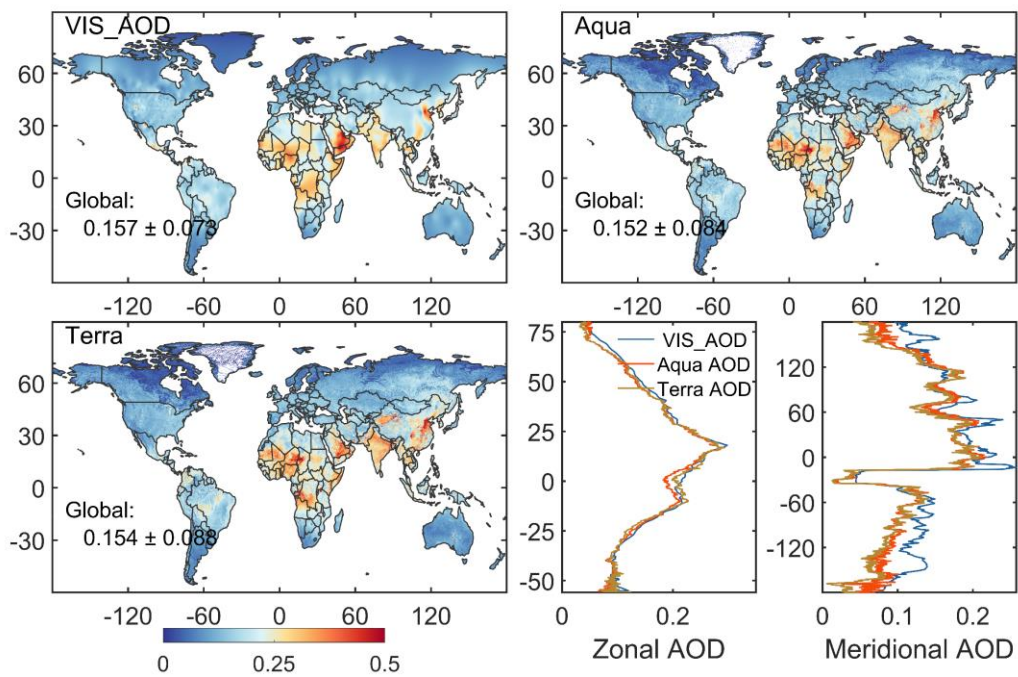
799

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

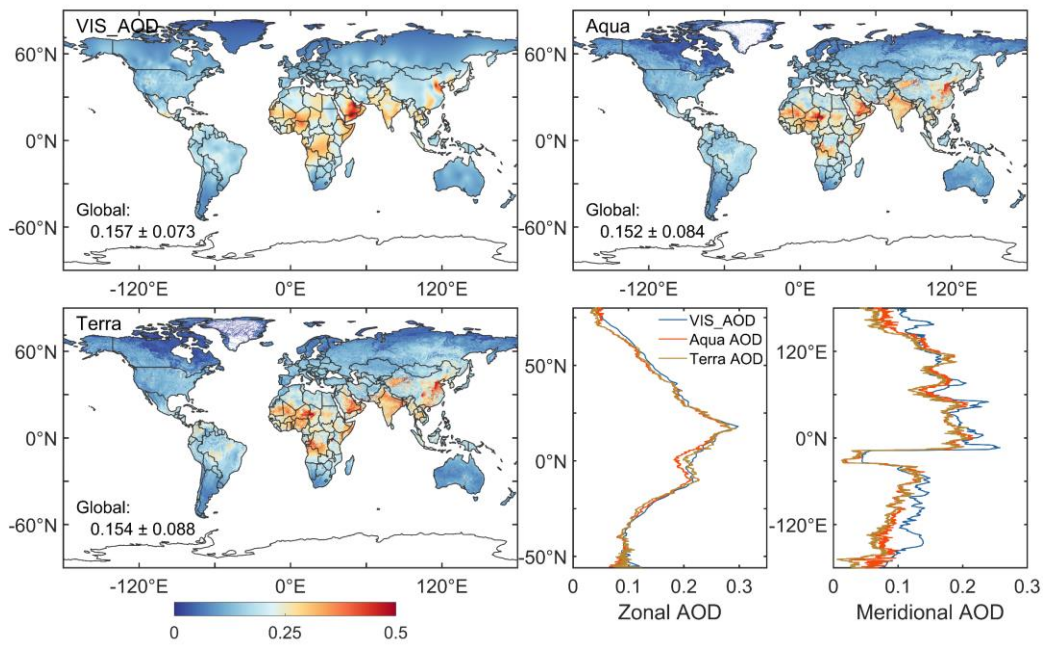
804 **3.4 Evaluation of gGridded visibility-derived AOD**

805 Figure 8-9 shows the gridded AOD based on ordinary kriging interpolation with the area-weighted
 806 method and compares the multi-year spatial, zonal, and meridional distributions of AOD with Aqua
 807 and Terra AOD over land from 2003 to 2021. The VIS_AOD is 0.157 ± 0.073 over land, which is
 808 almost equal to the Aqua (0.152 ± 0.084) and Terra (0.154 ± 0.088) AOD values with relative biases
 809 of 3.3%, and 1.9%, respectively. In order to compare the spatial correlation, Aqua and Terra MODIS
 810 AOD are averaged to the 0.5-degree resolution. In the heatmap (Figure 9 Figure 10), the R of

811 VIS_AOD and Aqua AOD is 0.7988, the RMSE is 0.049 with a bias of 32% compared to the mean,
812 and the MAE is 0.008, with a bias of 5% compared to the mean. Compared to Terra AOD, the R is
813 0.7879, and the RMSE is 0.051, with a bias of 33% compared to the mean, and the MAE is 0.005,
814 with a bias of 3% compared to the mean. ~~The R between Aqua and Terra AOD are highly similar,~~
815 ~~with an R of 0.980. By comparing the zonal and meridional distributions of AOD, VIS_AOD is~~
816 ~~consistent with Aqua and Terra AOD, with~~ The R values between VIS_AOD and Aqua and Terra
817 AOD are of 0.9957 and 0.9909 for the zonal distribution and 0.986 and 0.8979 for the meridional
818 distribution, respectively. In the low aerosol loading region, VIS_AOD exhibits a little
819 overestimation. Whether in meridional or zonal distribution, the peak and valley regions are
820 basically consistent (Tian et al., 2023). Due to the limitations of satellite inversion algorithms, a bias
821 appears on the bright surface, especially in northern North America with extensive snow cover
822 (Levy et al., 2013). All above results suggest that the gridded AOD is ~~highly~~ consistent with satellite
823 observations in spatial distribution.

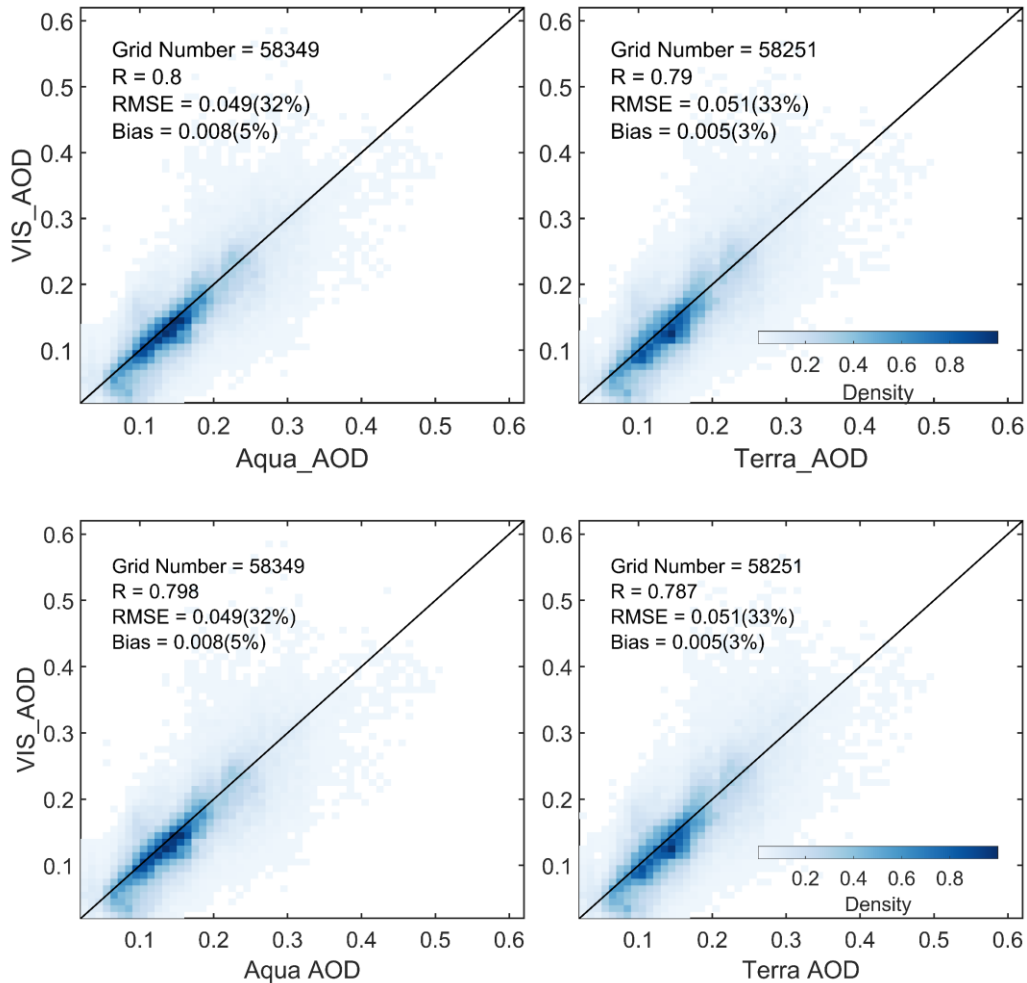


824



825

826 **Figure 8-9** The spatial, zonal and meridional distributions of the multi-year mean VIS_AOD, Aqua
 827 AOD, and Terra AOD over land from 2003 to 2021.



828

829

830 **Figure 109** Heatmap of multi-year mean gridded VIS_AOD and Aqua AOD and Terra AOD during
 831 2003-2021. Terra and Aqua AOD are averaged onto a grid of 0. 5°.

832 **3.5 Interannual variability and trend of visibility-derived AOD over global land**
 833 **Global spatiotemporal variation of AOD in 1980-2021**

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

839 ~~We first analyzed t~~The spatial distribution of multi-year average AOD ~~from 1980 to 2021~~ over land
 840 ~~is shown in Figure 11 (a).~~from 1980 to 2021 and separately for the Southern Hemispheres (SH, -60-
 841 0°N) and Northern Hemisphere (NH, 0-85°N) in Figure 10 (a). The mean AOD of land (-60-85°N),
 842 ~~NH and Northern Hemisphere (NH, 0-85°N), and the Southern Hemispheres (SH, -60-0°N)~~ SH is
 843 0.161 ± 0.074, 0.158 ± 0.076, and 0.173 ± 0.059, respectively. The AOD values of Africa, Asia,
 844 Europe, North America, Oceania, and South America are 0.241, 0.222, 0.110, 0.111, 0.129 and 0.117,
 845 respectively.

846 Due to the influence of geography, atmospheric circulation, population, and emissions, the AOD

847 varies in different latitudes. Figure 12 illustrates the multi-year average AOD in different latitude
848 ranges for land, the NH, and the SH from 1980 to 2021. Within [-20, 20°N], the global average AOD
849 reaches its maximum (0.234), and the maximum AOD NH is 0.256 in [0, 20°N]. The highest AOD
850 in SH is 0.217 in in [-15, 0°N]. The average AOD in SH rapidly decreases from -15°N to -35°N. In
851 NH, AOD is generally greater than in SH from 5°N to 65°N. When, the latitude is greater than 70°N,
852 the NH's AOD is smaller than the SH's.

853 There are many regions of Hhigh AOD values occur in ~~the~~NH, ~~and align~~with the distribution of
854 population density. Approximately 7/8 of the global population resides in the NH, with 50%
855 concentrated at 20°N-40°N (Kummu et al., 2016), indicating a significant impact of human activities
856 on aerosols. The highest AOD values are observed near 17°N, including the Sahara Desert, Arabian
857 Peninsula, and southeastern India, suggesting that in addition to anthropogenic sources, deserts also
858 play a crucial role in aerosol emissions. Lower AOD regions of the SH are from 25°S to 60°S
859 are found in the 25°S region of the SH, encompassing Australia, southern Africa, and southern South
860 America, indicating lower aerosol burdens in these areas. Additionally, North America also exhibits
861 low aerosol loading. Chin et al. (2014) analyzed the AOD over land from 1980 to 2009 with the
862 Goddard Chemistry Aerosol Radiation and Transport model, which is similar to the visibility-
863 derived AOD. The spatial distribution is consistent with the satellite results (Remer et al., 2008; Hsu
864 et al., 2012; Hsu et al., 2017; Tian et al., 2023). The AOD and extinction coefficient retrieved from
865 visibility show a similar distribution at global scale, with a correlation coefficient of nearly 0.6
866 (Mahowald et al., 2007). Similar global (Husar et al., 2000; Wang et al., 2009) and regional
867 (Koelemeijer et al., 2006; Wu et al., 2014; Boers et al., 2015; Zhang et al., 2017; Zhang et al., 2020)
868 spatial distributions have been reported.

869 AOD loadings exhibit significant seasonal variations worldwide, particularly over land. In this study,
870 a year is divided into four parts: December-January-February (DJF), March-April-May (MAM),
871 June-July-August (JJA), and September-October-November (SON), corresponding to winter
872 (summer), spring (autumn), summer (winter), and autumn (spring) in NH (SH), respectively. Figure
873 11 Figure 10(b-e) also depicts the spatial distribution of seasonal average AOD over land from 1980
874 to 2021. The global AOD in DJF, MAM, JJA, and SON is 0.158 ± 0.062 , 0.162 ± 0.081 , 0.175 ± 0.093 ,
875 and 0.153 ± 0.070 , respectively. The standard bias of AOD in JJA and MAM are greater than those
876 in DJF and SON. AOD exhibits seasonal changes, with the highest in JJA, followed by MAM, DJF,
877 and SON. From 1980 to 2021, the seasonal AOD in NH is 0.152 ± 0.064 (DJF), 0.161 ± 0.088 (MAM),
878 0.176 ± 0.090 (JJA), and 0.144 ± 0.060 (SON), and in SH is 0.184 ± 0.041 (DJF), 0.166 ± 0.044 (MAM),
879 0.169 ± 0.072 (JJA), and 0.19 ± 0.060 (SON).

880 In NH, the AOD ranking from high to low in season is summer > spring > winter > autumn. In SH,
881 the AOD ranking from high to low in season is spring > summer > winter > autumn. The highest
882 AOD is observed during JJA in NH, while in SH, the peak occurs during SON. The occurrence of
883 high AOD values is highly associated with the growth of hygroscopic particle and the photochemical
884 reaction of aerosol precursors under higher relative humidity~~the intensification of industrial~~
885 ~~activities~~ in Asia (JJA) (Remer et al., 2008) and Europe such as Russia (JJA), and biomass burning
886 in South America (SON), Southern Africa (SON), and biomass burning in Indonesia (SON)
887 (Ivanova et al., 2010; Krylov et al., 2014), ~~and the increased dust emissions in Middle East region~~
888 ~~related to the transport of dust from the Sahara region~~ (Remer et al., 2008; Prakash et al., 2014). On
889 the other hand, the lowest global AOD values are observed during autumn, which may be attributed

890 to the ~~weakening of influence of~~ monsoon systems (Li et al., 2016; Zhao et al., 2019).

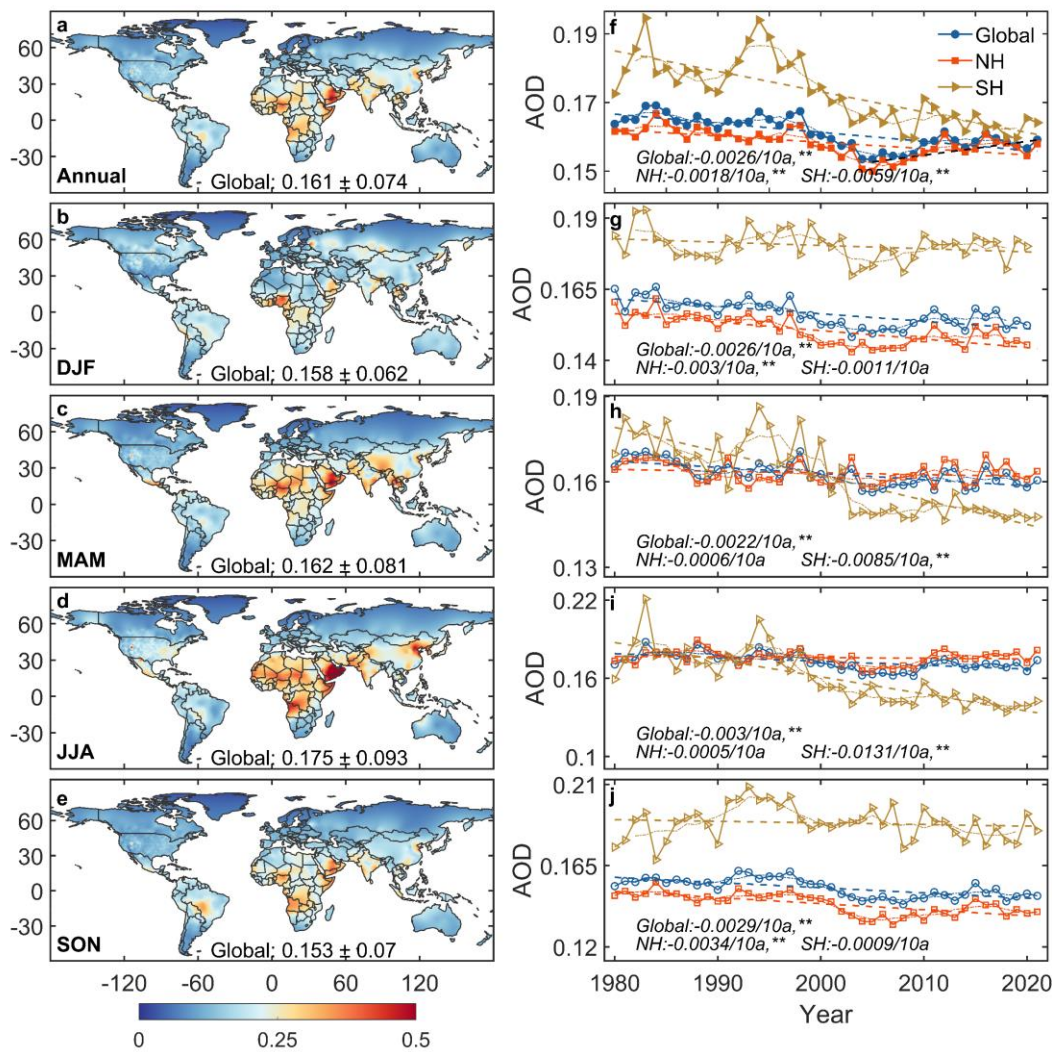
891 In addition to the spatial characteristics of AOD, the temporal variations in AOD have also been of
892 great interest due to the significant relationship between aerosols and climate change. ~~Figure~~
893 ~~40Figure 11~~ (f) shows the temporal trends of annual average AOD (** represents passing the
894 significance test, $p < 0.01$) over the global land, the SH and the NH during 1980-2021. The global
895 land, NH, and SH trends demonstrate decreasing trends of AOD with values of $-0.0026/10a$, $-$
896 $0.0018/10a$, and $-0.0059/10a$, respectively, with all passing the significance test with a confidence
897 level of 95%. Notably, the declining trend is much greater in the SH than in the NH. It may be
898 related to the decrease in the frequency of sandstorms and wildfires and the increase in precipitation,
899 such as in Australia. The MODIS satellite results (including oceans) indicate trends of $0.004/10a$,
900 $0.009/10a$, and $-0.002/10a$ for the global, SH, and NH, respectively, during the period of 2003-2020.
901 This findings suggest a growth trend in the global and NH and a declining trend in SH (Tian et al.,
902 2023). The trend of SeaWiFS AOD was $0.0058/10a$ over land during 1998-2010 (Hsu et al., 2012).
903 ~~Our study has the same downward signal as that in previous studies.~~ Two AOD peaks in 1983 and
904 1994 and two AOD valleys in 1980 and 1990 are observed before 2000. The two AOD peaks may
905 be attributed to large volcanic eruptions, which has been confirmed by previous studies. The
906 volcanic eruptions and their associated fires of the El Chichón volcano in Mexico in 1982 (Hirono
907 and Shibata, 1983) and Mount Pinatubo in the Philippines in 1991 (Tupper et al., 2005) resulted in
908 elevating global AOD levels in the following years. The AOD recovery to the previous low levels
909 after volcanic eruptions takes approximately 10 years (Chazette et al., 1995; Sun et al., 2019). This
910 further indicates the efficiency of our data capturing the volcanic eruption emission features. ~~also~~
911 ~~indicates that our data effectively captures this feature.~~

912 Due to the influence of geography, atmospheric circulation, population, and emissions, the trend of
913 global aerosols varies in different latitude Figure 12 illustrates the multi-year average AOD in
914 different latitude ranges for land, the NH, and the SH from 1980 to 2021. Within $[-20, 20^{\circ}N]$, the
915 global average AOD reaches its maximum (0.234), and the maximum AOD NH is 0.256 in $[0, 20^{\circ}N]$.
916 The highest AOD in SH is 0.217 in in $[-15, 0^{\circ}N]$. The average AOD in SH rapidly decreases from $-$
917 $15^{\circ}N$ to $-35^{\circ}N$. In NH, AOD is generally greater than in SH from $5^{\circ}N$ to $65^{\circ}N$. When, the latitude
918 is greater than $70^{\circ}N$, the NH's AOD is smaller than the SH's, which may be related to low emission
919 intensity and low population density in high latitude areas.

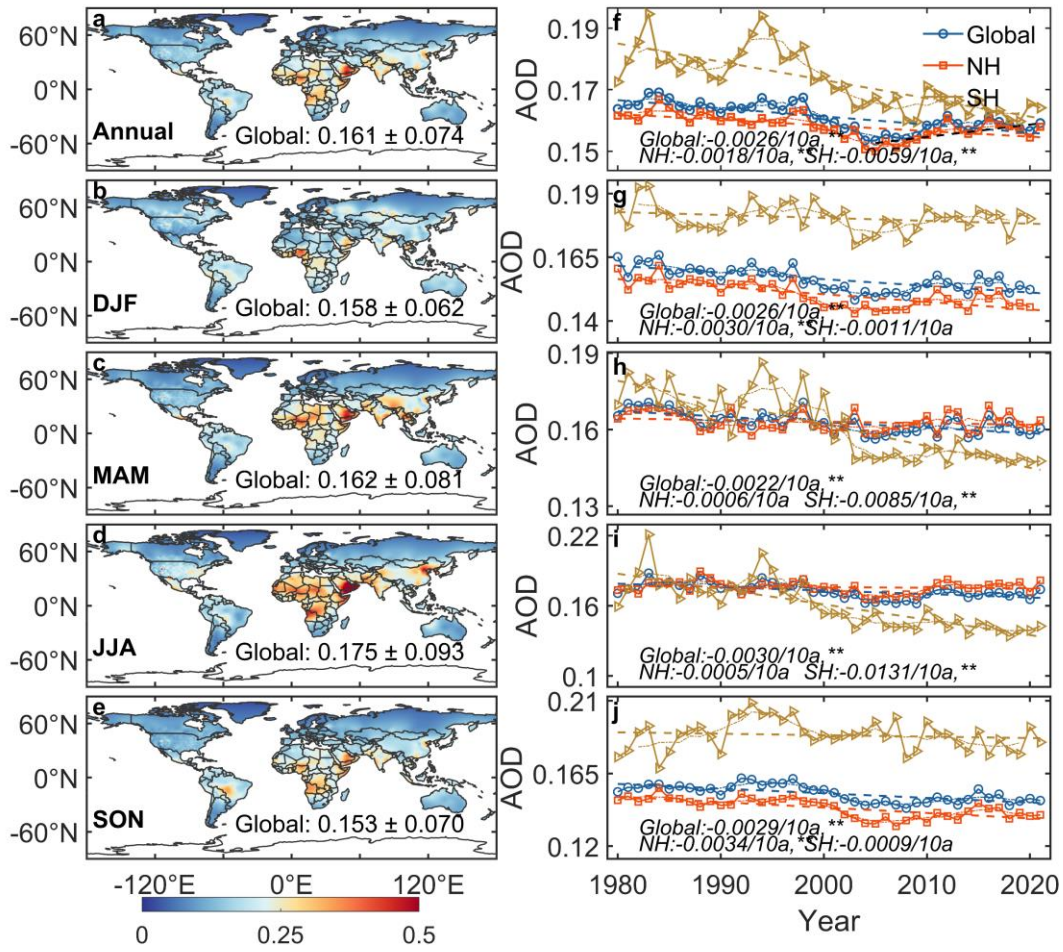
920 The ~~distinct~~ seasonal trends of AOD during 1980-2021 at the global and hemispheric scales are
921 shown in ~~Figure 10Figure 11~~ (g-j). The global AOD shows a decreasing trend in all seasons ($-$
922 $0.002 \sim -0.003/10a$). The large declining trends are observed in JJA and SON, with decreasing trend
923 values of $-0.003/10a$ and $-0.0029/10a$, respectively. DJF and MAM follow with decreasing trend
924 values of $-0.0026/10a$ and $-0.0022/10a$, respectively, all passing the significance test ($p < 0.01$). For
925 the NH, the AOD trends in different seasons are $-0.0030/10a$ (DJF), $-0.0006/10a$ (MAM), $-$
926 $0.0005/10a$ (JJA), and $-0.0034/10a$ (SON). DJF and SON pass the significance test ($p < 0.01$), while
927 MAM and JJA do not. In the SH, the trends are as follows: $-0.0011/10a$ (DJF), $-0.0085/10a$ (MAM),
928 $-0.0131/10a$ (JJA), and $-0.0009/10a$ (SON). Interestingly, in contrast to the NH, MAM and JJA pass
929 the significance test, while DJF and SON do not. The largest declining season in the NH is winter,
930 while in the SH, it is summer. The decreasing trend in the SH is more than four times greater than
931 that in the NH, particularly before the year 2000. While both the global and SH AOD exhibit a
932 decreasing trend since 2005, the NH has shown a significant increase in winter AOD, leading to an

933 overall increasing trend. Moreover, the NH shows an increasing trend of 0.004/10a from 2005 to
 934 2021.

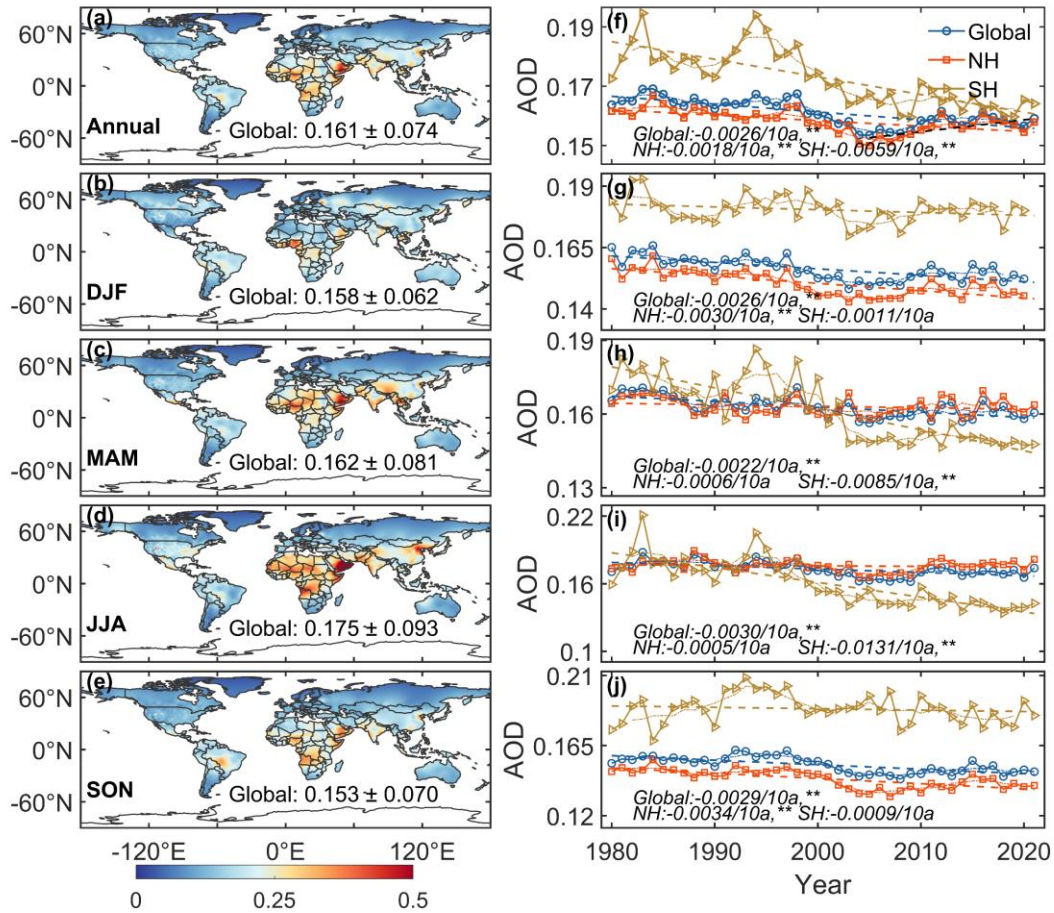
935 Annual SO₂ emissions increased from 9.4 to 15.3 TgS from 2000 to 2005, which ultimately ended
 936 up as sulfate aerosols, leading to a significant increase in sulfate aerosols (Hofmann et al., 2009).
 937 More relevantly, the frequent volcanic eruptions in tropical regions from 2002 to 2006, combined
 938 with seasonal circulation patterns during winter, led to the transport of aerosol particles to higher
 939 latitudes (Hofmann et al., 2009; Vernier et al., 2011; Sawamura et al., 2012; Andersson et al., 2015).



940

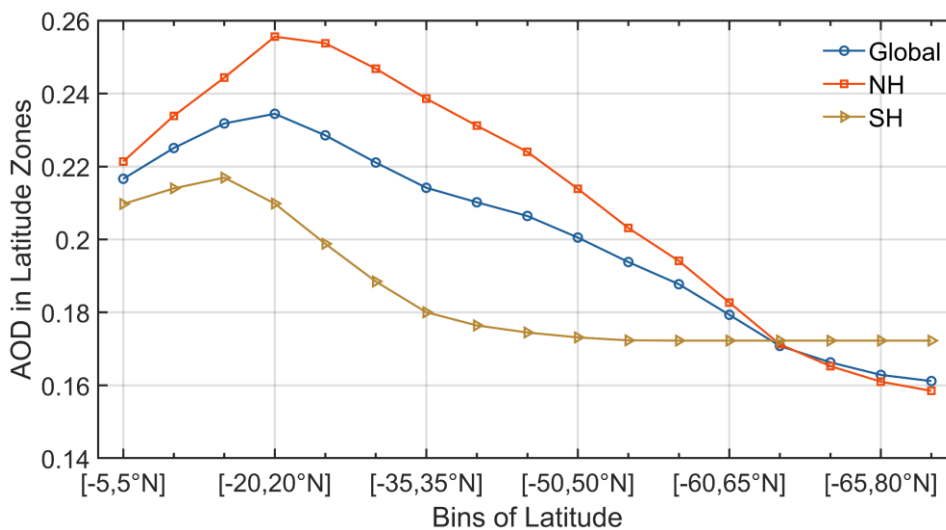


941



942

943 **Figure 10-11** The multi-year averages of VIS_AOD from 1980 to 2021. Global land (circle),
 944 northern hemisphere (NH,0-85°N) (triangle) and southern hemisphere (SH,0-60°S) (square) annual
 945 and seasonal AOD. The symbol, **, represents that the test passed at a significance level of 0.01.
 946 DJF represents December and next January and February. MAM represents March, April, and May.
 947 JJA represents June, July, and August. SON represents September, October, and November.—



948

949 **Figure 12** The global land (blue), northern hemisphere's (red) and southern hemisphere's (yellow)

950 multi-year average VIS_AOD from 1980 to 2021 in different latitude zones. The latitude range is
951 from -60 to 85°N, with a bin of 5°.

952 **3.6 Interannual variability and trend of visibility-derived AOD over regions** 953 **Regional spatiotemporal variation in AOD during 1980-2021**

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

959 To analyze the spatiotemporal characteristics and trends of AOD in different regions, we selected
960 12 representative regions that are influenced by various aerosol sources(Wang et al., 2009; Hsu et
961 al., 2012; Chin et al., 2014), such as desert, industry, anthropogenic emissions, and biomass burning
962 emissions, which nearly cover the most land and are densely populated regions_(Kummu et al.,
963 2016). These representative regions are Eastern Europe, Western Europe, Western North America,
964 Eastern North America, Central South America, Western Africa, Southern Africa, Australia,
965 Southeast Asia, Northeast Asia, Eastern China, and ~~the Middle East~~India, as shown in- Figure
966 1Figure 1. _-

967 We use multi-year average and seasonal average AOD to evaluate aerosol loadings (Figure 11Figure
968 13), the annual average of monthly anomalies to analyze interannual trends (Figure 14Figure 12),
969 and the seasonal average to analyze seasonal trends (Figure 15Figure 13) in 12 regions from 1980
970 to 2021. _

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

977 -
978 Figure 13Figure 11 shows the regions with high aerosol loadingsAOD level from 1980 to 2021
979 (multi-year average AOD > 0.2) are in West Africa, Northeast Asia, Eastern China, and ~~the Middle~~
980 ~~East~~India. The AOD values in Eastern North America, Central South America, South Africa, and
981 Southeast Asia range from 0.15 to 0.2-~~with middle aerosol loadings~~. The AOD values in Eastern
982 Europe, Western Europe, Western North America, and Australia are less than 0.15-~~with low aerosol~~
983 ~~loadings~~.

984 Europe is an industrial region with a low aerosol loading region, and the multi-year average AOD
985 in Eastern Europe (0.144±0.007) is higher than that in Western Europe (0.139±0.003) during 1980-
986 2021. Eastern Europe shows a greater downward trend in AOD (-0.0041/10a) compared to Western
987 Europe (-0.0021/10a). The highest AOD is observed in JJA, the dry period when solar irradiation
988 and boundary layer height increase, with Eastern Europe at 0.161 and Western Europe at 0.162,
989 which could be due to increases in secondary aerosols, biomass burning, and dust transport from

990 the Sahara (Mehta et al., 2016). However, there are seasonal variations. In Eastern Europe, the
991 seasonal AOD ranking from high to low is JJA (0.161) > DJF (0.147) > MAM (0.138) > SON
992 (0.131), while in Western Europe, it is JJA (0.162) > MAM (0.140) > SON (0.136) > DJF (0.117).
993 The differences among seasons are larger in Western Europe. AOD in Eastern Europe shows
994 declining trends in all seasons, while it does not pass the significance test in MAM. Among four
995 seasons, SON has the largest decline trend of AOD (-0.0058/10a). In Western Europe, DJF, JJA, and
996 SON exhibit declining trends of AOD that pass the significance test, while the MAM shows a
997 significant increase trend of AOD (0.0022/10a), which may be due to eruptions of the
998 Eyjafjallajökull volcano in Iceland in spring 2010 (Karbowska and Zembrzuski, 2016). Both
999 Western and Eastern Europe experienced increasing trends in AOD during the period of 1995-2005,
1000 with Western Europe showing a greater increase. However, after 2000, the decline rate accelerated
1001 in both regions. The downward trend in Europe is attributed to the reduction of biomass burning,
1002 anthropogenic aerosols, and aerosol precursors (such as sulfur dioxide)(Wang et al., 2009; Chin et
1003 al., 2014; Mortier et al., 2020).

1004 North America is also an industrial region with a low aerosol loading. The average AOD values for
1005 Eastern and Western North America during 1980-2021 are 0.153 ± 0.004 and 0.131 ± 0.005 ,
1006 respectively, with the Eastern region being higher than the Western region by 0.022. From 1980 to
1007 2021, both Eastern (-0.0021/10a) and Western North America (-0.0009/10a) show a downward trend;
1008 however, the decline in the Western region is not statistically significant. And the trend is -
1009 0.0172/10a from 1995 to 2005 and 0.0096/10a from 2005 to 2021. The average AOD values in DJF,
1010 MAM, JJA, and SON in Western North America are 0.1367, 0.1286, 0.1457, and 0.114, respectively,
1011 compared to 0.137, 0.145, 0.1913, and 0.138 in Eastern North America. The lowest AOD values of
1012 12 regions during DJF and SON are observed in Western North America (Remer et al., 2008).
1013 Specifically, in the Western region, there is a consistent increasing trend during MAM (0.004/10a)
1014 from 1980 to 2021, while JJA and SON also show an increase after 2000, except for DJF (-
1015 0.0032/10a). In contrast, the AOD trends in the Eastern region remain unchanged during the period
1016 1980-2021, except for MAM, which shows a stable increasing trend (0.0033/10a), while DJF, JJA,
1017 and SON exhibit decreasing trends (-0.0023/10a, -0.0040/10a, -0.0053/10a, respectively). In the
1018 Western region, the annual mean AOD started to increase after 2005, while in the Eastern region,
1019 the increase was not significant. The upward trend may be due to low rainfall and increased wildfire
1020 activities (Yoon et al., 2014). The decrease in AOD in Eastern North America is related to the
1021 reduction of sulfate and organic aerosols, as well as the decrease in anthropogenic emissions caused
1022 by environmental regulations (Mehta et al., 2016).

1023 Central South America is a relatively high aerosol loading region, sourced from biomass burning,
1024 especially in SON (Remer et al., 2008; Mehta et al., 2016), with a multi-year average AOD of
1025 0.192 ± 0.017 . There is a clear downward trend (-0.0100/10a) from 1980 to 2021, which is slightly
1026 greater than the trend (-0.0090/10a) from 1998 to 2010 (Hsu et al., 2012) and AOD decreased from
1027 1980 to 2006 (Streets et al., 2009) and from 2001 to 2014 (Mehta et al., 2016). Although DJF (0.199)
1028 and SON (0.226) have higher values compared to MAM (0.180) and JJA (0.163), the large declining
1029 trends are observed in MAM (-0.0126/10a) and JJA (-0.0167/10a). It indicates that although AOD
1030 has decreased overall, the aerosol loading caused by seasonal deforestation and biomass combustion
1031 is still large (Mehta et al., 2016).

1032 Africa is also one of the regions with a high aerosol loading worldwide. In West Africa, the average
1033 AOD is 0.275 ± 0.01246 during 1980-2021, and the annual AOD shows a downward trend ($-$
1034 $0.0008/10a$, $p > 0.05$). The world's largest desert (Sahara Desert) is in West Africa, with much dust
1035 aerosol discharged. AOD values in all seasons are above 0.25, with JJA (0.301) and MAM (0.300)
1036 reaching 0.3, and DJF and SON being 0.252 and 0.250, respectively. ~~In addition to the dust source,~~
1037 ~~frequent forest fires and biomass burning result in high AOD in JJA (Tian et al., 2023).~~ The AOD
1038 in DJF ($-0.0135/10a$, $p < 0.01$) and SON ($-0.0026/10a$, $p > 0.05$) exhibit decreasing trends, while JJA
1039 ($0.0088/10a$, $p < 0.01$) and MAM ($0.0037/10a$, $p > 0.05$) show an opposite trend. The multi-year
1040 average AOD in South Africa is 0.177 ± 0.020 , lower than that of West Africa. The annual mean AOD
1041 in South Africa shows a significant decrease ($-0.0096/10a$). The AOD values range from 0.12 to 0.2
1042 during 2000-2009, dominated by fine particle matter from industrial pollution from biomass and
1043 fossil fuel combustion (Hersey et al., 2015). The average AOD values in DJF, MAM, JJA, and SON
1044 are 0.189, 0.162, 0.147, and 0.210, respectively. JJA ($-0.0268/10a$, $p < 0.01$), MAM ($-0.0126/10a$,
1045 $p < 0.01$) and SON ($-0.0001/10a$, $p > 0.05$) exhibit a downward AOD trend, while DJF ($0.0006/10a$,
1046 $p > 0.05$) shows an upward trend. AERONET and simulation results also show a decreasing trend of
1047 AOD (Chin et al., 2014).

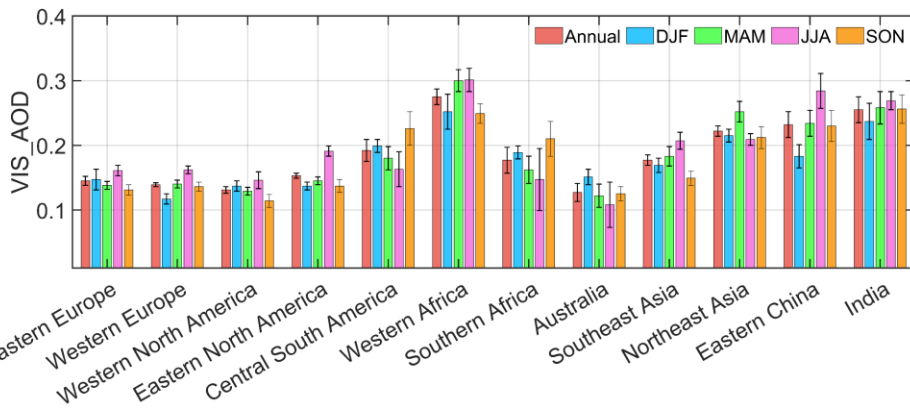
1048 Australia is a region with a low aerosol loading. The multi-year mean AOD is 0.127 ± 0.014 during
1049 1980-2021. The AOD ranges from 0.05 to 0.15 from AERONET during 2000-2021, and ~~dust and~~
1050 ~~biomass burning wildfires~~ are an important contributors to the aerosol loading (Yang et al., 2021a).
1051 There is a downward trend of AOD ($-0.0081/10a$, $p < 0.01$), which may be related to a decrease in
1052 ~~BC dust~~ and ~~biomass burning OC~~ (Yoon et al., 2016; Yang et al., 2021a). In addition, research has
1053 shown that the forest area in Australia has increased sharply since 2000 (Giglio et al., 2013),
1054 surpassing the forest fire area of the past 14 years. The seasonal average of AOD in MAM, JJA,
1055 SON, and DJF are 0.122, 0.108, 0.125, and 0.151. The AOD in JJA is the lowest among all seasons
1056 and regions. The highest AOD is in DJF with an increasing trend ($0.0056/10a$, $p < 0.01$), while the
1057 trends during MAM, JJA and SON are $-0.0096/10a$ ($p < 0.01$), $-0.0231/10a$ ($p < 0.01$) and $-0.0042/10a$
1058 ($p < 0.01$), respectively. Ground-based and satellite observations indicate that wildfires, biomass
1059 burning and sandstorms lead to high AOD in DJF and SON. The low AOD of MAM and JJA is due
1060 to a decrease in the frequency of sandstorms and wildfires and an increase in precipitation (Gras et
1061 al., 1999; Yang et al., 2021a; Yang et al., 2021b).

1062 Asia is also a high aerosol loading area with various sources. In Southeast Asia, the multi-year
1063 average AOD is 0.177 during 1980-2021 with a downward trend of AOD ($-0.0003/10a$, $p > 0.05$). It
1064 is also a biomass-burning area. The seasonal average AOD ranking from high to low is JJA (0.207) >
1065 MAM (0.183) > DJF (0.169) > SON (0.149). The trends in DJF ($-0.0035/10a$, $p < 0.05$), JJA ($-$
1066 $0.0007/10a$, $p > 0.05$) and SON ($-0.0021/10a$, $p > 0.05$) are opposite to MAM ($0.0050/10a$, $p < 0.01$).
1067 ~~Natural emissions were predominant in 1992 and 1997, because of the volcanic eruptions and forest~~
1068 ~~fires.~~ Southeast Asia has no clear long-term trend in estimated AOD or observed surface solar
1069 radiation (Streets et al., 2009). In Northeast Asia, the multi-year average AOD is 0.222 during 1980-
1070 2021, with no significant temporal trend. The seasonal AOD values are 0.252 in MAM, 0.215 in
1071 DJF, 0.212 in SON and 0.209 in JJA. AOD in MAM is significantly higher than other seasons, which
1072 may be related to sandstorms in East Asia, and the reason for the high AOD in winter may be related
1073 to the ~~low boundary layer height transportation~~. The trends of AOD in DJF ($-0.0025/10a$, $p > 0.05$),
1074 MAM ($0.0031/10a$, $p > 0.05$), JJA (0) and SON ($-0.0006/10a$, $p > 0.05$) are not significant. In Eastern

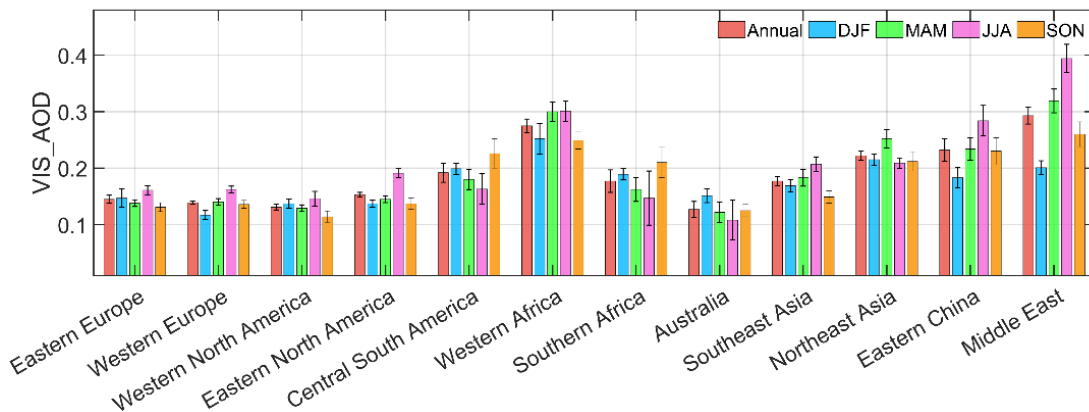
1075 China, the multi-year average AOD is 0.233, with an increasing trend (0.0071/10a, $p < 0.01$). The
1076 trend is 0.0151/10a from 1980 to 2006 and -0.0469/10a from 2006 to 2021. The seasonal average
1077 AOD ranking from high to low is JJA (0.284), MAM (0.234), SON (0.230) and DJF (0.183). The
1078 AOD trends in DJF (0.0093/10a, $p < 0.01$), MAM (0.0092/10a, $p < 0.01$), JJA (0.0038/10a, $p > 0.05$)
1079 and SON (0.0065/10a, $p < 0.05$) are all positive but the trend in JJA does not pass the significance
1080 test. We can see that there are three stages of changes in AOD: 1980-2005, 2006-2013 and 2014-
1081 2021. In the first stage, AOD increased steadily. In the second stage, AOD maintained a larger
1082 positive anomaly accompanied by oscillations-high level of volatility. The third stage experienced a
1083 rapid decline, reaching the level of the 1980s by 2021. The increasing trend of AOD before 2006
1084 may be due to the significant increase in industrial activity, and after 2013, the significant decrease
1085 is closely related to the implementation of air quality-related laws and regulations, along with
1086 adjustments in the energy structure (Hu et al., 2018; Cherian and Quaas, 2020).

1087 India is a high aerosol loading area. The multi-year average AOD is 0.255, with an upward trend
1088 (0.0096/10a, $p < 0.01$) from 1980 to 2021. Dust and biomass burning has an influence on AOD level.
1089 There are three stages: 1980-1997 (0.0032/10a, $p < 0.01$), 1997-2005 (-0.0420/10a, $p < 0.01$), 2005-
1090 2021 (0.0481/10a, $p < 0.01$). Although the trend is downward in the second stage, the larger positive
1091 trend is in the third stage. The seasonal average AOD values are 0.237 in DJF, 0.258 in MAM, 0.269
1092 in JJA, and 0.256 in SON. The largest AOD is in JJA. In winter and autumn, it affected by biomass
1093 burning, and in spring and summer, it is also affected by dust, transported from the Sahara under
1094 during the monsoon period (Remer et al., 2008). The trends in DJF (0.0152/10a, $p < 0.01$), MAM
1095 (0.0091/10a, $p < 0.01$), JJA (0.0025/10a, $p > 0.05$), and SON (0.0107/10a, $p < 0.05$) are positive. There
1096 largest trend is in winter. In the Middle East, aerosols are influenced by local deserts and aerosols
1097 transport from Africa and petroleum-related industries, resulting in high aerosol loading (Wei et al.,
1098 2019a; Wei et al., 2019b). The multi-year average AOD is 0.293, which is the highest among all 12
1099 study regions, with an upward trend (0.0027/10a, $p > 0.05$). The aerosol loading was higher during
1100 1980-1990 and 2000-2021 and lower during 1990-2000. The seasonal average AOD values are
1101 0.201 in DJF, 0.319 in MAM, 0.394 in JJA, and 0.26 in SON. The trends of AOD in DJF (-
1102 0.0039/10a, $p < 0.05$) and SON (-0.0012/10a, $p > 0.05$) show an upward trend, while the trends in
1103 MAM (0.0067/10a, $p < 0.05$) and JJA (0.0095/10a, $p < 0.01$) are opposite. This increasing trend is
1104 related to sand and dust emissions (Klingmüller et al., 2016).

1105
1106 To summarize, there are significant differences in the spatial distribution, annual trends, and
1107 seasonal trends of AOD across different regions from 1980 to 2021. The high aerosol loadings
1108 from 1980 to 2021 are in West Africa, Middle EastIndia and Asia, and low aerosol loading regions
1109 are in Europe, Western North America, and Australia. Eastern China and -Middle EastIndia show
1110 an increasing trend, Southeast Asia and Northeast Asia show no significant trend, and the other
1111 regions show downward trends. However, not all regional seasonal trends are consistent with their
1112 annual trends. The results in this study have supplemented the long-term trend and distribution of
1113 AOD over land.



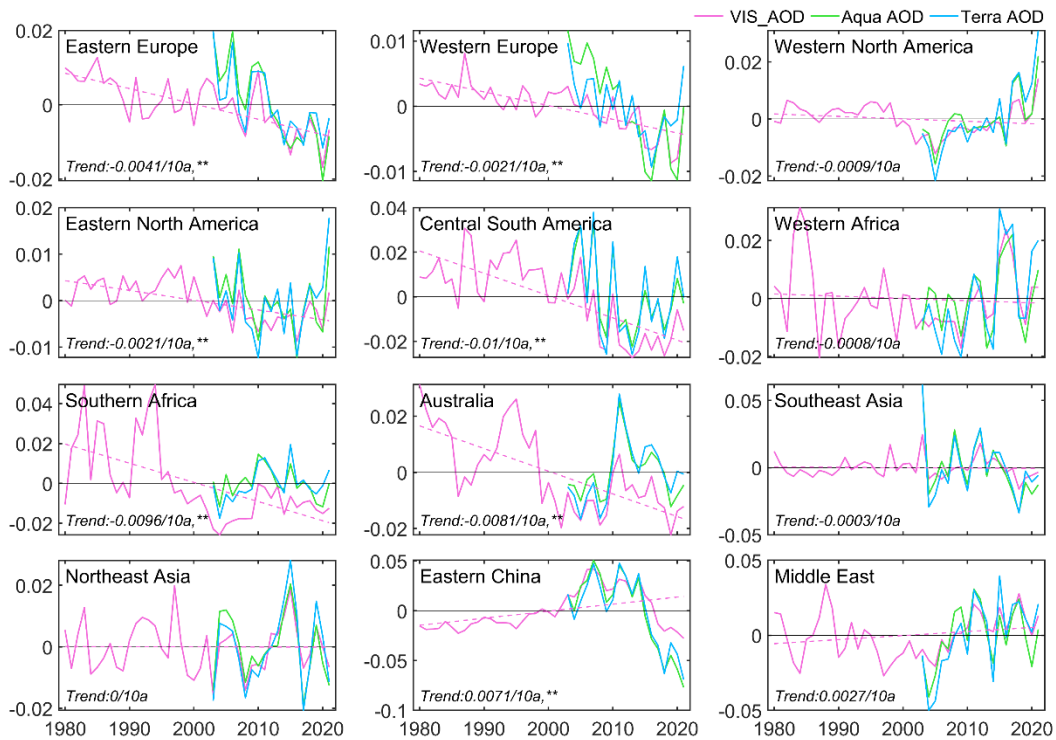
1114



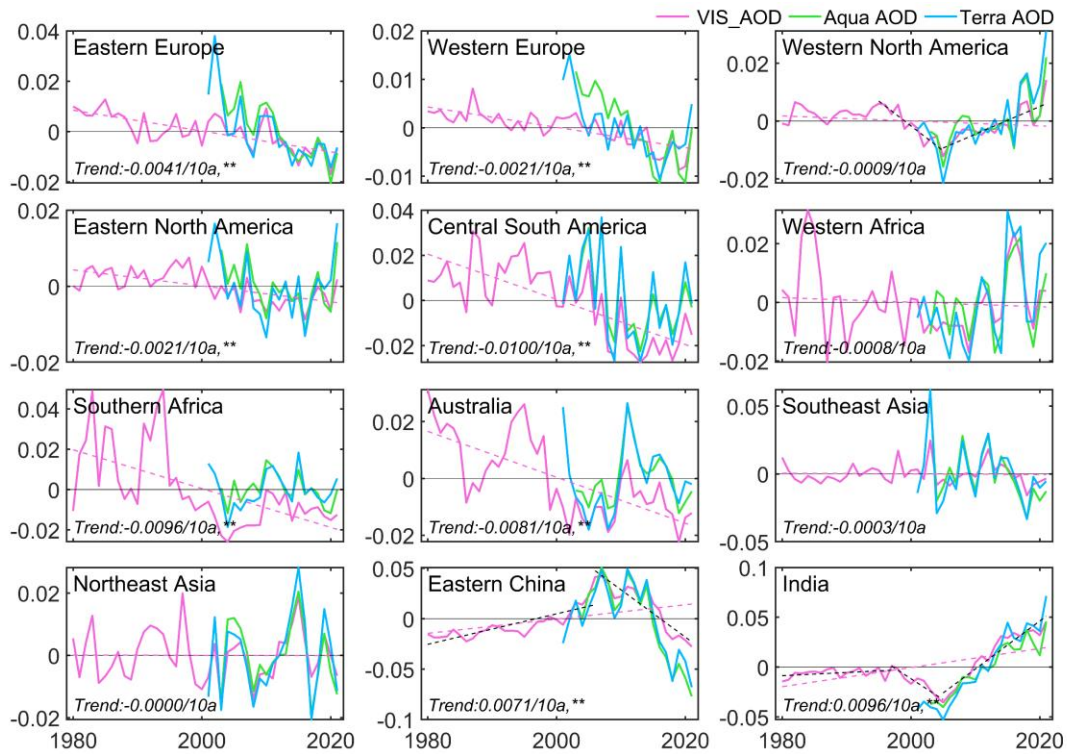
1115

1116

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

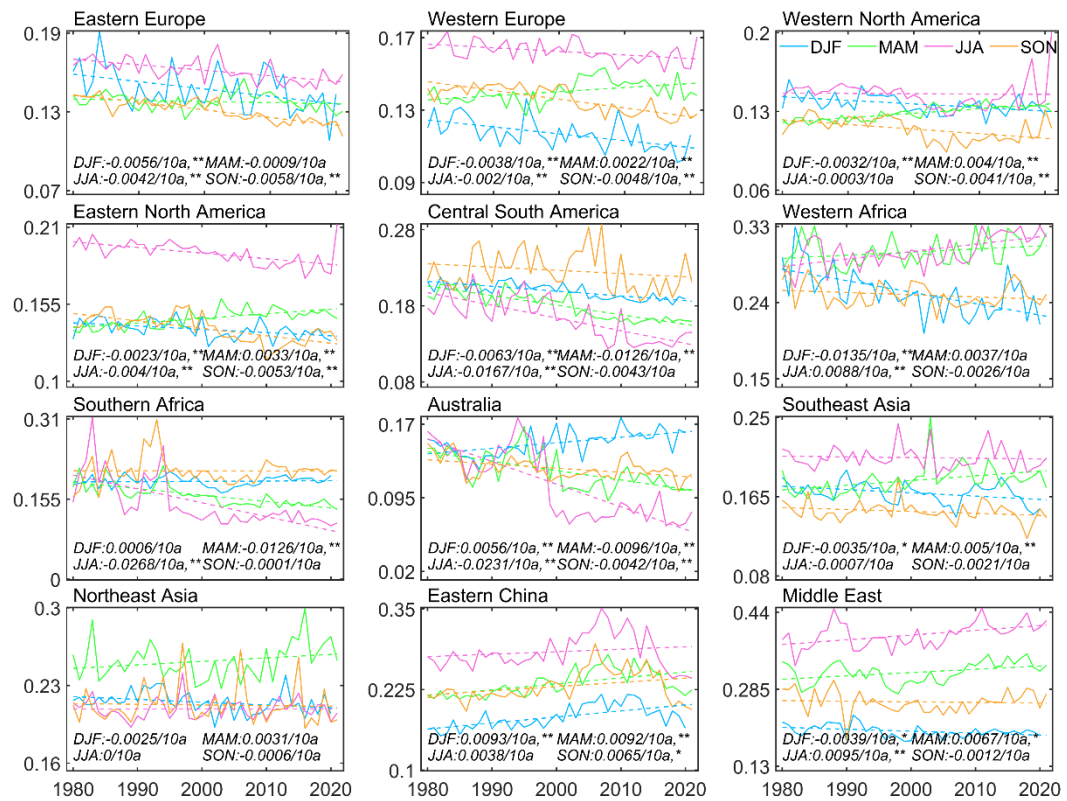


1117

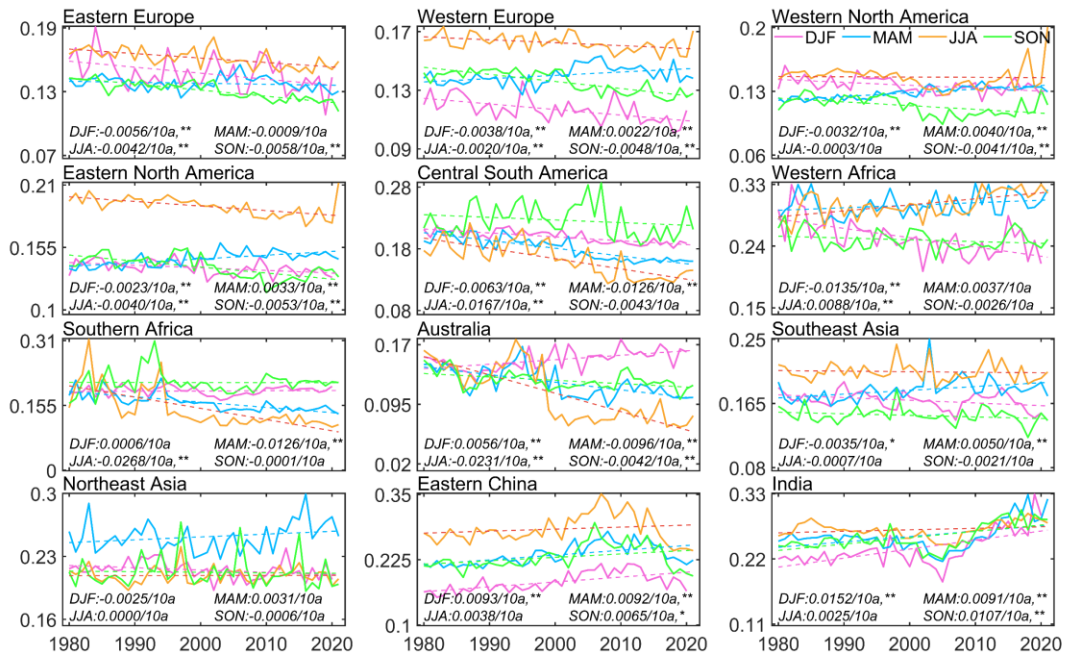


1118

1119 **Figure 12** Annual averages of monthly anomaly gridded VIS_AOD (pink line), Aqua
 1120 (green line), and Terra (blue line) MODIS AOD in 12 regions. The dotted line is the trend line.
 1121 **VIS_AOD has good temporal consistency with Aqua and Terra MODIS AOD from 2003 to 2021.**



1122



1123

1124 **Figure 15** Seasonal averages of gridded VIS_AOD during 1980 to 2021 in 12 regions
 1125 (Eastern Europe, Western Europe, Western North America, Eastern North America, Central South
 1126 America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China,
 1127 and **Middle East** India). The dotted line is the trend line.

1128 4 Data availability

1129 The visibility-derived AOD at station and grid scales over global land from 1980 to 2021 are
 1130 available at National Tibetan Plateau / Third Pole Environment Data Center
 1131 (<https://doi.org/10.11888/Atmos.tpdc.300822>) (Hao et al., 2023).

1132 5 Conclusions

1133 In this study, we employed a machine learning technique to derive AOD for over 5000 land stations
 1134 worldwide, based on satellite data, visibility, and related parameters. Monthly AOD was interpolated
 1135 onto a 0.5° grid using ordinary kriging with area weighting. The method was trained with Aqua
 1136 MODIS AOD. The accuracy and performance of the derived AOD were assessed and validated
 1137 against Terra MODIS AOD as well as AERONET ground-based observations of AOD for the
 1138 corresponding stations. Evaluation of the gridded AOD was conducted using Aqua and Terra
 1139 MODIS AOD. We obtained daily AOD for global land stations from 1980 to 2021, as well as
 1140 monthly gridded AOD. The two datasets complement the shortcomings of AOD in terms of time
 1141 scale and spatial coverage. Finally, the spatiotemporal variation in AOD was analyzed for global
 1142 land, the Southern Hemisphere, the Northern Hemisphere, and 12 regions in the past 42 years.
 1143 Several key findings have been obtained in this study as follows.

1144 **1. Modeling and gridding evaluation performance.** The mean RMSE, MAE, and R of all stations
1145 are 0.078, 0.044, and 0.750, respectively. The RMSE of 93% stations is less than 0.11, the MAE of
1146 91% is less than 0.06, and the R of 88% is greater than 0.7, respectively.

1147 ~~2. The gridded AOD is highly consistent with the satellite observations. Compared to Aqua and~~
1148 ~~Terra, the average biases of multi-year-gridded AOD compared to Aqua and Terra are 3.3% and~~
1149 ~~1.9%, and respectively. The spatial correlation coefficients are 0.80 and 0.79, with the zonal~~
1150 ~~correlation coefficients are of 0.997 and 0.99, and the meridional correlation coefficients are of~~
1151 ~~0.9986 and 0.90.~~

1152 **2. Model validation.** For the daily scale, the R, RMSE and MAE of between VIS AOD and Aqua
1153 AOD is 0.799, 0.079 and 0.044, respectively. The percentage of sample point falling within the EE
1154 envelopes is 84.12%. The R between VIS AOD and Terra AOD is 0.542, with a RMSE of 0.125
1155 and MAE of 0.078. The percentage falling within the EE envelopes is 64.76%. The R between
1156 VIS AOD and AERONET AOD is 0.546, with a RMSE of 0.186 and MAE of 0.099. The percentage
1157 falling within the EE envelopes is 57.87%. For the monthly and annual scales, RMSE and MAE
1158 show a significant decrease between VIS AOD and Aqua, Terra, and AERONET AOD, and R and
1159 percentages falling within EE show a significant increase.

1160 **3. Error analysis.** The average bias is 0.015 (AOD < 0.1), with 83% of data within the EE envelopes.
1161 As pollution level increases, the negative mean bias becomes significant and the underestimation
1162 increases. There is a negative bias in the low elevation (<=0.5km) with a percentage of 60%-64%
1163 falling within the EE envelopes and a positive bias in high elevation (0.5-1.2km) with a percentage
1164 of 50%-65% falling within the EE envelopes. The elevation of AERONET's site caused a bias in
1165 high elevation. When the elevation difference is negative (the elevation of the meteorological station
1166 is lower than that of the AERONET site), there is a significant positive bias. When the difference is
1167 positive, the mean bias approaches 0 or is positive. The bias does not change significantly with
1168 increasing distance between the meteorological station and AERONET site.

1169 **4. Global land AOD.** The global, NH, and SH AOD values from 1980 to 2021 are 0.161 ± 0.074 ,
1170 0.158 ± 0.076 , and 0.173 ± 0.059 , respectively. Trends in AOD for the global, NH, and SH
1171 demonstrate a decreasing trend of $-0.0026/10a$, $-0.0018/10a$, and $-0.0059/10a$, respectively ($p < 0.01$).
1172 The seasonal AOD ranking from high to low is JJA > MAM > DJF > SON over the global land and in
1173 the NH, while in the SH, it is DJF > JJA > MAM > SON. The largest declining trends are observed in
1174 NH summer and SH winter.

1175 **4. Regional AOD.** From 1980 to 2021, regions with high aerosol loadings (AOD > 0.2) were found
1176 in West Africa, Northeast Asia, Eastern China, and ~~the Middle East~~ India. Regions with moderate
1177 aerosol loadings (AOD between 0.15 and 0.2) are Eastern North America, Central South America,
1178 South Africa, and Southeast Asia. Eastern Europe, Western Europe, Western North America, and
1179 Australia are regions with low aerosol loadings (AOD < 0.15). The trends are $-0.0041/10a$, $-$
1180 $0.0021/10a$, $-0.0009/10a$, $-0.0021/10a$, $-0.0100/10a$, $-0.0008/10a$, $-0.0096/10a$, $-0.0081/10a$, $-$
1181 $0.0003/10a$, $-0.0000/10a$, $0.0071/10a$, and $0.0096/10a$ in Eastern Europe, Western Europe, Western
1182 North America, Eastern North America, Central South America, Western Africa, Southern Africa,
1183 Australia, Southeast Asia, Northeast Asia, Eastern China, and India, respectively.

1184 Competing interests

1185 The contact author has declared that none of the authors has any competing interests.

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1188 (2022YFF0801302) and the National Natural Science Foundation of China (41930970). The hourly
1189 visibility data were download from <https://mesonet.agron.iastate.edu/ASOS>. The Aerosol Robotic
1190 Network (AERONET) ~~daily~~-15-minute aerosol optical depth (AOD) data were download from
1191 which can be downloaded from <https://aeronet.gsfc.nasa.gov>. The MODIS AOD data were
1192 download from <https://ladsweb.modaps.eosdis.nasa.gov/>.-

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