1 Visibility-derived aerosol optical depth over global land from 1959 to

2 **2021**

3 Hongfei Hao¹, Kaicun Wang², Chuanfeng Zhao³, Guocan Wu¹, Jing Li³

¹Global Change and Earth System Science, Faculty of Geographical Science, Beijing Normal
 University, Beijing 100875, China

- ²Institute of Carbon Neutrality, Sino French Institute of Earth System Science, College Urban and
 Environmental Sciences, Peking University, Beijing 100871, China
- 8 ³Institute of Carbon Neutrality, Department of Atmospheric and Oceanic Sciences, School of

9 Physics, College Urban and Environmental Sciences, Peking University, Beijing 100871, China

10 Corresponding Author: Kaicun Wang (kcwang@pku.edu.cn)

11 Abstract

Long-term and high spatial resolution aerosol optical depth (AOD) data are essential for climate 12 change detection and attribution. Global ground-based AOD observations are sparsely distributed, 13 14 and satellite AOD retrievals have a low temporal frequency, as well low accuracy before 2000 over land. In this study, AOD at 550 nm is derived from visibility observations collected at more than 15 16 5000 meteorological stations over global land from 1959 to 2021. The AOD retrievals (550 nm) of 17 the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua Earth observation satellite are used to train the machine learning model, and the ERA5 reanalysis boundary layer 18 height is used to convert the surface visibility to AOD. Comparison with independent dataset 19 20 (AERONET ground-based observations) shows that the predicted AOD has a correlation coefficient 21 of 0.55 at daily scale. The correlation coefficients are higher at monthly and annual scales, which 22 are 0.61 for the monthly and 0.65 for the annual, respectively. The evaluation shows consistent 23 predictive ability prior to 2000, with a correlation coefficient of 0.54, 0.66 and 0.66 at daily, monthly, 24 and annual scales, respectively. Due to a small number and sparse visibility stations prior to 1980, 25 the global/regional analysis in this study is from 1980 to 2021. From 1980 to 2021, the mean 26 visibility-derived AOD over the global land, the Northern Hemisphere, and the Southern 27 Hemisphere are 0.177, 0.178, and 0.175, with a trend of -0.0029/10a, -0.0030/10a, and -0.0021/10a 28 from 1980 to 2021. The regional means (trends) of AOD are 0.181 (-0.0096/10a), 0.163 (-29 0.0026/10a), 0.146 (-0.0017/10a), 0.165 (-0.0027/10a), 0.198 (-0.0075/10a), 0.281 (-0.0062/10a), 0.182 (-0.0016/10a), 0.133 (-0.0028/10a), 0.222 (0.0007/10a), 0.244 (-0.0009/10a), 0.241 (0.0130 30 /10a), and 0.254 (0.0119/10a) in Eastern Europe, Western Europe, Western North America, Eastern 31 32 North America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, 33 Northeast Asia, Eastern China, and India, respectively. However, the trends are decreasing 34 significantly in Eastern China (-0.0572/10a) and Northeast Asia (-0.0213/10a) after 2014 and the 35 lager increasing trend is found after 2005 in India (0.0446/10a). The visibility-derived daily AOD 36 dataset at 5032 stations over global land from 1959 to 2021 are available at National Tibetan Plateau / Third Pole Environment Data Center (https://doi.org/10.11888/Atmos.tpdc.300822) (Hao et al., 37

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42 **1 Introduction**

Atmospheric aerosols are composed of solid and liquid particles suspended in the atmosphere. 43 Aerosol particles are directly emitted into the atmosphere or formed through gas-particle 44 45 transformation (Calvo et al., 2013), with diverse shapes and sizes (Fan et al., 2021), optical properties, and components (Liao et al., 2015; Zhang et al., 2020; Li et al., 2022). Most atmospheric 46 47 aerosols are concentrated in the troposphere, especially in the boundary layer (Liu et al., 2022), with a high concentration near emission sources (Kulmala et al., 2004), and a small portion are distributed 48 49 in the stratosphere. Atmospheric aerosols severely impact the atmospheric environment and human health. They deteriorate air quality, reduce visibility, and cause other environmental issues (Wang 50 51 et al., 2012; Boers et al., 2015). They impair human health or other organisms' conditions by 52 increasing cardiovascular and respiratory disease incidence and mortality rates (Chafe et al., 2014; 53 Yang et al., 2022). The Global Burden of Disease shows that global exposure to ambient PM_{2.5} 54 (particulate matter suspended in air with an aerodynamic diameter of less than 2.5 micrometers) resulted in 0.37 million deaths and 9.9 million disability-adjusted life years (Chafe et al., 2014). 55

56 Aerosols are inextricably linked to climate change. Atmospheric aerosols alter the Earth's energy 57 budget and affect the climate (Li et al., 2022). They cool the surface and heat the atmosphere by 58 scattering and absorbing solar radiation (Forster et al., 2007; Chen et al., 2022). Aerosols, such as black carbon and brown carbon, also absorb solar radiation (Bergstrom et al., 2007), heat the local 59 60 atmosphere and suppress or invigorate convective activities (Ramanathan et al., 2001; Sun and Zhao, 61 2020). Aerosols also alter the optical properties and life span of clouds (Albrecht, 1989). 62 Atmospheric aerosols strongly affect regional and global short-term and long-term climates through 63 direct and indirect effects (Mcneill, 2017).

Tropospheric aerosols are considered as the second largest forcing factor for global climate change 64 65 (Li et al., 2022), and they reduce the warming due to greenhouse gases by -0.5°C (IPCC, 2021). However, aerosols are also regarded as the largest contributor to the uncertainty of present-day 66 climate change attribution (IPCC, 2021). The uncertainties are caused by the deficiencies of the 67 68 global descriptions of aerosol optical properties (such as scattering and absorption) and 69 microphysical properties (such as size and component), and the impact on cloud and precipitation, 70 further affecting the estimation of aerosol radiative forcing (Lee et al., 2016; IPCC, 2021). Therefore, 71 sufficient aerosol observations are crucial. In aerosol measurements, aerosol optical depth (AOD) 72 is often used to describe its column properties, which represents the vertical integration of aerosol 73 extinction coefficients. AOD is an important physical quantity for estimating the content, 74 atmospheric pollution and climatology of aerosols (Zhang et al., 2020).

AOD data usually from ground-based and satellite-borne remote sensing observations. They have both advantages and disadvantages. Ground-based lidar observation is an active remote sensing technology. Lidar generally emits laser and receives backscattered signals to invert the extinction 78 coefficient of aerosols at different heights (Klett, 1985). By using the depolarization ratio, the type 79 of aerosol, such as fine particles or dust, can be distinguished (Bescond et al., 2013). The AOD within a certain height can be calculated by integrating the extinction coefficients; however, 80 scattering signals are usually not received near the ground, leading to blind spots (Singh et al., 2019). 81 82 At present, there are many ground-based lidar worldwide and regional networks, which provides 83 important support of vertical changes in aerosols, such as the NASA Micro-Pulse Lidar Network 84 (MPLNET) in the early 1990s (Welton et al., 2002), the European Aerosol Research Lidar Network 85 (EARLINET) since 2000 (Bösenberg and Matthias, 2003), the Latin American Lidar Network 86 (LALINET) since 2013 (Guerrero-Rascado et al., 2016).

87 Ground-based remote sensing observations supply aerosol loading data (such as AOD), by measuring the attenuation of radiation from the top of the atmosphere to the surface (Holben et al., 88 89 1998). This type of observation mainly uses weather-resistant automatic sun and sky scanning 90 spectral radiometers to retrieve optical and microphysical aerosol properties (Che et al., 2014). The 91 Aerosol Robotic Network (AERONET) is a popular global network composed of NASA and 92 multiple international partners that provides high-quality and high-frequency aerosol optical and 93 microphysical properties under various geographical and environmental conditions (Holben et al., 94 1998; Dubovik et al., 2000). The AERONET observations are extensively used to validate satellite remote sensing observations and model simulations, as well as climatology study (Dubovik et al., 95 96 2002b). There are many regional networks of sun photometers, such as the Maritime Aerosol Network (MAN), which use a handheld sun photometer to collect data over the ocean and is merged 97 98 into AERONET (Smirnov et al., 2009), the China Aerosol Robot Sun Photometer Network 99 (CARSNET) (Che et al., 2009), the Canadian sub-network of AERONET (AEROCAN) (Bokoye et 100 al., 2001), Aerosol characterization via Sun photometry: Australian Network (AeroSpan) (Mukkavilli et al., 2019), and the sky radiometer network (SKYNET) in Asia and Europe (Kim et 101 102 al., 2004; Nakajima et al., 2020). Another very valuable global network is the NOAA/ESRL 103 Federated Aerosol Network (FAN), which uses integrated nephelometers distinct from sun photometers, mainly located in remote areas, providing background aerosol properties over 30 sites 104 105 (Andrews et al., 2019).

106 Satellite remote-sensing is a space-based method that can provide aerosol properties worldwide. 107 With the development of satellite remote sensing technology since 1970s, aerosol distributions can be extracted with the advantage of sufficient real-time and global coverage from multiple satellite 108 109 sensors (Kaufman and Boucher, 2002; Anderson et al., 2005). The Advanced Very High Resolution 110 Radiometer (AVHRR) is the earliest sensor used for retrieving AOD over ocean (Nagaraja Rao et 111 al., 1989). The Moderate Resolution Imaging Spectroradiometer (MODIS), on board the Terra (launched in 1999) and Aqua (launched in 2002) satellites is a popular sensor with 36 channels, 112 which have been used for AOD retrieval over both ocean and land based on the Dark Target and the 113 Deep Blue algorithms (Remer et al., 2005; Levy et al., 2013). The latest MODIS AOD data version 114 is the Collection 6.1, which provides global AOD over 20 years (Wei et al., 2019). There are also 115 many other satellite sensors that can be used to retrieve AOD, such as the Polarization and 116 117 Directionality of the Earth's Reflectances (POLDER) during 1996-1997, 2003 and 2004-2013 118 (Deuzé et al., 2000), Sea-viewing Wide Field-of-view Sensor (SeaWIFS) during 1997-2007 119 (O'reilly et al., 1998), the Multi-angle Imaging Spectroradiometer (MISR) on Terra since 1999 (Diner et al., 1998). The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) has also 120

derived aerosols in the vertical direction since 2006 (Winker et al., 2009).

122 These measurements provide important data for studying the global and regional spatiotemporal 123 variabilities and climate effect of aerosols. However, ground-based remote sensing observations 124 only provide aerosol properties with low spatial coverage. There were only about 150 ground 125 stations worldwide in 2002 and even fewer sites were available for climate analysis (Holben et al., 126 1998; Chu et al., 2002), which limited aerosol climate research by spatial coverage (Bright and Gueymard, 2019). Satellite remote sensing overcomes the limitations of spatial coverage. The 127 128 AVHRR has been used to retrieve AOD since 1980, but it is limited by a few channel number, low 129 spatial resolution, and insufficient validation through ground-based observations before 2000 (Hsu 130 et al., 2017). Many studies have only investigated the trends and distributions of aerosols after 2000 (Bösenberg and Matthias, 2003; Winker et al., 2013; Xia et al., 2016; Tian et al., 2023), because of 131 the lack of long-term and global cover AOD products, which is the bottleneck for aerosol climate 132 133 change detection and attributions.

134 To overcome these limitations and enrich aerosol data, alternative observation data could be utilized

to derive AOD. Atmospheric horizontal visibility is a suitable alternative (Wang et al., 2009; Zhang
 et al., 2020), because it has the advantages of the long-term records with a large number of stations

137 worldwide.

138 Atmospheric visibility is a physical quantity that describes the transparency of the atmosphere through manual and automatic observations, and the automatic observations of visibility usually 139 measure atmospheric extinction (scattering coefficient and transmissivity). Koschmieder (1924) 140 141 first proposed the relationship between the meteorological optical range and the total optical depth. 142 Elterman (1970) futher established a formula between AOD and visibility by assuming an 143 exponential decrease in aerosol concentration with altitude, considering the extinction of molecules 144 and ozone to analyze air pollution, which called the Elterman model. Qiu and Lin (2001) corrected 145 the Elterman model by considering the influence of water vapor and used two water vapor pressure correction coefficients to retrieve AOD of 16 stations in China in 1990. Wang et al. (2009) analyzed 146 147 the trend of AOD using visibility-based retrivals from 1973 to 2007 over land. Lin et al. (2014) 148 retrieved the AOD in eastern China in 2006 using visibility and aerosol vertical profiles provided 149 by GEOS-Chem. Wu et al. (2014) and Zhang et al. (2017) parameterized the constants in the 150 Elterman model and use satellite retrieved AOD to solve the parameters in the models at different stations, to retrive the long-term AOD in China. 151

152 Zhang et al. (2020) reviewed the methods of visibility retrieval of AOD, indicating that visibility-153 based retrieval of AOD can compensate for the shortcomings of long-term aerosol observation data. 154 Simultaneously, various parameters, such as station altitude, consistency of visibility data, water 155 vapor and aerosol vertical profiles (scale height), were discussed with modified suggestions 156 proposed. These studies have enriched AOD data regionally. These studies have enriched aerosol 157 data in some extent. At present, there are very few studies on global visibility-retrieved AOD and to 158 analyze climatology of aerosols.

159 The two physical quantities of visibility and AOD have both connections and differences, making it 160 challenging to retrieve AOD from visibility. Visibility represents the maximum horizontal visible 161 distance near the surface which is impaired by surface aerosols, while AOD represents the total 162 column attenuation of solar radiation by aerosols from the surface to top of atmosphere. The

visibility of automatic observation is dependent on the local horizontal atmosphereic extinction 163 (NOAA et al., 1998). Visibility has not a simple linear relationship with meteorological factors. The 164 vertical structure of aerosols is the greatest challenge to obtain, as it is not a simple hypothetical 165 curve in complex terrain and circulation conditions (Zhang et al., 2020). These limitations make it 166 more complex to derive AOD. Machine learning methods can effectively address complex nonlinear 167 168 relationships between variables and have been widely applied in remote sensing and climate research fields. Li et al. (2021) used the random forest method to predict PM_{2.5} in Iraq and Kuwait 169 170 based on satellite AOD during 2001-2018. Kang et al. (2022) applied LightGBM and random forest to estimate AOD over East Asia, and the results showed a consistency with AERONET. Dong et al. 171 172 (2023) derived aerosol single scattering albedo from visibility and satellite AOD over 1000 global 173 stations. Hu et al. (2019) used a deep learning method to retrieve horizontal visibility from MODIS 174 AOD. These studies have confirmed the ability of machine learning to effectively solve complex 175 relationships among variables. 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 176 177 visibility over global land by using the machine learning method.

178 In this study, we propose a machine learning method to derive AOD, where satellite AOD is the 179 target value, and visibility and other related meteorological variables are the predictors. We explain the model's robustness, and evaluate the model's predictive ability, and validate the model's 180 181 predictions using independent ground-based AOD, satellite retrievals and reanalysis AOD, and analyze the mean and trend of AOD across land and regions. A station-scale dataset of long-term 182 AOD is generated. The Section 2 introduces the data and method. The Section 3 is the evaluation 183 and validation of the visibility-derived AOD, and the distribution and trends are discussed at global 184 185 and regional scales. The Section 5 presents the conclusions. This study is dedicated to supporting 186 the research of aerosols in climate change detection and attribution.

187 2 Data and method

188 2.1 Study area

189 The study area is global land. A total of 5032 meteorological stations and 395 AERONET sites are 190 selected in this study, shown in Figure 1. Twelve regions are selected for regional analysis, including Eastern Europe, Western Europe, Western North America, Eastern North America, Central South 191 192 America, Western Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, 193 and India and the number of stations in the regions is 187, 494, 390, 1759, 132, 72, 78, 86, 76, 140, 194 26, and 51, respectively. The meteorological observations data including visibility are available 195 since 1959. The time period for global and regional analysis is from 1980 to 2021, during which the visibility observations are sufficient with a uniform spatial distribution. As shown in Figure 1, the 196 number of active stations has exceeded 2000 during the period of 1980-1990 and the number of 197 active stations has exceeded 3000 since 2000. 198

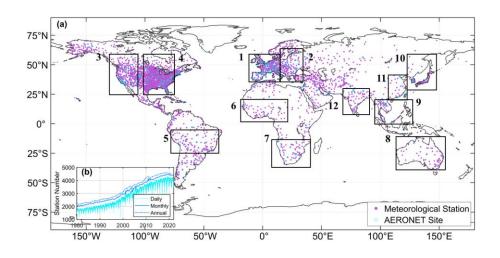


Figure 1: Study area (a) and the meteorological station number (b) at daily, monthly, and annual
scale. The number of meteorological stations (filled circles) is 5032. The number of AERONET
sites (empty circles) is 395. The box regions of labelled with number 1-12 are Eastern Europe,
Western Europe,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

205 2.2 Meteorological data

The ground-based hourly meteorological data from 1959 to 2021 is collected from 5032 206 207 meteorological stations of airports over land, which can be downloaded at 208 https://mesonet.agron.iastate.edu/ASOS. Over 1000 stations belong to the Automated Surface 209 Observing System (ASOS), and others are sourced from airport reports around the world. The 210 visibility measurements can be divided into automatic observation and manual observation. 211 Automatic visibility observations reduce errors associated with human involvement in data 212 collection, processing, and transmission. The visibility and other meteorological data are extracted 213 from the Meteorological Terminal Aviation Routine Weather Report (METAR). The World 214 Meteorological Organization (WMO) sets guidelines for METAR reports, including report format, 215 encoding, observation instruments and methods, data accuracy, and consistency, which ensures the 216 consistency and comparability of METAR reports globally. Some international regulations can be 217 https://community.wmo.int/en/implementation-areas-aeronautical-meteorologyreferenced at 218 programme.

The daily average visibility is calculated using harmonic mean in equation (1). The reciprocal of visibility is proportional to the extinction coefficient (Wang et al., 2009). Experiments have found that harmonic average visibility can better detect the weather phenomena than arithmetic average visibility, when visibility decline quickly (NOAA et al., 1998). Therefore, daily visibility will have greater representativeness.

224
$$V = n/(\frac{1}{v_1} + \frac{1}{v_2} + \dots + \frac{1}{v_n}),$$
 (1)

where V is the harmonic mean visibility, n is the daily record number, and V_1 , V_2 ,... V_n are the individual hourly visibility. In addition to hourly visibility (VIS), other variables closely related to aerosol properties are selected, including relative humidity (RH), dew point temperature (DT), temperature (TMP), wind speed (WS) and sea-level pressure (SLP). Because air temperature affects atmospheric stability and the rate of secondary particle formation, and humidity influences the size and hygroscopic growth, and wind speed and pressure significantly impact the transport and deposition. Sky conditions (cloud amount) and hourly precipitation are also selected to remove the records of extensive cloud cover and precipitation.

234 We have processed the meteorological data as follows. The records with high missing value ratio 235 are eliminated (Husar et al., 2000). When over 80% overcast or fog, the records of sky conditions 236 are eliminated, though such situations occur less than 1% of the time over land (Remer et al., 2008). 237 The records with 1-hour precipitation greater than 0.1 mm are eliminated. We calculate the 238 temperature dew point difference (dT). The low visibility records under "blowing snow" weather 239 are eliminated at high latitude region (> 65° N), when wind speed is great than 4.5m/s (Husar et al., 240 2000). When the RH is greater than 90%, it is impossible to distinguish whether it is fog or haze, or 241 both, and even precipitation. Therefore, the records with the RH greater than or equal to 90% are 242 eliminated. When the RH is less than 30%, the hygroscopic effect of aerosols is very low or even 243 negligible. When the RH is between 30% and 90%, the hygroscopic effect of aerosols is high, and visibility is converted to dry visibility (Yang et al., 2021c), as shown in equation (2). At least 3 244 245 hourly records of meteorological variables are required when calculating the daily average ($n \ge 3$).

246
$$VISD = VIS/(0.26 + 0.4285 * log(100 - RH)),$$
 (2)

247 where VISD is the dry visibility.

248 2.3 Boundary layer height

249 The hourly boundary layer height (BLH) data from 1980 to 2021 are available from the Fifth 250 Generation reanalysis of the European Medium-Range Weather Forecast Center (ERA5) with a 251 resolution of 0.25° x 0.25° (https://cds.climate.copernicus.eu), which is the successor of ERA-252 Interim and has undergone various improvements (Hersbach et al., 2020). The atmospheric 253 boundary layer is the layer closest to the Earth's surface and exhibits complex turbulence activities, 254 and its height undergoes significant diurnal variation. The boundary layer plays a crucial role in 255 regulating and adjusting the distribution of atmospheric aerosols, such as vertical distribution, 256 concentration changes, transport, and deposition (Ackerman et al., 1995). The boundary layer height serves as an approximate measure of the scale height for aerosols (Zhang et al., 2020). 257

- Compared to observations of 300 stations over world from 2012 to 2019, the ERA5 BLH is
 underestimated by 131.96m, and it is closest to the observations compared to JRA-55, and NECP2 BLH (Guo et al., 2021). The hourly BLH data is temporally and spatially matched with visibility
 and other meteorological data before calculating the daily average.
- Because the reciprocal of visibility is proportional to the extinction coefficient and positively related
 to AOD (Wang et al., 2009), we calculate the reciprocal of visibility (VISI) and the reciprocal of dry
 visibility (VISDI). Due to the influence of boundary layer height on the vertical distribution of
 particles (Zhang et al., 2020), we calculate the product (VISDIB) of VISDI and BLH. Therefore,
 the Predictor (Figure 2) is composed of 11 variables (TMP, Td, dT, RH, SLP, WS, VIS, BLH, VISI,
 VISDI, and VISDIB).

268 2.4 MODIS AOD products

Satellite daily AOD data are available from the Moderate Resolution Imaging Spectroradiometer 269 270 (MODIS) Level 3 Collection 6.1 AOD products of the Aqua (MYD09CMA) satellite from 2002 to 271 2021 and Terra (MOD09CMA) satellite from 2000 to 2021 with a spatial resolution of 0.05° x 0.05° 272 at a wavelength of 550 nm (https://ladsweb.modaps.eosdis.nasa.gov). Terra (passing 10:30 am at 273 local time) and Aqua (passing 1:30 pm at local time) are successfully launched in December 1999 274 and May 2002, respectively. MODIS, carried on the Terra and Aqua satellites is a crucial instrument 275 in the NASA Earth Observing System program, which is designed to observe global biophysical 276 processes (Salomonson et al., 1987). The 2330 km-wide swath of the orbit scan can cover the entire 277 globe every one to two days. MODIS has 36 channels and more spectral channels than previous satellite sensors (such as AVHRR). The spectrum ranges from 0.41 to 15 µm representing three 278 279 spatial resolutions: 250 m (2 channels), 500 m (5 channels), and 1 km (29 channels). The aerosol retrievals use seven of these channels (0.47-2.13 µm) to retrieve aerosol characteristics and use 280 281 additional channels in other parts of the spectrum to identify clouds and river sediments. Therefore, 282 it has the ability to characterize the spatial and temporal characteristics of the global aerosol field.

283 The MODIS aerosol product actually uses different algorithms to retrieve aerosols over land. The 284 Dark Target (DT) algorithm is applied to densely vegetated areas because the surface reflectance 285 over dark-target areas is lower in the visible channels and has nearly fixed ratios with the surface 286 reflectance in the shortwave and infrared channels (Levy et al., 2007; Levy et al., 2013). The Deep 287 Blue (DB) algorithm is originally applied to bright land surfaces (such as deserts), and later extended 288 to cover all cloud-free and snow-free land surfaces (Hsu et al., 2006; Hsu et al., 2013). MODIS 289 Collection 6.1 aerosol product is released in 2017, incorporating significant improvements in 290 radiometric calibration and aerosol retrieval algorithms.

291 The aerosol retrievals usually are evaluated by the expected error. For the DT algorithm, the 292 expected error is $\pm (0.05 + 15\% \text{AOD}_{\text{AFRONET}})$. The coverage of retrieval products varies by season based on the DT algorithm over land. Higher spatial coverage is observed in August and September, 293 294 reaching 86-88%. During December and January, due to the presence of permanent ice and snow 295 cover in high-latitude regions of the Northern Hemisphere, the spatial coverage is 78-80%. Thus, 296 challenges remain in retrieving AOD values in high-latitude regions (Wei et al., 2019). However, 297 visibility observations are available in high-latitude regions, thereby partially addressing the lack in these regions. In this study, the Terra and Aqua MODIS AOD are temporally and spatially matched 298 299 with the meteorological stations. Aqua MODIS AOD is used as the Target when training the model, 300 and Terra MODIS AOD is used in the evaluation and validation of the model results, as shown in 301 the flowchart (Figure 2).

302 2.5 Ground-based AOD

303 Ground-based 15-minute AOD observations are available from the Aerosol Robotic Network 304 (AERONET) Version 3.0 Level 2.0 product at 395 sites (Figure 1), which can be downloaded from 305 https://aeronet.gsfc.nasa.gov. The AERONET program is a federation of ground-based remote sensing aerosol networks established by NASA and PHOTONS, including many subnetworks (such 306 307 as AeroSpan, AEROCAN, NEON, and CARSNET). The sun photometer (CE-318) measures 308 spectral sun and sky irradiance in the 340-1020 nm spectral range. AERONET has three levels of AOD products: Level 1.0 (unscreened), Level 1.5 (cloud screened), and Level 2.0 (cloud screened 309 310 and quality assured). Compared to Version 2, the Version 3 Level 2.0 database has undergone further

8

- 311 cloud screening and quality assurance, which is generated based on Level 1.5 data with pre- and
- 312 post-calibration and temperature adjustment and is recommended for formal scientific research
- 313 (Giles et al., 2019). AERONET provides AOD products at wavelengths of 440, 675, 870, and 1020
- nm. When the aerosol loading is low, the error is significant. When the AOD at 440 nm wavelength
- is less than 0.2, the error is 0.01, which is equivalent to the error of the absorption band in the total
- optical depth (Dubovik et al., 2002a). The total uncertainty in AOD under cloud-free conditions is
- less than \pm 0.01, when the wavelength is more than 440 nm, and \pm 0.02 when the wavelength is less than 440 nm (Holben et al., 1998). AERONET AOD is usually considered as the 'true' value. The
- than 440 nm (Holben et al., 1998). AERONET AOD is usually considered as the 'true' value. The
 AOD at 440 nm and the Ångström index at 440-675 nm are used to calculate AOD at 550 nm (not
- 320 provided by AERONET), as shown in equation (3).

321
$$\tau_{550} = \tau_{440} (\frac{550}{440})^{-\alpha},$$
 (3)

322 where τ_{440} and τ_{550} are the AOD at a wavelength of 440 nm and 550 nm, and α is the Ångström 323 index.

The daily average AOD requires at least two observations within 1 hour (\pm 30 minutes) of Aqua/Terra transit time (Wei et al., 2019). The matching conditions between AERONET sites and meteorological stations are (1) a distance of less than 0.5 °, and (2) at least three years of observations. Finally, a total of 395 sites are selected.

328 2.6 AOD reanalysis dataset

329 The monthly AOD (550 nm) dataset of Modern-Era Retrospective Analysis for Research and 330 Applications version 2 (MERRA-2) from 1980 to 2021 is a NASA reanalysis of the modern satellite era produced by NASA's Global Modeling and Assimilation Office with a spatial resolution of 331 332 0.5×0.625° (Gelaro et al., 2017), available at https://disc.gsfc.nasa.gov. MERRA-2 AOD uses an 333 analysis splitting technique to assimilate AOD data at 550 nm. The assimilated AOD observations 334 are including (1) AOD retrievals from AVHRR (1979-2002) over global ocean, (2) AOD retrievals 335 from MODIS on Terra (2000-present) and Aqua (2002-present) over global land and ocean, (3) 336 AOD retrievals from MISR (2000–2014) over bright and desert surfaces, and (4) direct AOD 337 measurements from the ground-based AERONET (1999-2014) (Gelaro et al., 2017). The monthly 338 MERRA-2 AOD is used to evaluate the model's predictive ability before 2000 and after 2000.

339 2.7 Decision tree regression

340 2.7.1 Feature selection

Although a multidimensional dataset can provide as much potential information as possible for 341 342 AOD, irrelevant and redundant variables can also introduce significant noise in the model and reduce the model's accuracy and stability (Kang et al., 2021; Dong et al., 2023). Therefore, the F-343 test is used to search for the optimal feature subset in the Predictor, aiming to eliminate irrelevant 344 345 or redundant features and select truly relevant features, which helps to simplify the model's input 346 and improve the model's prediction ability (Dhanya et al., 2020). The F-test is a statistical test that 347 gives an f-score $(=-\log(p), p)$ represents the degree to which the null hypothesis is not rejected) by 348 calculating the ratio of variances. In this study, we calculate the ratio of variance between the 349 Predictors and Target, and the features are ranked based on the f-score values. A larger value of fscore means that the distances between Predictors and Target are less and the relationship is closer, thus, the feature is more important. We set p=0.05. When the score is less than -log (0.05), the variable in the Predictors is not considered.

353 **2.7.2 Data balance**

354 When the weather is clear, the AOD value is small (AOD < 0.5), and the variability of AOD is small, and 355 the data is concentrated near the mean value. When heavy pollution, the AOD value is large (AOD > 0.5). Compared to clear sky, the AOD sequence will show "abnormal" large values with low frequency, which 356 357 is a phenomenon of imbalance AOD data. When dealing with imbalanced datasets, because of the 358 tendency of machine learning algorithms to perform better on the majority class and overlook the 359 minority class, the model may be underfit (Chuang and Huang, 2023). Data augmentation techniques are 360 commonly employed to address the issue in imbalance data, which applies a series of transformations or 361 expansions to generate new training data, thereby increasing the diversity and quantity of the training 362 data of the minority class.

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

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

375 2.7.3 Decision tree regression model

376 The decision tree is a machine learning algorithm based on a tree-like structure used to solve 377 classification and regression problems. We use regression tree algorithm to construct a regression model 378 by analyzing the mapping relationship between object attributes (Predictor) and object values (Target). 379 The internal nodes have binary tree structures with feature values of "yes" and "no". In addition, each 380 leaf node represents a specific output for a feature space. The advantages of the regression tree include 381 the ability to handle continuous features and the ease of understanding the generated tree structure 382 (Teixeira, 2004; Berk, 2008). Before training the tree model, the variables (Input) are normalized to 383 improve model performance, and after prediction, the results are obtained by denormalization. The 10-384 fold cross-validation method is employed to improve the generalization ability of the model (Browne, 2000). 385

The core problems of the regression tree need to be solved are to find the optimal split variable and optimal split point. The optimal split point of Predictors is determined by the minimum MSE, which in turn determines the optimal tree structure. We set $Y = [y_1, y_2, ..., y_N]$ as the Target. We set X = $[x_1, x_2, ..., x_N]$ as the Predictors, $x_i = (x_i^1, x_i^2, ..., x_i^n)$, i = 1, 2, 3, ..., N, where n is the feature number, and N is the length of sample. We set a training dataset as $D = [(x_1, y_1), (x_2, y_2), ..., (x_N, y_N)]$. A regression tree corresponds to a split in the feature space and the output values on the split domains. Assuming that the input space has been divided into M domains $[R_1, R_2, ..., R_M]$ and there is a fixed output value on each R_M domain, the regression tree model can be represented as follows:

394
$$f(x) = \sum_{m=1}^{M} c_m I(x \in R_M), m = 1, 2, ..., M,$$
 (4)

395 where I is the indicator function, equation (5):

$$396 I = \begin{cases} 1, x \in R_m \\ 0, x \notin R_m \end{cases}, (5)$$

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

$$401 \quad \widehat{c_m} = ave(y_i|x_i \in R_m), \tag{6}$$

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

409
$$L(j,s) = \sum_{x_i \in R_1(j,s)} (y_i - c_1)^2 + \sum_{x_i \in R_2(j,s)} (y_i - c_2)^2,$$
(7)

410 When L(j,s) is the smallest, x^j is the optimal split variable and s is the optimal split point for the 411 x^j .

412
$$\min_{j,s} \left[\frac{\min_{c_1} \sum_{x_i \in R_1(j,s)} (y_i - c_1)^2 + \min_{c_2} \sum_{x_i \in R_2(j,s)} (y_i - c_2)^2 \right],$$
(8)

413 We use the optimal split variable x^{j} and the optimal split point *s* to split the feature space and calculate 414 the corresponding output value.

415
$$\widehat{c_1} = ave(y_i|x_i \in R_1(j,s)), \ \widehat{c_2} = ave(y_i|x_i \in R_2(j,s)),$$
(9)

416 We traverse all input variables to find the optimal split variable x^{j} , forming a pair (j, s). Divide the 417 input space into two regions accordingly. Next, repeat the above process for each region until the stop 418 condition is met. The regression tree is generated.

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

420
$$f(x) = \sum_{m=1}^{M} \widehat{c_m} I(x \in R_M), m = 1, 2, ..., M,$$
 (10)

421 **2.8 Evaluation metrics**

- 422 Evaluation metrics, including Root Mean Squared Error (RMSE), Mean Absolute Error (MAE) and
- 423 Pearson Correlation Coefficient (R), are used to evaluate the performance and accuracy of the model

424 results.

425
$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (y_i - \hat{y}_i)^2},$$
 (11)

426
$$MAE = \frac{1}{n} \sum_{i=1}^{n} |\mathbf{y}_i - \hat{\mathbf{y}}_i|, \qquad (12)$$

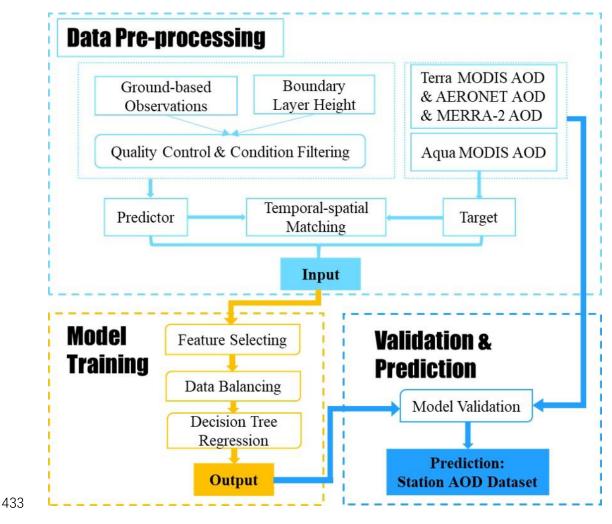
427
$$\boldsymbol{R} = \frac{\sum_{i=1}^{n} (y_i - \bar{y}) (\hat{y}_i - \bar{y})}{sqrt(\sum_{i=1}^{n} (y_i - \bar{y})^2 \sum_{i=1}^{n} (\hat{y}_i - \bar{y})^2)},$$
(13)

428 where y_i and \bar{y} are the predicted value and the average of the predicted values. \hat{y}_i and \bar{y} are 429 the target and the average of the target. i = 1, 2, ..., n is the length of sample.

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

431
$$EE = \pm (0.05 + 0.15 * \tau_{true}),$$
 (14)

432 where τ_{true} is the AOD at 550 nm from AERONET, satellite and reanalysis datasets.



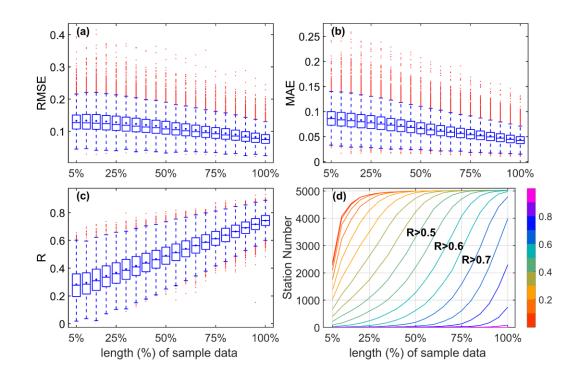
434 **Figure 2:** Flowchart for deriving aerosol optical depth (AOD).

435 **2.9 Workflow**

⁴³⁶ Figure 2 summarizes the flowchart and provides an overview of the structure of this study, which

437 involves four main parts: (1) data preprocessing, (2) model training, and (3) validation and438 prediction.

439 **3 Results and discussion**



440 **3.1 Dependence of model performance on training data length**

441



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

447 We build the models using different lengths of sample data (5% to 100%, with a 5% interval) by random 448 allocation without overlap and evaluate the predictive performance of each model. Figure 3 (a-c) depicts 449 RMSE, MAE, and R between the predicted values and target based on the training data of 5% to 100% 450 sample data at a station. As the volume of the training data increases, the RMSE and MAE values 451 decrease, and the R values increase. Compared to 5% of the sample data, the result of 100% sample data 452 shows a decrease in RMSE by 41.1%, a decrease in MAE by 50.1%, and an increase in R by 162.3%. 453 The relationship between the length of sample data and the model's performance is positive for each 454 station. Figure 3 (d) shows that R of approximately 70% stations is greater than 0.5 at 50% of the sample 455 data, while at 75%, the R of approximately 80% of stations is greater than 0.6. When 100% of the sample 456 data is used as sample data, the R of approximately 80% of stations is greater than 0.75, and the R of 457 about 97% is greater than 0.7. This finding indicates that the predictive capability and robustness of the 458 model increase as the amount of training data increases. It may be attributed to the model's ability to 459 capture more complex patterns and relationships among the input by multi-year data.

460 **3.2 Evaluation of model training performance**

461 Figure 4 shows the spatial distribution (a-c) and frequency and cumulative frequency (d-e) of RMSE,

462 MAE, and R of all stations. The mean values of RMSE, MAE and R are 0.078, 0.044, and 0.750,

respectively. The RMSE of 93% stations is less than 0.11, the MAE of 91% is less than 0.06, and the R

464 of 88% is greater than 0.7. The R values in Africa, Asia, Europe, North America, Oceania, and South

- 465 America are 0.763, 0.758, 0.736, 0.750, 0.759, and 0.738, respectively. Although the RMSE and MAE
- 466 of a few stations are high in America and Asia, the R is still high (> 0.6). Therefore, the results of the
- 467 model's errors demonstrate that the model performs well on almost all stations.

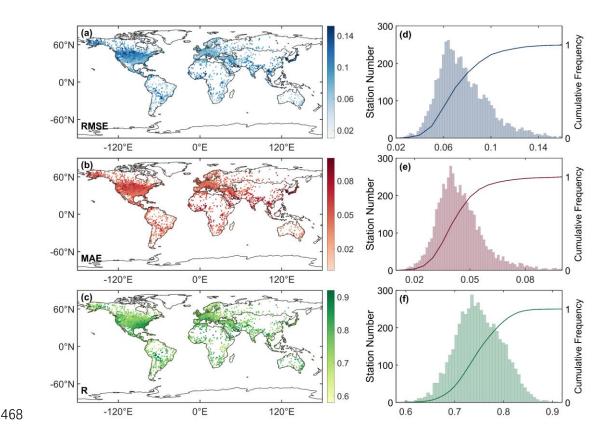


Figure 4: Spatial distribution (a-c) of root mean squared error (RMSE), mean absolute error (MAE),
and correlation coefficient(R) between the model's result and target with 100% sample data. Station
number (bar) and cumulative frequency (curve) (d-e) of RMSE, MAE, and R.

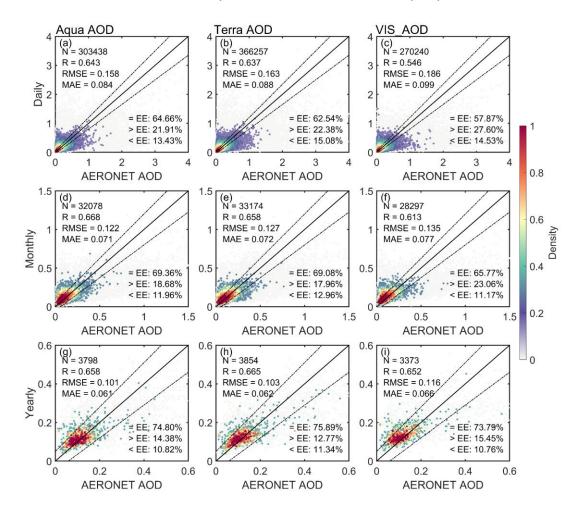
472 **3.3 Validation and comparison with MODIS and AERONET AOD**

473 **3.3.1 Validation over global land**

To validate the model's predictive ability, the visibility-derived AOD (for short, VIS_AOD) is compared
with Aqua, Terra, MERRA-2 and AERONET AOD at 550 nm for the global scale. Among them, Aqua
AOD has been used as training data, which is not an independent dataset. Terra AOD and AERONET
AOD have not been used as training data and can be regarded as independent datasets.

- First, the relationship among daily MODIS and AERONET AOD is evaluated, as shown in Figure 5 (ab, d-e, g-h). The R values with Aqua AOD and Terra AOD are 0.643 and 0.637 on the daily scale, and
 0.668 and 0.658 on the monthly scale, 0.658 and 0.665 on the yearly scale. The RMSE with Aqua AOD
- and Terra AOD are 0.158 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 scale. The MAE values with Aqua AOD and Terra AOD are 0.084 and 0.088 on
the daily scale, and 0.071 and 0.072 on the monthly scale, 0.061 and 0.062 on the yearly scale. The
percentages of sample point falling within the EE envelopes are 64.66% and 62.54% on the daily scale,
and 69.36% and 69.08% on the monthly scale, 74.80% and 75.89% on the yearly scale.



486

Figure 5: Scatter density plots between AERONET AOD (550 nm) and Aqua MODIS AOD, Terra 487 488 MODIS AOD and VIS AOD on the daily (a-c), monthly (d-f) and yearly (g-i) scale. The solid black line 489 represents the 1:1 line and the dashed lines represents expected error (EE) envelopes. The sample size 490 (N), correlation coefficient (R), mean absolute error (MAE), and root mean square error (RMSE) are 491 given. '= EE', '> EE', and '< EE' represent the percentages (%) of retrievals falling within, above, and 492 below the EE, respectively. The matching time for Aqua AOD and VIS AOD with AERONET AOD is 493 13.30 (\pm 30 minutes) at local time, and the matching time between Terra AOD and AERONET AOD is 494 $10.30 (\pm 30 \text{ minutes})$ at local time.

Figure 6 shows the scatter density plots and the EEs between VIS_AOD and Aqua AOD, Terra AOD, and AERONET AOD. Aqua AOD is not an independent validation, and Terra and AERONET AOD are independent validation. For the daily scale, the R, RMSE and MAE of between VIS_AOD and Aqua AOD (15,962,757 pairs data) is 0.799, 0.079 and 0.044, respectively. The percentage of sample point falling within the EE envelopes is 84.12% on the global scale (Figure 6 a). The R between VIS_AOD and Terra AOD (17,145,578 pairs data) is 0.542, with a RMSE of 0.125 and MAE of 0.078. The percentage falling within the EE envelopes is 64.76% (Figure 6 b). The R between VIS_AOD and

15

AERONET AOD (270,240 pairs data) at 395 sites is 0.546, with a RMSE of 0.186 and MAE of 0.099.
The percentage falling within the EE envelopes is 57.87% (Figure 6 c).

504 For the monthly and yearly scales, RMSE and MAE show a significant decrease between VIS AOD and 505 Aqua, Terra, and AERONET AOD, and R and percentages falling within EE show a significant increase 506 in Figure 6 (e-g, i-k). The monthly RMSEs are 0.029, 0.051, and 0.135, the monthly MAEs are 0.018, 507 0.031, and 0.077, and the monthly R values are 0.936, 0.808, and 0.613, respectively. The percentages falling within the EE envelopes are 98.34%, 93.25%, and 65.77%. The RMSEs on the yearly scale are 508 509 0.013, 0.024, and 0.116, the MAEs are 0.008, 0.015, and 0.066, and the R values are 0.976, 0.906, and 510 0.652, respectively. The percentages falling within the EE envelopes are 99.82%, 99.20%, and 73.79%. 511 The percentage falling within the EE envelopes against AERONET is smaller than that against Terra, 512 which may be related to the elevation of AERONET sites, the distance between AERONET and 513 meteorological stations, and observed time. The results highlighted above demonstrate a clear 514 improvement in performance on the monthly and yearly scales compared to the daily scale.

515 To further examine the predictive capability of historical data, we compare the VIS AOD with 516 AERONET AOD before 2000, as shown in Figure 6 (d, h, l). We match 43 AERONET sites, with a total 517 of 5166 daily records. The result indicates that the daily-scale R is close to that after 2000 (Figure 6 c), 518 with the percentages approaching 50% falling within the EE envelopes. The monthly and annual 519 correlation coefficients are even higher, with a percentage of 55% falling within the EE envelopes. 520 Although the sample size is small, it still demonstrates the excellent predictive ability of the model. 521 Compared with AERONET (an independent validation dataset), the performance of VIS AOD is almost 522 unchanged before and after 2000.

523 We also compare the VIS AOD with the MERRA-2 reanalysis AOD on the monthly scale, as shown in 524 Figure 7. The correlation coefficient between MERRA-2 and AERONET is 0.655 before 2000, slightly 525 lower than the correlation coefficient (0.657) between VIS AOD and AERONET. The correlation 526 coefficient between MERRA-2 and AERONET is 0.829 after 2000, significantly higher than that before 527 2000, while the correlation coefficient between VIS AOD and AERONET is 0.613. It suggests that 528 VIS AOD and MERRA-2 AOD have similar accuracy before 2000. The correlation of MERRA-2 after 529 2000 is higher and even performs better than MODIS retrievals (as shown in Figure 5) when evaluated 530 at AERONET sites. However, before 2000, the correlation coefficient of MERRA-2 and AERONET, 531 RMSE, and MAE all show significant changes and differences in consistency. The higher correlation 532 between MERRA-2 and AERONET AOD is partly because MERRA-2 has assimilated AERONET AOD 533 observations (Gelaro et al., 2017). Compared to AERONET, VIS AOD and Aqua/Terra MODIS have a 534 similar correlation coefficient. The correlation coefficient of VIS AOD before 2000 is even higher than 535 after 2000, and the changes in RMSE and MAE are not significant. It indicates good consistency of 536 VIS AOD. In conclusion, the predicted results have good consistency with AEONET AOD and Terra 537 AOD on the daily scale. The monthly and annual results have a significant improvement. The model shows good predictive capabilities before/after 2000, highlighting the stable accuracy of VIS_AOD. 538

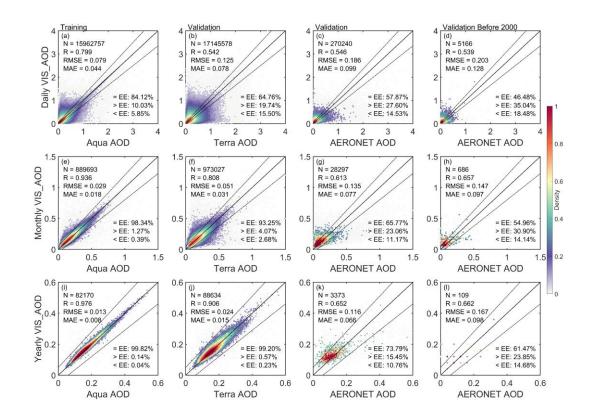


Figure 6: Scatter density plots between predicted AOD (VIS AOD) and Aqua MODIS AOD, Terra 540 MODIS AOD, AERONET AOD and AERONET AOD before 2000 on the daily (a-d), monthly (e-h) and 541 yearly (g-i) scale. The solid black line represents the 1:1 line and the dashed lines represents expected 542 543 error (EE) envelopes. The sample size (N), correlation coefficient (R), mean absolute error (MAE), and 544 root mean square error (RMSE) are given. '= EE', '> EE', and '< EE' represent the percentages (%) of retrievals falling within, above, and below the EE, respectively. Note Aqua AOD is not an independent 545 validation dataset for predicted results, while Terra and AERONET AOD are independent validation 546 547 datasets.

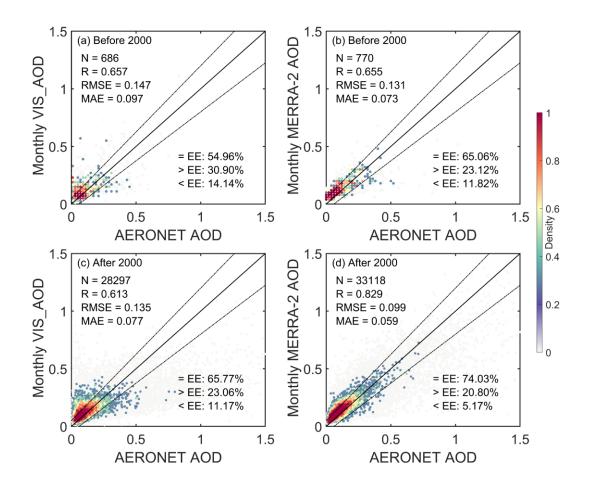


Figure 7: Scatter density plots between AERONET AOD and the predicted AOD (VIS_AOD) and MERRA-2 AOD before/after 2000 on the monthly scale. The solid black line represents the 1:1 line and the dashed lines represents expected error (EE) envelopes. The sample size (N), correlation coefficient (R), mean absolute error (MAE), and root mean square error (RMSE) are given. '= EE', '> EE', and '< EE' represent the percentages (%) of retrievals falling within, above, and below the EE, respectively.

554 3.3.2 Validation over regions

Aerosol loading exhibits spatial variability. Evaluation metrics for the relationships between visibility-derived AOD and AERONET AOD and Terra AOD for each region are listed in Table 1.

557 In Europe and North America, the results are similar to those of Terra and AERONET, with a large 558 number of data pairs, greater than 10^5 (AERONET) and greater than 10^7 except for Eastern Europe 559 (Terra) on the daily scale. Approximately 63% -70% data pairs fall within the EE envelopes. The 560 RMSE is approximately 0.11, except for western North America (~0.15), and the MAE is 561 approximately 0.07, and the correlation coefficient is between 0.44 and 0.54.

562 In Central South America, South Africa, and Australia, data pairs are about 10^{3-4} (AERONET) and 563 10^6 (Terra) on the daily scale. 52-60% fall within the EE envelopes compared to AERONET, and 564 58-67% compared to Terra. The RMSE is 0.03-0.05 compared to Terra, and 0.11-0.17 compared to 565 AERONET. The correlation coefficient ranges from 0.40 to 0.74, with the highest correlation 566 coefficient in South America at 0.74.

- 567 In Asia, India, and West Africa, the data pairs are only approximately 10⁴ (AERONET). 32% to 50%
- fall within the EE envelopes compared to AERONET, the RMSE value ranges from 0.20 to 0.50,
- and the MAE ranges from 0.11 to 0.36. Compared to Terra AOD, 51 to 58% of data pairs fall within
- 570 the EE envelopes, the RMSE is around 0.16, and the MAE is around 0.11. Compared to AERONET,
- in these high aerosol loading regions, RMSE and MAE increase, and the percentages falling within
- 572 the EE envelopes decrease, but the correlation coefficients do not significantly decrease.
- 573 Compared to Terra AOD, 55% -67% of data falls within the EE envelopes on the daily scale, 87% -574 96% on the monthly scale, and over 97% on the yearly scale. Compared to AERONET AOD, 32-575 68% of data falls within the EE envelopes, 24% -84% on the monthly scale, and 15% -97% on the 576 yearly scale. On both monthly and yearly scales, all metrics have shown a significant increase in 577 performance when compared to Terra. However, compared to AERONET, not all metrics increase 578 in some regions due to limited data pairs, such as West Africa, Northeast Asia, and India, which may
- be due to the spatial differences between AERONET sites and meteorological stations.

580 **3.3.3 Validation at a site scale**

- 581 Sites, especially AERONET, are not completely uniform across the world or in any region, and
- 582 different stations have different sample sizes, which may lead to a certain uncertainty. Therefore,
- 583 further analysis is conducted on the spatial distribution of different evaluation metrics. Figure 8
- shows the validation and comparison of daily VIS_AOD against Terra and AERONET AOD at a
- 585 site scale.
- 586 Compared to Terra daily AOD, the R of 67% stations is greater than 0.40, the mean bias of 83% is

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

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

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

- less than 0.01, the RMSE of 85% is less than 0.15, and the percentage falling within the EE of 67%
- 590 is greater than 60%. More than 85% of stations falling within the EE is greater than 60% in Europe,
- 591 North America, and Oceania, while 40-60% in South America, Africa, and Asia. The percentage of
- 592 expected error is low in South and East Asia, and Central Africa, with some underestimation. Above
- 593 60% in Africa, Asia, North America, and Europe have a correlation coefficient greater than 0.40.
- 594 The regions with lower correlation are the coastal regions of South America, eastern Africa, western
- 595 Australia, northeastern North America, and northern Europe. Above 90% of the RMSE in Europe,
- 596 North America, and Oceania are smaller than 0.15. High RMSE regions are in western North
- 597 America, Asia, central South America, and central Africa.
- 598 Compared to AERONET daily AOD, the R of 74% stations is greater than 0.40, and the spatial distribution is similar to Terra's. The mean bias of 44% is less than 0.01, the RMSE of 68% is less 599 600 than 0.15, and the percentage falling within the EE of 53% is greater than 60%. More than 70% of sites have a correlation coefficient greater than 0.40 in Africa, Asia, Europe, and North America. 601 602 More than 57% of sites have an expected error percentage of over 60% in Europe, North America, and Oceania, except for Asia. Over 72% of sites have a RMSE less than 0.15. Except for Oceania 603 604 and South America, over 71% of sites in other regions have MAEs less than 0.01. Almost all sites 605 in Asia show a negative bias, significantly underestimating. However, there is a significant 606 overestimation in western North America and western Australia. Most sites in Asia falling within 607 the expected error are less than 50%. High RMSE are in high emission and dust areas, such as Asia, 608 India, and Africa.
- 609 The validation and comparison on the site scale show a limitation similar to the MODIS DT 610 algorithm. In areas with high vegetation coverage, the AOD from visibility are better than those in bright areas. Although the correlation coefficients are high in high aerosol loading areas (Central 611 South America, West Africa, India, Eastern China, Northeast Asia), there are significant differences 612 613 in these areas with high RMSE values. As shown in Figure 6, some stations located in dusty and urban areas are overestimated or underestimated. Studies have shown that there is a significant 614 uncertainty in the MODIS retrievals in these regions, and the challenges of inversion algorithms are 615 616 significant in bright surfaces (desert and snow covered areas) and urban surface of densely populated complex structures (Chu et al., 2002; Remer et al., 2005; Levy et al., 2010; Wei et al., 617 618 2019; Wei et al., 2020). In India, the elevation difference between AERONET site and meteorological station reached 0.7 km may be a factor affecting the validation effect, as aerosol 619 varies greatly with altitude. In eastern China, the complex urban surface, emission sources, and 620 621 observations in different locations (AERONET site and meteorological station) may be the reasons 622 for underestimation. At the same time, visibility stations in desert areas are sparse, and the spatial 623 variability of dust aerosols is large, which also increases the difficulty to estimate VIS AOD.

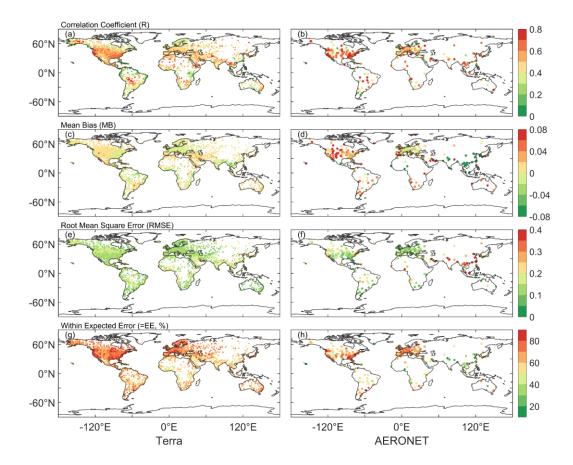


Figure 8: Validation of VIS_AOD against Terra and AERONET AODs at each site: (a–b)
correlation (R), (c-d) mean bias (MB), (e-f) root mean square error (RMSE), (g-h) percentage (%)
of VIS_AOD within the expected error envelopes.

628 3.3.4 Discussion and uncertainty analysis

The atmospheric visibility is a surface physical quantity, while AOD is a column-integrated physical 629 quantity. We have linked the two variables together using machine a learning method, which 630 631 partially compensates for the scarcity of AOD data. However, we have to face some limitations. 632 Although the boundary layer height is considered, it is not sufficient. Pollutants such as smoke from 633 biomass burning, dust, volcanic ash, and gas-aerosol conversion of sulfur dioxide to sulfate aerosols in the upper and lower troposphere can undergo long-range aerosol transport under the influence of 634 circulation. The pollution transport and aerosol conversion processes above the boundary layer are 635 636 still significant and cannot be ignored (Eck et al., 2023). Compared to surface visibility, bias occurs 637 when the aerosol layer rises and affects AERONET measurements and MODIS retrievals. Therefore, it should be considered when using this data. If there are sufficient historical vertical aerosol 638 639 measurements with high temporal and spatial resolution, the results of this data would be greatly 640 improved. Although some studies use aerosol profiles from pollution transport models or assumed profiles as substitutes for observed profiles (Li et al., 2020; Zhang et al., 2020), the biases introduced 641 642 by these non-observed profiles are still significant.

In machine learning, we use MODIS Aqua AOD as the target value for the model because the validation results for MODIS C6.1 product have a correlation coefficient of 0.9 or higher with AERONET AOD on the daily scale (Wei et al., 2019; Wei et al., 2020). Compared to AERONET, MODIS AOD provides more sample data with a high global coverage. However, apart from modeling errors, the systematic biases and uncertainties of MODIS Aqua AOD cannot be ignored (Levy et al., 2013; Levy et al., 2018; Wei et al., 2019). Averaging over time scale can reduce representation errors effectively, and emission sources and orography can increase representation errors (Schutgens et al., 2017). Therefore, the strong correlation at monthly and annual scales indicates a substantial reduction in errors. This is also one of the reasons why this dataset shows stronger correlation with Terra AOD and weaker correlation with AERONET in validation.

The spatial matching between meteorological stations and AERONET sites may cause some biases. 653 654 AERONET sites are usually not co-located with meteorological stations in terms of elevation and 655 horizontal distance, this is another reason for the weak correlation between VIS AOD and AERONET AOD. The meteorological stations are located at the airport. Different horizontal 656 distances may result in meteorological stations and AERONET sites being located on different 657 658 surfaces (such as urban, forest, mountainous). Differences in site elevation significantly impact the 659 relationship between AOD and measured visibility. When the AERONET site is at a higher elevation than the meteorological station, there may be fewer measurements of aerosols over the sea at the 660 AERONET site. 661

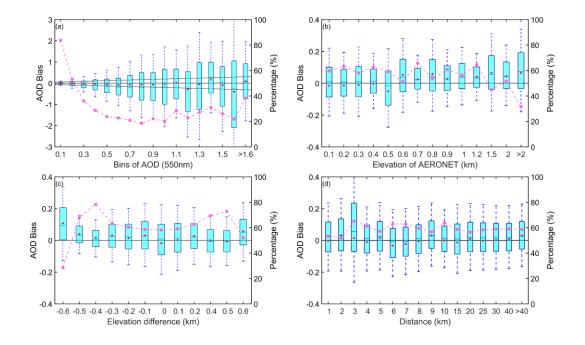
Different pollution levels and station elevation affect the AOD derived from visibility. The elevation
difference and distance between meteorological stations and AERONET sites also have an impact
on the validation results. Therefore, the error and performance of different AERONET AOD values,
station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

As the AOD increases, the variability of bias also increases in Figure 9 (a). Almost all mean bias values are within the envelope of EE, except for 1.1-1.2 and 1.5-1.6. The average bias is 0.015 (AOD < 0.1), with 83% of data within the EE envelopes. The mean bias is -0.0011 (AOD, 0.1-0.2), with 54% within the EE envelopes. The mean bias is negative (AOD, 0.3-1.0), with 20%-40% falling within the EE envelopes. There is a positive bias (AOD, 1.1, 1.4 and > 1.6), and there is a negative bias at 1.2-1.3 and 1.5-1.6. The results indicate that as pollution level increases, the negative mean bias becomes significant and the underestimation increases.

The contribution of aerosols near the ground to the column aerosol loading is significant. The elevation of the site affects the measurement of column aerosol loading in Figure 9 (b). There is a negative bias in the low elevation (≤ 0.5 km) with a percentage of 60%-64% falling within the EE envelopes and a positive bias in high elevation (0.5-1.2 km) with a percentage of 50%-65% falling within the EE envelopes. The percentage significantly decreases (> 1.2 km), and the average bias increases. Therefore, the elevation of AERONET's site will cause bias in validation, and the uncertainty greatly increases in high elevation.

680 Due to the elevation difference between the meteorological station and AERONET site in the 681 vertical direction, the uncertainty caused by elevation differences of site is analyzed in Figure 9 (c). When the elevation difference is negative (the elevation of the meteorological station is lower than 682 683 that of the AERONET station), there is a significant positive bias. When the difference is positive, 684 the mean bias approaches 0 or is positive. The percentage is greater than 60% (-0.5 km-0.5 km). 685 The positive mean bias is greater than the negative mean bias, and the uncertainty greatly increases 686 when the elevation of meteorological stations is lower than that of AERONET sites. It indicates that 687 the contribution of the near surface aerosol to the column aerosol loading is significant and cannot 688 be ignored.

The spatial variability of aerosols is significant. Meteorological stations and AERONET sites are 689 690 not collocated, resulting in a certain distance in spatial matching. In this study, the upper limit of 691 distance is 0.5 degree. Figure 9 (d) shows the error of the distance between stations, where the 692 degree is converted to the distance at WGS84 coordinates. The bias does not change significantly 693 with increasing distance. The average bias is around 0, with the maximum positive mean bias (0.0322) at a distance of 2 km and the maximum negative mean deviation (-0.0323) at 6 km. The 694 695 median is almost positive, except at 5 km and 6 km. The percentage falling within the EE envelopes 696 is over 50%, with the maximum percentage (66%) at 3km and the minimum (62%) at 2 km.



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Figure 9: Box plots of AOD bias and the percentage falling within the EE envelopes (curves): (a) 698 699 AERONET AOD levels, (b) elevation of AERONET sites, (c) elevation difference between 700 meteorological stations and AERONET sites, (d) distance (km) between meteorological stations and 701 AERONET sites. The black horizontal line represents the zero bias. For each box, the upper, lower, 702 and middle horizontal lines, and whiskers represent the AOD bias 75th and 25th percentiles, median, 703 and 1.5 times the interquartile difference, respectively. The black solid lines represent the EE 704 envelopes (\pm (0.05 + 0.15*AOD_{AERONET})). No site with a difference of + 0.3 km (x-axis label without 0.3) in (c). 705

706 **3.4 Interannual variability and trend of visibility-derived AOD over global land**

The multi-year average AOD from 1980 to 2021 over land is 0.177, as shown in Figure 10 (a). The average is 0.178 in Northern Hemisphere (NH, 4532 stations) and 0.174 in Southern Hemispheres (SH, 500 stations). Due to the influence of geography, atmospheric circulation, population, and emissions, the AOD varies in different latitudes. Figure 11 illustrates the multi-year average AOD in different latitude ranges from 1980 to 2021. The AOD value in the NH is higher than that over land, then higher than that in the SH. Within [-20, 20°N], the average AOD reaches its maximum (0.225), and the maximum AOD in the NH is 0.239 in [0, 20°N]. The highest AOD in the SH is 0.203 in in [-15, 0°N]. The average AOD rapidly decreases from -15°N to -35°N in the SH and from
20°N to 50°N in the NH.

716 There are many regions of high AOD values occur in the NH, with the distribution of high 717 population density. Approximately 7/8 of the global population resides in the NH, with 50% 718 concentrated at 20°N-40°N (Kummu et al., 2016), indicating a significant impact of human activities 719 on aerosols. The highest AOD values are observed near 17°N, including the Sahara Desert, Arabian 720 Peninsula, and India, suggesting that in addition to anthropogenic sources, deserts also play a crucial 721 role in aerosol emissions. Lower AOD regions of the SH are from 25°S to 60°S, encompassing 722 Australia, southern Africa, and southern South America, indicating lower aerosol burdens in these 723 areas. Additionally, North America also exhibits low aerosol loading. Chin et al. (2014) analyzed 724 the AOD over land from 1980 to 2009 with the Goddard Chemistry Aerosol Radiation and Transport 725 model, which is similar to the visibility-derived AOD. The spatial distribution is consistent with the satellite results (Remer et al., 2008; Hsu et al., 2012; Hsu et al., 2017; Tian et al., 2023). The AOD 726 727 and extinction coefficient retrieved from visibility show a similar distribution at global scale, with 728 a correlation coefficient of nearly 0.6 (Mahowald et al., 2007). Similar global (Husar et al., 2000; 729 Wang et al., 2009) and regional (Koelemeijer et al., 2006; Wu et al., 2014; Boers et al., 2015; Zhang 730 et al., 2017; Zhang et al., 2020) spatial distributions have been reported.

731 AOD loadings exhibit significant seasonal variations worldwide, particularly over land. In this study, 732 a year is divided into four parts: December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), and September-October-November (SON), corresponding to winter 733 734 (summer), spring (autumn), summer (winter), and autumn (spring) in the NH (SH), respectively. 735 Figure 10 (b-e) also depicts the spatial distribution of seasonal average AOD over land from 1980 736 to 2021. The global AOD in DJF, MAM, JJA, and SON is 0.161, 0.176, 0.204, and 0.164, 737 respectively. The standard bias of AOD in JJA and DJF are greater than those in DJF and SON. 738 AOD exhibits seasonal changes, with the highest in JJA, followed by DJF, MAM and SON.

- 739 In the NH, the AOD ranking is summer (0.210) > spring (0.176) > autumn (0.163) > winter (0.160).740 In the SH, the AOD ranking from high to low in season is spring (0.188) > summer (0.184) > autumn
- (0.164) > winter (0.152). The highest AOD is observed during JJA in the NH, while in the SH, the
- 742 peak occurs during SON. The high AOD value is highly associated with the growth of hygroscopic
- 743 particle and the photochemical reaction of aerosol precursors under higher relative humidity in Asia
- 744 (JJA) (Remer et al., 2008) and Europe such as Russia (JJA), and biomass burning in South America
- (SON), Southern Africa (SON), and Indonesia (SON) (Ivanova et al., 2010; Krylov et al., 2014). On
- the other hand, the lowest global AOD values are observed during winter, which may be attributedto the atmospheric circulation systems (Li et al., 2016; Zhao et al., 2019).
- The temporal variations in AOD have also been of great interest due to the significant relationship between aerosols and climate change. Figure 10 (f) shows the trends of annual average AOD (** represents passing the significance test, p < 0.01) over the global land, the SH and the NH during 1980-2021. The global land, NH, and SH trends demonstrate decreasing trends of AOD with values of -0.0029/10a, -0.0030/10a, and -0.0021/10a, respectively, with all passing the significance test. The declining trend is much greater in the NH than in the SH.
- The seasonal trends of AOD during 1980-2021 at the global and hemispheric scales are shown in
 Figure 10 (g-j). The trend over land is decreasing in DJF, JJA and SON, and increasing in MAM.

- The largest declining trend is observed in SON (-0.0055/10a). In the NH, the trends are -0.0044/10a (DJF), 0.0016/10a (MAM), -0.0024/10a (JJA), and -0.0064/10a (SON). In the SH, the trends are as follows: 0.0022/10a (DJF), -0.0044/10a (MAM), -0.0064/10a (JJA), and 0.0033/10a (SON). The largest declining trend is SON in the NH and JJA in the SH. However, the trends are positive in
- 760 MAM of the NH and DJF and SON of the SH.

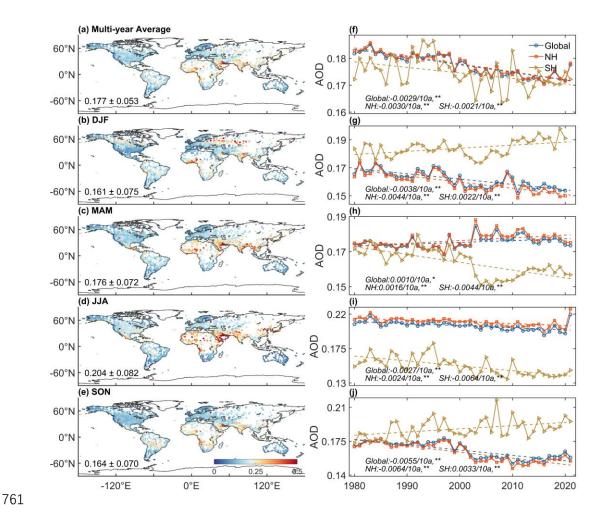
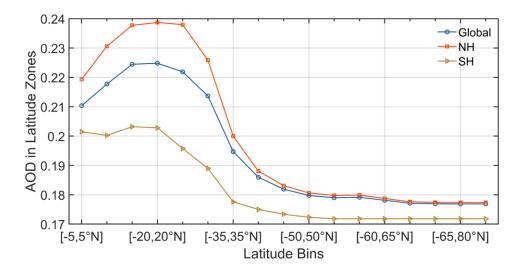


Figure 10: The map of annual and seasonal mean AOD (left) and global/regional mean time series from 1980 to 2021 (right). Global land (circle), northern hemisphere (NH) (triangle) and southern hemisphere (SH) (square) annual and seasonal AOD. The symbol, **, represents that the trend passed the test at a significance level of 0.01. The symbol, *, represents that the trend passed the test at a significance level of 0.05. DJF represents December and next January and February. MAM represents March, April, and May. JJA represents June, July, and August. SON represents September, October, and November.



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Figure 11: The global land (blue), northern hemisphere's (red) and southern hemisphere's (yellow)
multi-year average VIS_AOD from 1980 to 2021 in different latitude zones. The latitude range is
from -65 to 85°N, with a bin of 5°.

773 3.5 Interannual variability and trend of visibility-derived AOD over regions

774 The distribution of AOD over global land exhibits significant spatial heterogeneity. Large variations in aerosol concentrations exist among different regions, leading to a non-uniform spatial distribution 775 776 of AOD globally. Accurately assessing the long-term trends of aerosol loading is a key for quantifying aerosol climate change, and it is crucial for evaluating the effectiveness of 777 778 measurements implemented to improve regional air quality and reduce anthropogenic aerosol 779 emissions. Therefore, we select 12 representative regions to analyze the variability and trend of AOD, which are influenced by various aerosol sources (Wang et al., 2009; Hsu et al., 2012; Chin et 780 781 al., 2014), such as desert, industry, anthropogenic emissions, and biomass burning emissions, which 782 nearly cover the most land and are densely populated regions (Kummu et al., 2016). These 783 representative regions are Eastern Europe, Western Europe, Western North America, Eastern North 784 America, Central South America, Western Africa, Southern Africa, Australia, Southeast Asia, 785 Northeast Asia, Eastern China, and India, as shown in Figure 1.

- The multi-year average and seasonal average AOD (Figure 12), the trends of the annual average of
 monthly anomalies (Figure 13), and the seasonal trends (Figure 14) are analyzed in 12 regions from
 1980 to 2021.
- The regions with a high aerosol level (AOD > 0.2) are in West Africa, Southeast and Northeast Asia,
 Eastern China, and India. The AOD values range from 0.15 to 0.2 in Eastern Europe, Western
 Europe, Eastern North America, Central South America, and South Africa. The AOD values are less
- than 0.15 in Western North America and Australia.
- Europe is an industrial region with a low aerosol loading region, and the multi-year average AOD
- in Eastern Europe (0.181) is higher than that in Western Europe (0.163) during 1980-2021. Eastern
- Europe shows a greater downward trend in AOD (-0.0067/10a) compared to Western Europe (-
- 796 0.0026/10a). The highest AOD is observed in JJA, the dry period when solar irradiation and
- 797 boundary layer height increase, with AOD values of 0.201 in Eastern Europe and 0.162 in Western

- 798 Europe, which could be due to increases in secondary aerosols, biomass burning, and dust transport 799 from the Sahara (Mehta et al., 2016). However, there are seasonal variations. In Eastern Europe, the seasonal AOD ranking from high to low is JJA (0.201) > DJF (0.181) > MAM (0.175) > SON800 (0.161), while in Western Europe, it is JJA (0.193) > MAM (0.162) > SON (0.160) > DJF (0.138). 801 802 The differences among seasons are larger in Western Europe. AOD in Eastern Europe shows 803 declining trends (p < 0.01) in all seasons, and the largest declining trend is in DJF (-0.0096/10a). In Western Europe, the AOD in DJF, JJA, and SON exhibits declining trends, while the AOD in MAM 804 805 shows a significant increasing trend (0.0019/10a). The trends in both Western and Eastern Europe 806 are increasing in MAM from 1995 to 2005 with Western Europe showing a greater increasing trend. 807 However, after 2005, the decline rates accelerate in each season. Studies have shown the downward 808 trend in Europe is attributed to the reduction of biomass burning, anthropogenic aerosols, and 809 aerosol precursors (such as sulfur dioxide) (Wang et al., 2009; Chin et al., 2014; Mortier et al., 2020).
- 810 North America is also an industrial region with a low aerosol loading. The average AOD values in 811 Eastern and Western North America during 1980-2021 are 0.165 and 0.146, respectively, with the Eastern region being higher than the Western region by 0.019. From 1980 to 2021, both Eastern (-812 813 0.0027/10a) and Western North America (-0.0017/10a) show a downward trend. The AOD values 814 in DJF, MAM, JJA, and SON in Western North America are 0.141, 0.148, 0.163, and 0.130, respectively, and 0.138, 0.156, 0.216, and 0.149 in Eastern North America. Specifically, the trends 815 816 of the Western and Eastern region are increasing during MAM and decreasing during other seasons. 817 In the Western region, the trend is increasing after 2005, while in the Eastern region, there is no 818 increasing trend. The increasing trend may be due to low rainfall and increased wildfire activities 819 (Yoon et al., 2014). The decrease in Eastern North America is related to the reduction of sulfate and 820 organic aerosols, as well as the decrease in anthropogenic emissions caused by environmental 821 regulations (Mehta et al., 2016).
- 822 Central South America is a relatively high aerosol loading region, sourced from biomass burning, 823 especially in SON (Remer et al., 2008; Mehta et al., 2016), with a multi-year average AOD of 0.198. 824 There is a downward trend (-0.0075/10a) from 1980 to 2021. The trend is slightly lower than the 825 trend (-0.0090/10a) from 1998 to 2010 (Hsu et al., 2012) and the trend is decreasing from 1980 to 826 2006 (Streets et al., 2009) and from 2001 to 2014 (Mehta et al., 2016). The AOD values in DJF 827 (0.207) and SON (0.228) are higher compared to the values in MAM (0.185) and JJA (0.171), and the larger declining trends are observed in MAM (-0.0100/10a) and JJA (-0.0150/10a). The result 828 829 indicates that although AOD has decreased overall, the aerosol loading is still high, which is caused 830 by deforestation and biomass burning (Mehta et al., 2016).
- 831 Africa is a high aerosol loading region worldwide. In West Africa, the multi-year average AOD is 0.281, and the trend is decreasing (-0.0062/10a) from 1980 to 2021. The world's largest desert 832 833 (Sahara Desert) is in West Africa, with much dust emission. The AOD values in JJA (0.296), MAM 834 (0.292), DJF (0.276) and SON (0.261) are above 0.26. The trends in DJF (-0.0145/10a), MAM (-0.0015/10a), JJA (-0.0019/10a) and SON (-0.0078/10) are decreasing. For South Africa, the multi-835 836 year average AOD is 0.182, lower than that of West Africa. The trend is decreasing (-0.0016/10a). 837 The results of AERONET observations and simulation also show a decreasing trend (Chin et al., 838 2014). The AOD values range from 0.12 to 0.20 during 2000-2009, dominated by fine particle 839 matter from industrial pollution from biomass and fossil fuel combustion (Hersey et al., 2015). The

average AOD values in DJF, MAM, JJA, and SON are 0.207, 0.173, 0.135, and 0.21, with trends of
0.0044/10a, -0.0089/10a, -0.0089/10a and 0.0063/10a, respectively.

842 Australia is a region with a low aerosol loading. The multi-year average AOD is 0.133 during 1980-843 2021. The AOD ranges from 0.05 to 0.15 from AERONET during 2000-2021, and dust and biomass 844 burning are important contributors to the aerosol loading (Yang et al., 2021a). There is a downward 845 trend of AOD (-0.0028/10a), which may be related to a decrease in dust and biomass burning (Yoon et al., 2016; Yang et al., 2021a). In addition, a research has shown that the forest area in Australia 846 847 has increased sharply since 2000 (Giglio et al., 2013), surpassing the forest fire area of the past 14 848 years. The seasonal average of AOD in MAM, JJA, SON, and DJF are 0.130, 0.107, 0.132, and 849 0.161. The AOD in JJA is the lowest in all seasons and in all regions. The trends in DJF and SON are increasing, and the trends in MAM and JJA are decreasing. Ground-based observations and 850 satellite retrievals indicate that wildfires, biomass burning and sandstorms lead to high AOD in DJF 851 and SON. The low AOD of MAM and JJA is due to a decrease in the frequency of sandstorms and 852 wildfires and an increase in precipitation (Gras et al., 1999; Yang et al., 2021a; Yang et al., 2021b). 853

854 Asia is also a high aerosol loading area with various sources. In Southeast Asia, the multi-year 855 average AOD is 0.222 during 1980-2021 with a downward trend of AOD (0.0007/10a). It is also a 856 biomass-burning area. The seasonal average AOD ranking is MAM (0.251) > DJF (0.216) > SON857 (0.212) > JJA (0.209). The trend in DJF (-0.0018/10a) is decreasing and the tends in MAM (0.033/10a), JJA (0.0008/10a) and SON (0.0006/10a) are increasing. However, the trends are 858 859 insignificant. Southeast Asia has no clear long-term trend in the estimated AOD or ground-based observations (Streets et al., 2009). In Northeast Asia, the multi-year average AOD is 0.244 during 860 861 1980-2021, with a trend of -0.0009/10a. The trend is increasing (0.0018/10a) during 1980-2014 and 862 decreasing (-0.0213/10a) during 2014-2021. The seasonal AOD values are 0.196 in DJF, 0.260 in MAM, 0.287 in JJA and 0.236 in SON. The high aerosol level is related to dust and aerosol 863 864 transportation in East Asia. The trends in DJF (0.0016/10a), MAM (0.0062/10a) are increasing, and 865 the trends in JJA (-0.0043/10a) and SON (-0.0070/10a) are decreasing. In Eastern China, the multiyear average AOD is 0.241, with an increasing trend (0.0130/10a). The trend is 0.0196/10a from 866 867 1980 to 2014 and -0.0572/10a from 2014 to 2021. The seasonal ranking of AOD from high to low is 868 JJA (0.287), MAM (0.249), SON (0.236) and DJF (0.216). The AOD trends in DJF (0.0133/10a), 869 MAM (0.0179/10a), JJA (0.0107/10a) and SON (0.0105/10a) are all positive. The trend can be divided into three stages: 1980-2005, 2006-2013 and 2014-2021. In the first stage, AOD values are 870 increasing steadily. In the second stage, AOD values maintain a high level. In the third stage, the 871 872 AOD values experience a rapid decline, reaching the level in 1980s by 2021. The increasing trend 873 of AOD before 2006 may be due to the significant increase in industrial activity, and after 2013, the 874 significant decrease is closely related to the implementation of air quality-related laws and regulations, along with adjustments in the energy structure (Hu et al., 2018; Cherian and Quaas, 875 2020). 876

India is a high aerosol loading area. The multi-year average AOD is 0.254, with an increasing trend
(0.0119/10a) from 1980 to 2021. Dust and biomass burning have an influence on AOD. There are
three stages: 1980-1997 (0.0050/10a), 1997-2005 (-0.0393/10a), 2005-2021 (0.0446/10a). The
seasonal average AOD values are 0.238 in DJF, 0.251 in MAM, 0.271 in JJA, and 0.257 in SON.
The largest AOD is in JJA. In winter and autumn, the aerosol level is affected by biomass burning,
and in spring and summer, it is also affected by dust, transported from the Sahara under during the

monsoon period (Remer et al., 2008). The trends in DJF (0.0186/10a), MAM (0.0143/10a), JJA
(0.0012/10a), and SON (0.0129/10a) are positive.

885 The above results have supplemented the existing estimates of long-term AOD variability and trend

- over land. The AOD level at regional scale is significant differences from 1980 to 2021, which is
- significantly related to the aerosol emission source types, transportation and the implementation of
- laws and regulations about pollution control.
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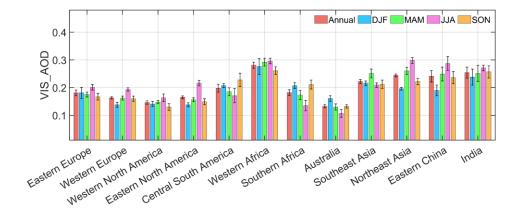


Figure 12: Annual and seasonal mean AOD in 12 regions (Eastern Europe, Western Europe,
Western North America, Eastern North America, Central South America, Western Africa, Southern
Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India) during the period of
1980-2021.

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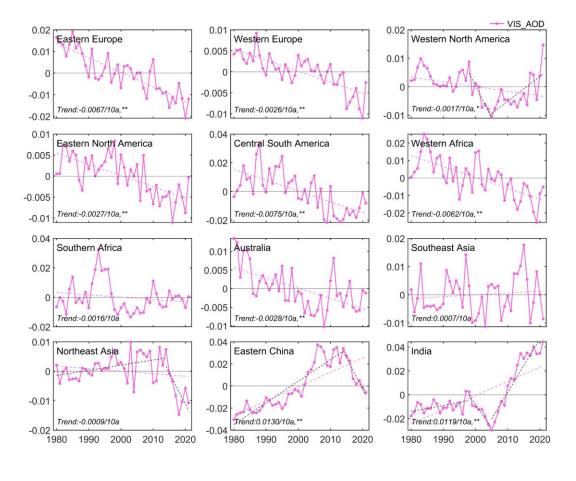


Figure 13: Annual anomaly of VIS_AOD from 1980 to 2021 in 12 regions (Eastern Europe,
Western Europe, Western North America, Eastern North America, Central South America, Western
Africa, Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India). The
dotted line is the trend line.

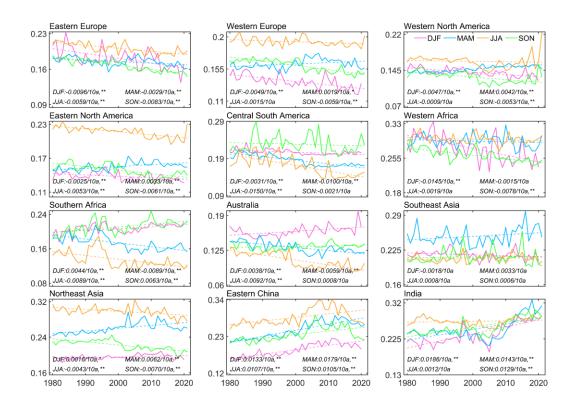


Figure 14: Seasonal mean VIS_AOD from 1980 to 2021 in 12 regions (Eastern Europe, Western
Europe, Western North America, Eastern North America, Central South America, Western Africa,
Southern Africa, Australia, Southeast Asia, Northeast Asia, Eastern China, and India). The dotted
line is the trend line.

909 4 Data availability

We provide the daily visibility-derived AOD data at 5032 stations over global land from 1959-2021,
which is available at National Tibetan Plateau / Third Pole Environment Data Center,
<u>https://doi.org/10.11888/Atmos.tpdc.300822</u> (Hao et al., 2023). Due to a small number and sparse
visibility stations prior to 1980, the global/regional analysis in this study is from 1980 to 2021. The
following is a description to the AOD dataset.

The station-scale AOD files are in 'Station Daily AOD 1959 2021.zip'. The station-scale AOD 915 files can be directly opened by a text program (such as Notepad). The details station information is 916 in the file of '0A0A-Station In Information.txt'. There are eight columns in each text file, separated 917 by commas and the column names are Datetime, TEMP (°C), DEW (°C), RH (%), WS (m/s), SLP 918 (hPa), DRYVIS (km), and VIS AOD (550nm). The first column name is the date. The column name, 919 'VIS AOD (550nm)', is the AOD at 550 nm. The 2-7th column names are temperature (unit: °C), 920 dew temperature (unit: °C), relative humility (unit: %), wind speed (unit: m/s), sea level pressure 921 922 (unit: hPa), and dry visibility (unit: km). The more details are in '0A0B-ReadMe.txt'.

923 **5 Conclusions**

In this study, we employ a machine learning method to derive daily AOD at 550 nm from 1959 to 924 925 2021 at 5032 land stations worldwide, based on visibility, satellite retrieval, and related meteorological variables. In the model, Aqua MODIS AOD (550 nm) is set as the target and 926 visibility and related meteorological variables are set as the predictor. The performance and 927 928 predictive ability of the model are evaluated and validated against AERONET ground-based 929 observations, Terra MODIS AOD and MRRRA-2 AOD. We provide a long-term daily AOD (550 930 nm) dataset at 5032 global land stations from 1959 to 2021. The dataset has complemented the 931 shortcomings of AOD data in terms of time scale and spatial coverage over land. Finally, the 932 variability and trend of AOD are analyzed at global and regional scales in the past 42 years. Several 933 key findings have been given in this study as follows.

1. Modeling evaluation. For all stations, the mean RMSE, MAE, and R of the model are 0.078,
0.044, and 0.75, respectively. The RMSE of 93% stations is less than 0.110, the MAE of 91% is less
than 0.060, and the R of 88% is greater than 0.70.

2. Model validation. For the daily scale, the R, RMSE and MAE between VIS AOD and Aqua 937 AOD are 0.799, 0.079 and 0.044, respectively. The percentage of sample point falling within the EE 938 939 envelopes is 84.12%. The R between VIS AOD and Terra AOD is 0.542, with a RMSE of 0.125 940 and MAE of 0.078. The percentage falling within the EE envelopes is 64.76%. The R between 941 VIS AOD and AERONET AOD is 0.546, with a RMSE of 0.186 and MAE of 0.099. The percentage 942 falling within the EE envelopes is 57.87%. For the monthly and annual scales, RMSE and MAE show a significant decrease between VIS AOD and Aqua, Terra, and AERONET AOD, and R and 943 944 percentages falling within EE show a significant increase. Compared to AERONET AOD and 945 MERRA-2 AOD prior to 2000, the model has consistent predictive ability.

946 3. Error analysis. As the AOD value increases, the average bias increases. When the pollution level 947 is low (AOD < 0.1), the average bias is 0.015, with 83% of data within the EE envelopes. As 948 pollution level increases, the negative average bias becomes significant and the underestimation 949 increases. The elevation of AERONET's site also causes a bias. In low elevation (≤ 0.5 km), there is a negative bias, with a percentage of 60%-64% falling within the EE envelopes. In high elevation 950 951 (0.5-1.2 km), there is a positive bias, with a percentage of 50%-65% falling within the EE envelopes. 952 When the elevation difference is negative (the elevation of the meteorological station is lower than 953 that of the AERONET site), there is a significant positive bias. When the difference is positive, the 954 mean bias approaches 0 or is positive. The influence of distance between the meteorological station and AERONET site on bias is not significant. 955

9564. Global land AOD. The mean AOD from 1980 to 2021 is 0.177 over land, 0.178 in the NH and9570.174 in the SH, with a trend of -0.0029/10a, 0.0030/10a and -0.0021/10a, respectively. The seasonal958AOD rankings are JJA (0.204) > MAM (0.176) > SON (0.164) > DJF (0.161) over global land, and959JJA (0.210) > MAM (0.176) > SON (0.163) > DJF (0.160) in the NH, SON (0.188) > DJF (0.184) >960MAM (0.14) > JJA (0.152) in the SH. The largest decreasing trends are in SON of the NH (-0.0064/10a) and in JJA of the SH (-0.0064/10a). The increasing trends are in MAM of the NH and962in DJF and SON of the SH.

963 5. Regional AOD. The high aerosol loading (AOD > 0.2) regions are West Africa, Southeast and
964 Northeast Asia, Eastern China, and India, with a trend of -0.0062/10a, 0.0007/10a, -0.0009/10a,
965 0.0133/10a, and 0.0119/10a, respectively. However, the trends are decreasing in Eastern China (-

- 966 0.0572/10a) and Northeast Asia (-0.0213/10a) after 2014 and the lager increasing trend is found
- after 2005 in India (0.0446/10a). The moderate aerosol loading (AOD between 0.15 and 0.2) regions
- are Eastern Europe, Western Europe, Eastern North America, Central South America, and South
- 969 Africa, with a trend of -0.0067/10a, -0.0026/10a, -0.0027/10a, -0.0062/10a, and -0.0016/10a,
- 970 respectively. The low aerosol loading (AOD < 0.15) regions are Western North America and
- Australia, with a trend of -0.0017/10a and -0.0028/10a. However, the trends in Southern Africa,
- 972 Southeast Asia and Northeast Asia are not significant.

973 Competing interests

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

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978 visibility data are downloaded from <u>https://mesonet.agron.iastate.edu/ASOS</u>. The Aerosol Robotic

- 979 Network (AERONET) 15-minute AOD data are downloaded from <u>https://aeronet.gsfc.nasa.gov</u>.
- 980 The MODIS AOD data are downloaded from <u>https://ladsweb.modaps.eosdis.nasa.gov</u>.

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