# Global aerosol typing classification using a new hybrid algorithm utilizing Aerosol Robotic Network data

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### 12 Abstract

Aerosols have great uncertainty owing to the complex changes in their composition in 13 different regions. The radiation properties of different aerosol types differ 14 15 considerably and are vital in studying aerosol regional and/or global climate effects. Traditional aerosol-type identification algorithms, generally based on cluster or 16 empirical analysis methods, are often inaccurate and time-consuming. In response, 17 18 our study Hence, we aimed to develop a new aerosol-type classification model using 19 an innovative hybrid algorithm to improve the precision and efficiency of aerosoltype identification. This novel algorithm incorporates an optical database, constructed 20 using the Mie scattering model, and employs a random forest algorithm to classify 21 different aerosol types based on the optical data from the database. The complex 22 23 refractive index was used as a baseline to assess the performance of our hybrid 24 algorithm against the traditional Gaussian kernel density clustering method for aerosol type identification. An optical database was built using Mie scattering and a complex 25 refractive index was used as a baseline to identify different aerosol types by applying 26 a random forest algorithm to train the aerosol optical parameters obtained from the 27 28 Aerosol Robotic Network sites. The hybrid algorithm demonstrated impressive consistency rates of The consistency rates of the new model with the traditional 29 Gaussian were 90%, 85%, 84%, 84%, and 100% for dust, mixed-coarse, mixed-fine, 30

urban/industrial, and biomass burning aerosols, respectively. <u>Moreover, it achieved</u> <u>remarkable precision, with F-score and accuracy scores of The corresponding</u> precision of the new hybrid algorithm (F-score and accuracy scores) was 95%, 89%, 91%, and 89%. Lastly, a global map of aerosol types was generated using the new model to characterize aerosol types across the five continents. This study utilizing a novel approach for the classification of aerosol will help improve the accuracy of aerosol inversion and determine the sources of aerosol pollution.

Keywords: Aerosol typing classification, Hybrid algorithm, Complex refractive index,
AERONET

## 40 **1. Introduction**

Atmospheric aerosols are tiny solid or liquid particles suspended in the 41 atmosphere. Aerosols indirectly affect the energy budget and water cycle of the earth's 42 gas system by absorbing and scattering solar radiation or by changing the optical 43 properties and life cycle of the cloud as condensation nuclei of cloud droplets 44 (Redemann et al. 2000; Ramanathan et al. 2001). Additionally, desert dust, biomass 45 smog, and anthropogenic emissions of air pollutants can affect visibility, air quality, 46 and human health (Hess et al., 1998; Tong et al., 2017; Siomos et al., 2020). 47 48 Evaluating the impact of aerosols on radiative transfer is complex, primarily because of the uncertainty of radiative forcing caused by the high spatiotemporal dynamic 49 variation of aerosol optical and physical characteristics in different regions 50 (Kaskaoutis et al., 2011;Che et al., 2018; Elham et al., 2023).The aerosol type 51 52 embodies the long-term average physicochemical properties of aerosols in a certain area (Kiehl & Briegleb, 1993;Lu et al., 2023). Therefore, accurate identification of 53 aerosol types can drive the study of the climatic effects of aerosols, tracking and 54 control of environmental pollution sources, and precision of radiation transmission 55 models. 56

57 Aerosol types are defined based on the radiation properties of different types of 58 aerosol particles owing to the large variation in their optical, physical, and chemical

properties. Currently, aerosol types are classified by two ways using two different 59 clustering techniques (Kumar et al., 2018). First, based on different sources and 60 61 properties at different observation points worldwide, aerosols are classified as follows: dust aerosols from deserts, biomass combustion aerosols from forests or grasslands, 62 and urban/industrial (U/I) aerosols from fuel combustion in densely populated urban 63 areas (Dubovik et al., 2002; Pawar et al., 2015; Yousefi et al., 2020). Second, based on 64 the size of the radiation absorption rate, aerosols into four categories: carbonaceous 65 (fine-absorbing mode), soil dust (coarse absorption mode), sulfates (nonabsorbing 66 fine-grained mode), and sea salt aerosols (nonabsorbing coarse-grained mode) (Kim 67 et al., 2007;Levy et al., 2007). The second one is a type of subcategorize 68 anthropogenic aerosol. The first one is commonly used for aerosol retrieval. Therefore, 69 the first aerosol type classification is more common in research. The optical properties 70 of aerosols observed at ground stations are commonly used to construct a two-71 dimensional identification space to obtain the aerosol types by clustering techniques. 72 Many combinations of optical properties and parameters are available; They include 73 74 EAE440-870nm (extinction angstrom exponent) vs. SSA440nm (single-scattering albedo), 75 AAE440-870nm (absorption angstrom exponent) vs. EAE440-870nm, AAE440-870nm vs. FMF<sub>550nm</sub> (fine mode fraction), and SSA<sub>440nm</sub> vs. EAE<sub>440-870nm</sub> (Lee et al., 2010;Shin et 76 al., 2019; Choi, et al., 2021). Studies have highlighted the importance of selecting 77 appropriate aerosol properties for accurate aerosol type identification (Giles et al., 78 2012; Che et al., 2018). 79

Among the aerosol-type classification methodologies developed, those using 80 81 threshold and empirical analyses have the greatest potential for large-area and fixed-82 period applications (Eck et al., 1999; Omar et al., 2005; Yang et al., 2009). 83 Traditionally, the aerosol-type classification algorithm mainly distinguishes different aerosol types based on their optical properties and determines the threshold of their 84 optical properties based on clustering. However, the composition of aerosols changes 85 86 rapidly with time and location, owing to the combined influence of natural conditions and human activities (for example, tornadoes and various anthropogenic activities) 87 (Sheridan et al., 2001). Unfortunately, determining aerosol types accurately and 88

rapidly is a challenge when using traditional methods (Bahadur et al., 2012;Shin et al.,
2019;Lin et al., 2021). Nevertheless, with advancements in data science, artificial
intelligence techniques have aided the accurate and rapid recognition of different
aerosol types.

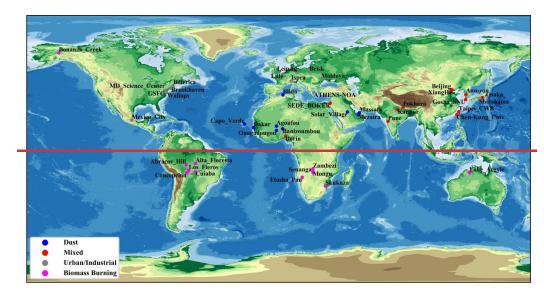
Artificial intelligence algorithms can receive multiple aerosol characteristic 93 parameters as input, thus preventing the sole reliance of aerosol classification on a 94 limited number of features (Li et al., 2022; Wang et al., 2023). For example, Boselli 95 96 (2012) performed a k-means clustering analysis of single scattering albedo (SSA), aerosol optical depth (AOD), electrical asymmetry effect (EAE), and asymmetry 97 parameter (g) datasets for the central Mediterranean Sea for the classification of 98 aerosol into four: dusty, continental, oceanic, or mixed aerosols. Nicolae (2018) 99 developed a neural network algorithm to estimate the aerosol typing of Lidar data and 100 Hamill (2016) introduced the Mahalanobis Distance for aerosol classification to 101 determine a specific aerosol type for each reference cluster. Li (2022) generated 102 spatial contiguous aerosol type map in China with an empirical aerosol type retrieval 103 104 algorithm. Overall, limited information on the optical properties of aerosols can reasonably determine the type of aerosol (Hamill et al., 2016). However, some 105 challenges remain in identifying aerosol types through machine learning. First, the 106 amount of valid ground aerosol property data that can be used for training is less due 107 to cloud removal and quality control. Second, the accuracy of machine learning 108 depends on the labeled aerosol typing dataset, and finding a suitable classification 109 method to classify the dataset is challenging. Third, evaluating the accuracy of the 110 final trained model is also tedious (Zhang & Li, 2019;Siomos et al., 2020; Choi, et al., 111 112 2021a,b)

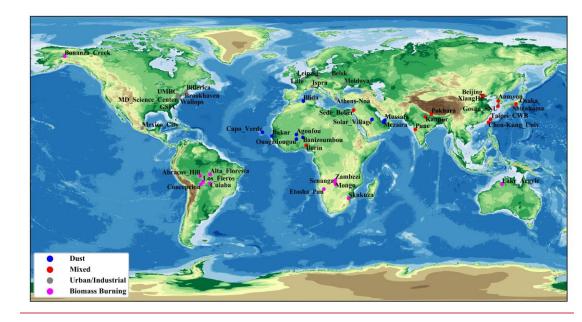
The traditional aerosol type identification methods are easily limited by time and space, and most of them only classify aerosol types using two optical property parameters, limiting the complete characterization of aerosols. Considering these limitations, we aimed to (1) develop a new algorithm that can accurately and quickly identify aerosol types to overcome existing problems such as low accuracy, insufficient data, and difficulty in setting labels; (2) investigate the characteristics of the regional spatial distribution of global aerosol types obtained using the new machine learning algorithms, considering the large regional differences in aerosol types. To achieve this, we propose a new aerosol-type classification algorithm based on a Gaussian cluster and random forest algorithm to generate an aerosol-typing map over several representative regions of the world.

#### 124 **2. Study area and data**

125 Figure 1 illustrates the research area and the distribution of the Aerosol Robotic 126 Network (AERONET) sites, strategically encompassing major global regions to validate the universality of the research algorithm. The study utilized 47 marked 127 128 aerosol sites across five continents, leveraging them to train and validate the machine 129 learning approach based on a comprehensive literature review. Figure 1 shows the study area and the Aerosol Robotic Network (AERONET) site distribution, which 130 covers major regions of the world, to ensure the generalizability of the research 131 algorithm. We used 47 aerosol sites as marked on the map that were distributed over 132 five continents to train and verify machine learning by literature review. The 47 sites 133 represent different aerosol-type properties of different aerosol source regions, 134 135 including dust, mixed (mixed coarse and mixed fine aerosols), U/I, and biomass burning (BB) aerosols (Table 1 and Figure 1). Marine aerosols were not considered 136 because their low optical thickness values (generally <0.4) can result in a less valid 137 data scale that would not meet the study requirements. Here, the aerosol source region 138 refers to the area affected by one dominant emission source, where the aerosol types 139 are fixed and not easily confused (Giles et al., 2012;Hamill et al., 2016). Table 2 140 presents the optical properties and microphysical characteristic parameters of aerosols 141 at four bands of AERONET (440, 675, 870, and 1020 nm). These parameters were 142 used to construct a database of SSA, AOD, and asymmetry parameters. Further, 143 typical sites dominated by different aerosol types worldwide were selected for 144 compositional analysis using the new model. The selected sites are distributed across 145 different regions of the world and represent a specific aerosol-dominated type and 146 147 aerosol source region.

148 For dust aerosols, five AERONET sites, namely Banizoumbou, Capo\_Verde, 149 Dakar, and Ouagadougou in Africa and Solar\_Solar\_Village in West Asia, influenced 150 by the Saharan Desert, were considered. The Dakar and Cape-Capo\_Verde sites are located at the tip of the CapeCapo\_Verde Peninsula—the westernmost part of Africa, 151 bordering the Atlantic Ocean. Although these two sites are located in the ocean, they 152 are dominated by dust aerosols influenced by aerosol plumes in the Saharan Desert. 153 Moreover, the Banizoumbou and Ouagadougou sites are in the middle of Africa. Here, 154 the northeasterly winds prevail in winter, and northwesterly winds prevail in summer, 155 which can bring dust aerosols from the Saharan Desert. For mixed aerosols, the 156 AERONET sites Ilorin, Kanpur, Sede\_Boker, and XiangHe were selected. For U/I 157 aerosols, the AERONET sites GSFC, Ispra, Mexico\_City, and Moldova were selected. 158 159 Four AERONET sites, namely, Alta\_Floresta, Abracos\_Hill, Lake\_Argyle, and Mongu, were selected as BB aerosol-dominant sites. 160





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**Figure 1**. Study area and 47 AERONET sites selected by literature review.

## **Table 1**. 47 AERONET sites selected by literature review.

Aerosol Type	Sites for Training	Sites for Testing	
Dust	Agoufou,Capo_Verde,Dakar,-Mezaira,	Banizoumbou,	
	Mussafa,-Ouagadougou	Solar_Village,	
		Blida	
Mixed	Anmyon, Beijing, Chen-Kung_Univ, Ilorin, Kanpur,	Osaka, XiangHe,	
	Sede_Boker, Gosan_SUN, Pune, Taipei_CWB	Pokhara	
Urban/Industry	Brookhaven, Billerica, Belsk, GSFC, Ispra, UMBC, Lille,	Athens_Noa,Shirahama,	
	Mexcio_City,Moldova,MD_Science_Center,Wallops	Leipzig	
Biomass	Abracos_Hill,Alta_Floresta,Cuiaba,Concepcion	Bonanza_Creak,	
Burning	Los_Fieros,Mongu,Senanga,-Skukuza,Zambezi	Etosha_Pan, Lake_Argyle	

# **Table 2**. The optical and microphysical properties for aerosol type identification.

	Parameters	Variables (band waves)
	Ångström Exponent (AE)	EAE (440-870) <sup>1</sup>
	Aerosol Optical Depth (AOD)	AOD (440,675,870,1020) <sup>1</sup>
Optical	Single Scattering Albedo (SSA)	SSA (440,675,870,1020) <sup>1</sup>
Properties	Asymmetry Parameter	g (440,675,870,1020) <sup>1</sup>
	Imaginary Part of the Complex Refractive Index	REFI (440,675,870,1020) <sup>1</sup>
	Real Part of the Complex Refractive Index	REFR(440,675,870,1020) <sup>3</sup>
Microphysical	Effective Radius	EffRad-F <sup>2</sup> , EffRad-C <sup>2</sup>
Properties	Standard Deviation of Effective Radius	StaDev-F <sup>2</sup> , StaDev-C <sup>2</sup>
	Size Distribution	Vol-Con (0.05-15µm)

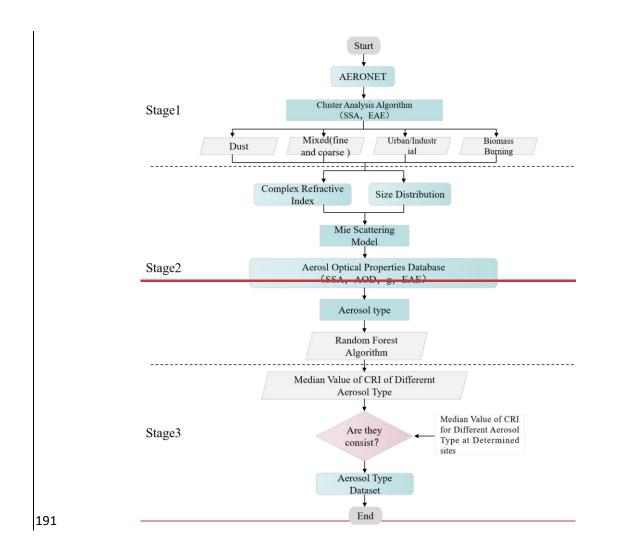
166 Note: <sup>1</sup> refers to wavelength in nm; <sup>2</sup> refers to different modes; EAE is Extinction Ångström Exponent; REFI is Imaginary Part of the

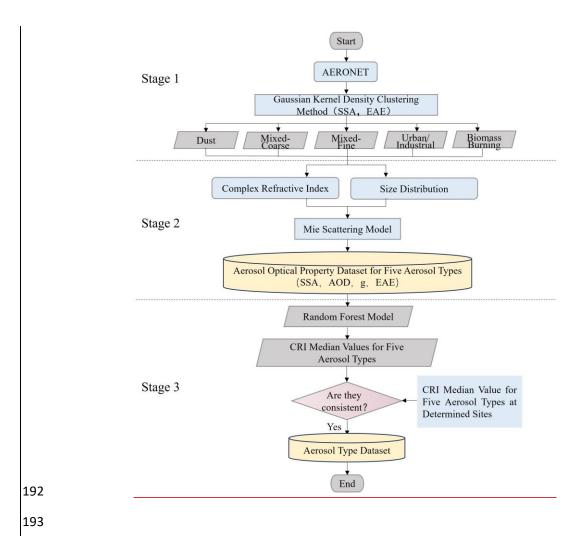
167 Complex Refractive Index; REFR is Real Part of the Complex Refractive Index; F refers to fine mode; C refers to coarse mode; EffRad is

168 Effective Radius; StaDev is standard deviation; Vol-Con is Volume concentration

## 169 **3. Methods**

A new aerosol classification typing hybrid approach that provides insight into 170 spatiotemporal variations in aerosol pollution and climate impacts on a global scale is 171 proposed in this study. In this approach, an aerosol optical properties database using 172 the Mie scattering model was built for calculating rapidly unique aerosol-type features. 173 174 Additionally, the approach introduced, for the first time, the median value of the 175 complex refractive index (CRI) as the criterion for identifying the aerosol type. CRI, a key microphysical characteristic of aerosols, plays a significant role in determining 176 177 their intrinsic optical properties, such as their ability to scatter and absorb light (Raut and Chazette, 2008). The CRI is also vital for determining aerosols' chemical and 178 physical compositions (Dubovik and King, 2000) and the CRI value is known for pure 179 180 aerosol components (Nandan et al., 2021). Unlike the mean, the median CRI value is employed in this research for it represents the central tendency of data, especially 181 beneficial in skewed distributions or when outliers are present. This is particularly 182 183 useful when an average value of a specific aerosol-type might be influenced by the presence of other aerosol types. Moreover, Further, we have selected the aerosol 184 classification based on the source (as described in Section 1), according to the 185 parameters applied in this study and the requirements for AOD retrieval. Figure 2 186 187 shows the working flowchart of the new hybrid aerosol-type identification approach, including three stages: aerosol typing preliminary classification, aerosol optical 188 database generation, and global aerosol typing identification and validation. The 189 details of these three stages are as follows. 190





**Figure 2**. Flow chart of the new hybrid algorithm in aerosol type identification.

## **3.1** Aerosol typing preliminary classification (Stage 1)

Stage 1 aimed to solve the problem of obtaining a feature parameter dataset for 196 197 the baseline aerosol type. In previous studies, the Gaussian kernel density clustering algorithm showed great potential for distinguishing the optical properties of different 198 199 aerosol types and determining their corresponding thresholds rapidly (Kalapureddy et 200 al. 2009; Pathak et al. 2012). The high concentration value in each cluster generally 201 represents the dominant pattern of a specific aerosol type, particularly the data within the window, taking the cluster centroid as the center and a specific distance as the 202 radius. Preliminary aerosol-type datasets can be generated by digging deep into the 203 distribution information of the effective radius, variance, and refractive index of the 204 data within the window. The spectral absorbability and particle size of aerosols guide 205 the identification of dust, carbonaceous, or hygroscopic aerosols; SSA indicates the 206

absorption of aerosol particles; and EAE describes aerosol particle size (Giles et al.,
208 2012). Consequently, in this study, SSA<sub>440nm</sub> and EAE<sub>440-870nm</sub> of 47 AERONET sites
and the <u>Gaussian kernel density clusteringGaussian kernel density</u> method was used
to estimate the relative densities and determine the primary patterns of the dominant
aerosol types; here, the aerosol type was classified as a dust aerosol. Eqs. (1) and (2)
represent the kernel density and <u>Gaussian kernel density clusteringGaussian kernel</u>
density methods (Rosenblatt, 1956).

214 
$$f_{X(v)} = \frac{1}{L} \sum_{i=1}^{L} k_{\sigma} (\frac{\vec{x} - \vec{x}_i}{\sigma}) , \qquad (1)$$

where  $f_{X(v)}$  denotes the kernel density and  $k_{\sigma}$  indicates the kernel function. x<sub>1</sub>, x<sub>2</sub>... x<sub>L</sub> are the sample points of independent identical distribution. Mathematically, kernel functions are symmetric, normalized, and sample-centric when used for density estimation; this is best described by the Gaussian kernel equation given by Eq. (2).

219 
$$k_{\sigma} = \frac{1}{\sqrt{2\pi\sigma}} \exp(\frac{-|\vec{x} - \vec{x}_i|^2}{2\sigma^2}), \qquad (2)$$

220

where  $\sigma$  is the kernel size used as a smoothing factor (Moraes et al., 2021).

The mixed aerosols comprised fine- and coarse-mode aerosols, indicated by 221 EAE > 0.8 and  $EAE \le 0.8$ , respectively. Figure 3 shows the clustering distribution of 222 223 EAE and SSA using the Gaussian kernel density clustering Gaussian kernel density method for different aerosol types at the 47 AERONET sites. For the dust aerosol 224 cluster, the density core area EAE was 0.1-0.3, and SSA was 0.89-0.94, implying that 225 it contained many coarse aerosol particles with moderate absorptivity. Furthermore, 226 227 the mixed aerosols had two distinct centers: one for the coarse-mode aerosols with a median EAE value of 0.4, indicating that the cluster contained massive high-228 absorption aerosols, and the other for fine-mode aerosols with a median EAE value of 229 1.3. Low-absorption aerosols were dominant in the cluster, similar to U/I aerosols. 230 231 Additionally, the density core region EAE of U/I aerosol was 1.5-1.8, and SSA was 0.94–0.97, implying the dominance of fine and low-absorption aerosols. Conversely, 232 BB aerosols had two indistinct centers. This is because, during biomass combustion, 233

gas and particulate matter emissions are limited by the combustion conditions, divided 234 into combustion and simmering. Combustion produces black smoke, and simmering 235 produces white smoke. Combustion, such as burning flames (grass) with high black 236 carbon content, has a strong absorption capacity, resulting in a low SSA. Simmering, 237 such as burning wood (i.e., trees), tends to be smoldering, lasts longer, has a weaker 238 239 absorption capacity, and has a higher SSA value. Therefore, despite possessing different absorption characteristics, BB aerosols are defined as one aerosol type with 240 241 an unseparated center of combustion and simmering.

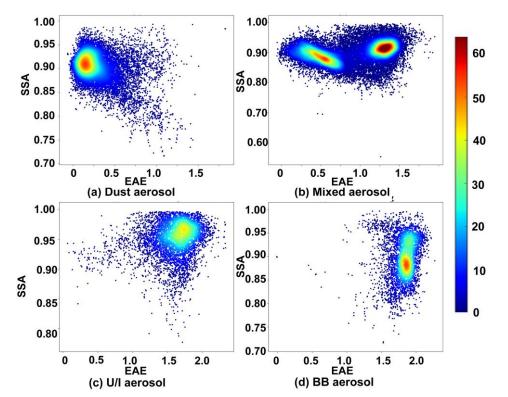


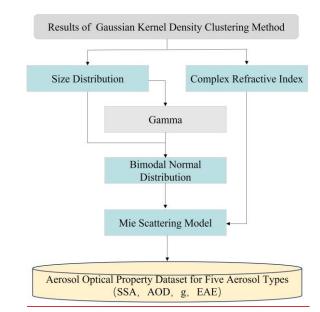
Figure 3. The clustering distribution of EAE and SSA using the <u>Gaussian kernel density clustering</u>
 method for different aerosol types.

# 245 **3.2 Aerosol optical database generation (Stage 2)**

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In stage 2, the aerosol optical parameter database was built using the aerosol size distribution parameters, CRI, and Mie scattering model. The main reasons for constructing an aerosol optical parameter database instead of using the AERONET data directly are as follows: 1) many data are missed in AERONET, particularly those for sites dominated by biomass combustion, which does not meet the requirements of machine learning methods or traditional aerosol type identification algorithms; 2)

Calculating the optical properties of aerosols based on a fixed refractive index can 252 accurately determine aerosol types. Therefore, once the aerosol spectral distribution 253 254 parameters, such as effective radius, variance, and refractive index of the five aerosol types, are determined in stage 1, the aerosol optical parameter database can be 255 constructed using the Mie scattering model in stage 2, assuming that aerosols are 256 257 spherical particles. The Mie scattering model is a simple, practical, and ideal spherical particle model commonly used in radiation transport models (Michael et al., 1994). 258 259 Figure 4 shows the details involved in the building aerosol optical database. The aerosol optical database has four major parameters (AOD, EAE, SSA, and g) at four 260 wavelengths (440, 675, 870, and 1020 nm, respectively). 261



262

**Figure 4**. The diagram of building aerosol optical property database.

As shown in Figure 4, size distribution is a major parameter in building aerosol optical databases. Table 3 presents the aerosol size distribution parameters, including the effective radius and standard deviation range for the five aerosol types in the coarse and fine modes, which were calculated using the data in the window determined by the Gaussian kernel density algorithm. These aerosol size distribution parameters were used to build the aerosol optical database for the Mie scattering model.



Aerosol type	<b>REFF-fine</b>	<b>REFF-coarse</b>	Std-fine	Std-coarse
Dust	0.05-0.42	1.3-2.65	0.5-0.8	0.4-0.7
Mixed-coarse	0.05-0.25	1.25-3.5	0.4-0.8	0.4-0.7
Mixed-fine	0.05-0.27	1.2-4.5	0.3-0.6	0.5-0.8
U/I	0.05-0.26	1.45-3.5	0.3-0.6	0.5-0.8
BB	0.05-0.17	1.35-4.5	0.3-0.5	0.5-0.8

272 Table 3 presents the aerosol size distribution parameters, including the effective 273 radius and standard deviation range for the five aerosol types in coarse and fine modes, which were derived from the data window set by the Gaussian kernel density 274 275 clustering algorithm. These aerosol size distribution parameters and the median CRI 276 value were utilized to construct the optical database for the Mie scattering model. 277 Many studies proven it is a reliable model with the advantage of lower computing load and high calculation accuracy (Zhao et al., 2008; Fu et al., 2009; Quirantes et al, 278 279 2019; Nandan et al., 2021).

The Mie scattering model has various size distribution functions, including lognormal, power-law, and bimodal log-normal distributions, which describe the aerosol type. According to the particle radii provided by AERONET, the size distributions of different aerosol types can be divided into coarse and fine modes. The bimodal lognormal function [Eq. (3)] is reportedly the most suitable size distribution function for modeling aerosol particle size distribution (Remer et al., 2009):

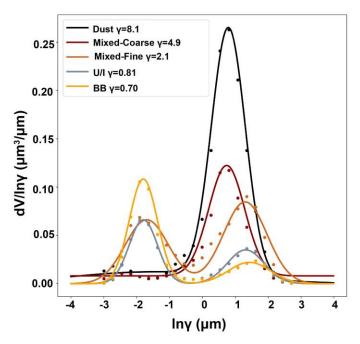
286 
$$n(r) = cons \tan t \times r^{-4} \{ \exp(-\frac{(\ln r - \ln r_{g_1})^2}{2\ln^2 \sigma_{g_1}}) + \gamma \exp(-\frac{(\ln r - \ln r_{g_2})^2}{2\ln^2 \sigma_{g_2}}) \} , \quad (3)$$

where n(r) is the number of particles at different radii; constant is obtained by fitting; While  $r_{g1}$  and  $r_{g2}$  denote the radii,  $\sigma_{g1}$  and  $\sigma_{g2}$  denote the variances of the aerosol in the coarse and fine modes, respectively; and  $\gamma$  is determined by the volume distribution. In the bimodal normal distribution model,  $\gamma$  is the ratio of coarse to fine modes, which can be fitted by the volume distribution from AERONET; notably, volume distribution is the average of the standard aerosols obtained after clustering at the training sites.

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Figure 5 shows the volume distributions of the different aerosol types. The

aerosol volume distribution of dust aerosol-dominant sites focuses on the large radius; 294 the peak value of  $\gamma$  was 8.1, and the radius of dust aerosols was 1.5–2.0  $\mu$ m. 295 296 Additionally, the mixed-coarse aerosol with the radius in the range of  $0.04-0.2 \ \mu m$ and 4.9 as the maximum value of  $\gamma$ . The mixed-fine aerosol had two obvious peaks: 297 one with a large radius, namely the coarse mode, with a radius of  $2.2-3 \mu m$  and 2.1 a s298 299 the peak point of  $\gamma$ ; a second with a small radius of 0.1–0.22 µm and 0.14 as the peak point of  $\gamma$ . Moreover, the volume distributions of U/I and BB aerosols were similar. 300 301 Both had a relatively low range of  $\gamma$  values at large radii and relatively high values at small radii, with peak values of 0.81 and 0.7 for U/I and BB aerosols, respectively. 302



303

**Figure 5.** Volume distribution of the five aerosol types.

305 The CRI of aerosols is another key parameter among aerosol optical properties; it 306 determines inherent optical properties of aerosols, such as scattering and absorption 307 (Raut and Chazette, 2008). The CRI is vital for determining aerosols' chemical and 308 physical compositions (Dubovik and King, 2000). Aerosols in the real atmosphere are 309 usually mixed with different types of particles, which a single refractive index cannot identify; however, the CRI represents the entire aerosol model in the atmosphere 310 (Redemann et al., 2000). Ideally, the CRI and aerosol components can be mutually 311 determined (Wu et al., 2021). Table 4 depicts the CRI standard values for the five 312

313 aerosol types obtained by calculating the median value of the CRI of the dominant 314 aerosol type after Gaussian density clustering. These values were used as a baseline 315 for identifying the aerosol types in subsequent studies. As presented in Table 4, the minimum imaginary index part is represented by the dust aerosol with CRI of 316 0.003396, 0.000731, 0.000639, and 0.000597 at 440, 675, 870, and 1020 nm, 317 318 respectively, owing to the weakest absorption of dust aerosols. Moreover, the imaginary index part of the mixed-fine aerosols (0.01) was close to that of the BB 319 320 aerosols (0.02) because of their similar absorption properties.

321 Table 4. Real and imaginary index of CRI for the five aerosol types (Bands:440/675/870/1020
322 nm).

Aerosol Type	Imaginary Index	Real Index
Dust	0.003396/0.000731/0.000639/0.000597	1.4584/1.4681/1.4513/1.4376
Mixed-coarse	0.005766/0.002921/0.002383/0.002043	1.4291/1.4787/1.4745/1.4695
Mixed-fine	0.01075/0.008444/0.009147/0.008955	1.5001/1.5044/1.5056/1.4977
U/I	0.004315/0.004331/0.004419/0.004432	1.4372/1.4280/1.4264/1.4214
BB	0.01828/0.017862/0.018125/0.017858	1.5051/1.5190/1.5228/1.5185

323	The CRI is an inherent optical property of aerosols. Aerosols in the real
525	The CRI is an inherent optical property of actosols. Actosols in the real
324	atmosphere are usually mixed with different types of particles, which a single
325	refractive index cannot identify; however, the CRI represents the entire aerosol model
326	in the atmosphere (Redemann et al., 2000). Ideally, the CRI and aerosol components
327	can be mutually determined (Wu et al., 2021). The CRI can effectively characterize
328	the main properties of the aerosols and accurately quantify the difference between
329	aerosol-type identification algorithms. Table 4 depicts the CRI standard values for the
330	five aerosol types obtained by calculating the median value of the CRI of the
331	dominant aerosol type after Gaussian kernel density clustering. These values were
332	used as a baseline for identifying the aerosol types in subsequent studies. As presented
333	in Table 4, the minimum imaginary index part is represented by the dust aerosol with
334	CRI of 0.003396, 0.000731, 0.000639, and 0.000597 at 440, 675, 870, and 1020 nm,
335	respectively, owing to the weakest absorption of dust aerosols. Moreover, the
336	imaginary index part of the mixed-fine aerosols (0.01) was close to that of the BB

aerosols (0.02) because of their similar absorption properties.

Lastly, by fixing the CRI, changing the size distribution, and using the Mie 338 scattering model, we generated the aerosol optical property database for five aerosols, 339 including the data for AOD, EAE, SSA, and g. In the aerosol optical property 340 database, AOD is the value obtained after eliminating the influence of the aerosol 341 concentration. The AOD was obtained from the extinction cross section (Cext) 342 calculated using the Mie scattering model in Eqs. (3) and (4), where  $\beta_{ext}$  is the 343 extinction coefficient, n(r) is the aerosol spectral distribution, and N(z) is the variation 344 of aerosol concentration with height. Notably, the effect of aerosol concentration 345 needs to be removed from the AOD when referring to aerosol optical properties. The 346 AOD was normalized by dividing the aerosol optical thickness at the four 347 wavelengths by the optical thickness at 440 nm. The other parameters (EAE, SSA, 348 and g) were calculated using Eqs. (6) - (8). 349

350 
$$\beta_{e/s} = \int_{\gamma_{min}}^{\gamma_{max}} C_{ext/sca} n(r) dr , \qquad (4)$$

351 
$$\tau_{e/s} = \int_0^{Z_{top}} \beta_{ext/sca} N(z) dz, \qquad (5)$$

352 
$$EAE_{440-870nm} = -\frac{\ln(\tau_{440nm}) - \ln(\tau_{870nm})}{\ln(440) - \ln(870)}$$
(6)

$$SSA = \frac{\tau_s}{\tau_e}, \tag{7}$$

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I

355 
$$g = \langle \cos\Theta \rangle = \frac{1}{2} \int_{-1}^{1} p(\cos\Theta) \cos\Theta d \cos\Theta , \qquad (8)$$

and

where  $\tau_{440}$  and  $\tau_{870}$  are the extinction optical depths of the aerosol at 440 and 870 nm, respectively, EAE<sub>440-870</sub> nm is the extinction Ångström index from the 440 to 870 nm band, and  $\Theta$  denotes the scattering angle.

359 The amount of data for the five aerosol types calculated using the Mie scattering
 360 model is presented in Table 5. The least amount of data was observed for the mixed-

361 fine aerosol owing to its small distribution range of effective variance. The largest data was observed for dust and mixed aerosols owing to their widely distributed 362 363 effective radii. A total of 326400 datasets were present in the aerosol optical database, which meets the requirements for random forest algorithm.he amount of data for the 364 five aerosol types calculated using the Mie scattering model is presented in Table 5. 365 The least amount of data was observed for the mixed-fine aerosol owing to its small 366 distribution range of effective variance. The largest data was observed for dust and 367 mixed acrosols owing to their widely distributed effective radii. A total of 326400 368 datasets were present in the aerosol optical database, which meets the requirements 369 370 for random forest algorithm.

#### 371 **Table 5**. The data size of optical database simulated by Mie scattering model.

Total	<b>Dust</b>	Mixed-coarse	Mixed-fine	<del>U/I</del>	BB
<del>326400</del>	<del>88200</del>	<del>96000</del>	<del>42000</del>	<del>51840</del>	<del>48360</del>

## **372 3.3** Global aerosol type identification and validation (Stage 3)

In stage 3, the random forest model was introduced to the aerosol-type 373 374 identification algorithm. The random forest model is an integrated model based on 375 classification and regression trees, in which multiple trees are aggregated using majority voting and averaging for classification and regression (Breiman, 2001). The 376 model has a high prediction accuracy, excellent tolerance for abnormal values and 377 noise, and a hard overfit. In a comparison by Fernandez (2014), the random forest 378 algorithm ranked as the top performer among 179 classification algorithms. In 379 380 addition, the evaluation matrix was brought into this study, and it further quantitatively assesses the performance of the Gaussian density clustering algorithm 381 382 and the new hybrid algorithm. The metric indexes include accuracy, recall, precision, and F-scores (Reddy et al., 2022). Here, the indexes are adjusted to micro-precision, 383 micro-recall, micro-F1-score, and accuracy to solve the multi-classification problem. 384 Micro refers to the weighted average of the five aerosol types rather than the 385 arithmetic mean, due to the large difference in sample size among the five aerosol 386 types, the arithmetic mean is highly susceptible to the influence of very large or very 387

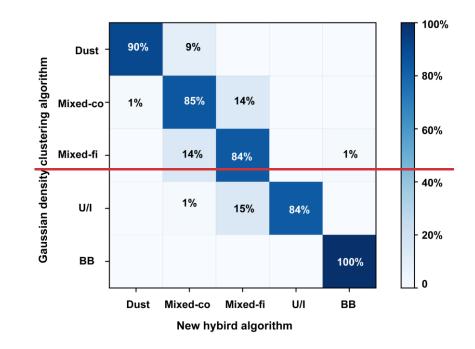
388 <u>few sample size aerosol types.</u>The random forest model is an integrated model based 389 on classification and regression trees, in which multiple trees are aggregated using 390 majority voting and averaging for classification and regression (Breiman, 2001). The 391 model has a high prediction accuracy, excellent tolerance for abnormal values and 392 noise, and a hard overfit. In a comparison by Fernandez (2014), the random forest 393 algorithm performed the best among 179 classification algorithms. Moreover

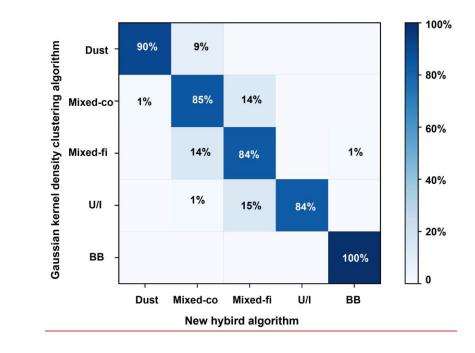
The input parameters for random forest model training, including SSA<sub>440nm</sub>, 394 395 SSA675nm, SSA870nm, SSA1020nm, g440nm, g675nm, g870nm, g1020nm, normalized AOD440nm, AOD<sub>675nm</sub>, AOD<sub>870nm</sub>, AOD<sub>1020nm</sub>, and EAE<sub>440-870nm</sub>, were selected from the aerosol 396 optical property database, and the expected output values were the specific aerosol 397 types. The random forest model was optimized and the parameters were determined 398 using the grid-searching method. The parameters, including n\_estimators (classifier), 399 max\_features (maximum feature value), and min\_samples\_leaf (minimum number of 400 samples for nodes), were set as 160, 10, 12, and 12, respectively. Then, based on the 401 trained and optimized model, aerosol typing of any AERONET site in different 402 403 regions of the world can be identified quickly. -Generating the aerosol type distribution map on a global scale is vital for regional and global climate studies and 404 ground remote sensing. 405

406 4 Results

#### 407 **4.1 Algorithm comparison**

To demonstrate the effectiveness of the new hybrid algorithm, its performance 408 409 was compared with that of Gaussian kernel density clustering Gaussian density elustering algorithm. Figure 6 shows the confusion matrix between the new hybrid 410 and Gaussian kernel density clustering algorithms in identifying aerosol types. The 411 412 results of the <u>new</u> hybrid algorithm showed 90% consistency with that from the 413 Gaussian kernel density clustering algorithm, in delineating dusty aerosols, indicating 414 that its efficiency in identifying dust. For mixed-coarse aerosols, the consistency 415 reached 85%, with 14% identified as mixed-fine aerosols, 1% as dust by the new 416 hybrid algorithm, and 15% as mixed-coarse aerosols by the Gaussian kernel density 417 clustering algorithm. Similarly, for mixed-fine aerosols, both algorithms showed 84% 418 consistency, with 14% identified as a mixed-coarse aerosol by thethe new -hybrid algorithm and as a mixed-fine aerosol by the Gaussian kernel density cluster 419 420 algorithm. Furthermore, both algorithms identified 84% of U/I aerosols correctly, with the remaining 16% identified as mixed aerosols (fine and coarse). Lastly, the 421 422 classification of BB aerosols using these two methods was the same. Overall, the 423 Gaussian kernel density clustering and <u>new hybrid</u> algorithms were consistent in dust, mixed-coarse, U/I, and BB aerosol identification. 424





427 Figure 6. The confusion matrix between Gaussian <u>kernel</u> density clustering and <u>new</u> hybrid
428 algorithm.

Table 5 shows the metric index value of the random forest algorithm in the new
hybrid algorithm. The micro-precision, micro-recall, micro-F1 score, and accuracy are
0.95, 0.89, 0.91, and 0.89, respectively. These metrics are derived from the core
values of the window, as determined by the Gaussian density clustering algorithm.
Consequently, the strong performance of these indicators further confirms the efficacy
and reliability of the newly developed hybrid algorithm.

Table 5. Matrix evaluation between new hybrid classification algorithm and Gaussian kernel
 density clustering algorithm

		Micro-Precision	Micro-Recall	Micro-F1-Score	Accuracy
	New Hybrid algorithm	<u>0.95</u>	<u>0.89</u>	<u>0.91</u>	<u>0.89</u>
437	As described i	in the Methods	section, a specif	ic aerosol type the	oretically has a
438	fixed CRI owing	to its constant	composition. Tl	ne CRI characteriz	the mixture
439	composition of ae	erosol particles	<u>and is a key p</u>	arameter controllir	ng the inherent
440	scattering and abso	orption characte	ristics of aeroso	l particles. To furt	her analyze the
441	accuracy of the ne	ew algorithm, th	e aerosol CRI v	<u>vas applied as a k</u>	ey criterion for
442	aerosol identification	on. The CRI has	s two parts: imag	inary and real. The	<u>e imaginary part</u>
443	indicates radiation	absorption by	aerosols, with	<u>a small value sig</u> r	nifying a small

<u>absorption. Because the radiation of aerosols is more dependent on the imaginary than</u>
<u>the real part, the imaginary part is essential for inferring the optical properties and</u>
<u>aerosol types. Hence, we compared the real and imaginary parts of the CRI calculated</u>
<u>using the new hybrid and Gaussian kernel density clustering algorithms.</u>

As described in the Methods section, a specific aerosol type theoretically has a 448 fixed CRI owing to its constant composition. The CRI characterizes the mixture 449 composition of aerosol particles and is a key parameter controlling the inherent 450 451 scattering and absorption characteristics of aerosol particles. To further analyze the accuracy of the new algorithm, the aerosol CRI was applied as a key criterion for 452 aerosol identification. The CRI has two parts: imaginary and real. The imaginary part 453 indicates radiation absorption by aerosols, with a small value signifying a small 454 455 absorption. Because the radiation of aerosols is more dependent on the imaginary than the real part, the imaginary part is essential for inferring the optical properties and 456 aerosol types. Hence, we compared the real and imaginary parts of the CRI calculated 457 using the hybrid and Gaussian density clustering algorithms. 458

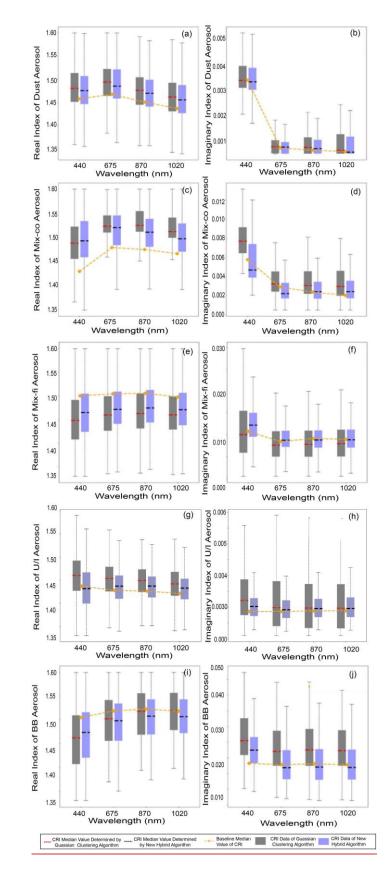
Figure 7 shows box plots of the aerosol CRI for dust, mixed-coarse, mixed-fine, U/I, and BB aerosols using the <u>new</u> hybrid classification and Gaussian <u>kernel</u> density clustering algorithms. Based on the principle that the CRI of aerosols is fixed under ideal conditions, the closer the median value of the CRI of the identified aerosol type is to the median value of the benchmark CRI, the more accurate is the identification method.

As shown in Figures 7 (a) and (f), the median values of the CRI real part for dust 465 aerosol are in the range 1.45–1.53 at four bands, and those of the imaginary part are 466 0.003-0.004 at 440 nm; further, the values in other bands decrease rapidly as 467 wavelength increases. The imaginary part of CRI represents the absorption of light by 468 the aerosol, with a small absorption indicating strong scattering. The results of the 469 imaginary part are consistent with the spectral dependence properties of dust-based 470 aerosols according to the wavelength. This is primarily because dust aerosols, 471 composed of clay, quartz, and hematite, exhibit strong absorption in the blue band 472 (440 nm) and low absorption in the visible and near-infrared bands. For the dust 473

aerosols, the CRI determined by the two methods did not differ much. However, the
median value of the CRI obtained using the <u>new hybrid algorithm was slightly closer</u>
to the benchmark CRI than that obtained using the Gaussian <u>kernel</u> density clustering
algorithm for dust aerosols. Therefore, the <u>new hybrid algorithm was concluded to be</u>
more accurate in identifying dust aerosol.

Figures 7 (b) and (g) show the median values of the CRI real part for mixedcoarse aerosol is 1.47–1.55 at four bands using the new hybrid algorithm, but the imaginary part is 0.004–0.009 at 440 nm. However, the real part is 1.44-1.50 at four bands determined by Gaussian <u>kernel</u> density clustering algorithm, and the imaginary part is 0.006–0.009 at 440nm. The median value of the hybrid algorithm was closer to the baseline median value than that of the Gaussian <u>kernel</u> density clustering algorithm for both the real and imaginary parts.

486 Figures 7 (c) and (h) show the median value of the CRI real part for mixed-fine 487 aerosols determined using the new hybrid and Gaussian kernel density clustering algorithms, which was 1.42-1.51 at four bands. This result is close to the range (1.44-488 489 1.52) reported by Wu (2021) in Beijing using a random forest algorithm. The median CRI of the real part at four bands and imaginary part at the (675-870-1020 nm) bands 490 were close to the baseline median value for the new algorithm. Additionally, the 491 492 median value of the imaginary part was lower than that of the new hybrid algorithm and further from baseline data for the identifying aerosol type results mixed with 14% 493 494 coarse aerosols. Mixed coarse aerosols result in weaker absorption. Hence, the new

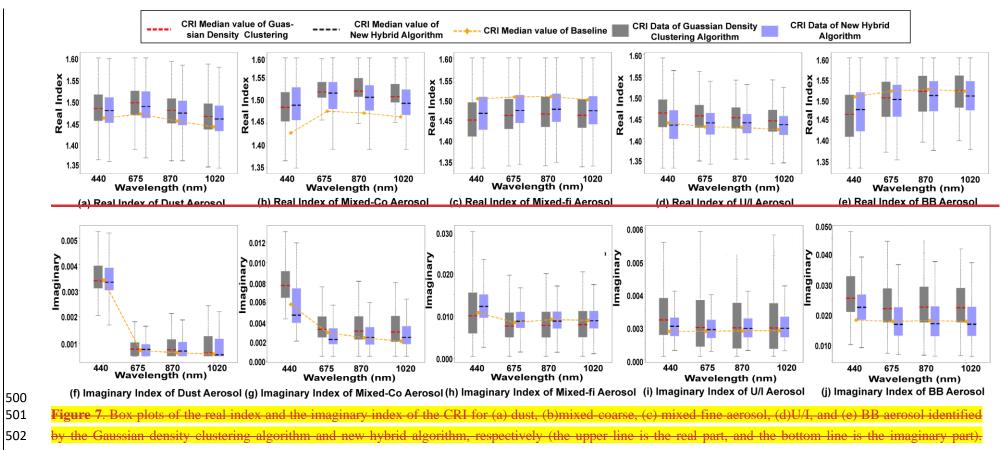


495

**Figure 7**. Box plots of the real index (left) and the imaginary (right) index of the CRI for (a-b)

497 <u>dust, (c-d) mixed-coarse, (e-f) mixed-fine aerosol, (g-h) U/I, and (i-j) BB aerosol identified by the</u>

498 Gaussian kernel density clustering algorithm and new hybrid algorithm, respectively.



503 hybrid algorithm performed better at identifying mixed-fine aerosols than the
504 Gaussian kernel density clustering algorithm.

505 Similarly, as seen in Figures 7 (d) and (i), the median value of the CRI real part 506 for U/I aerosol identified using the new hybrid algorithm was 1.39–1.47. This median 507 value was lower than that of the mixed-fine aerosols. This is because the real part 508 indicates the absorption ability of aerosols, and the absorption ability of U/I aerosols 509 was less than that of mixed-fine aerosols. For the imaginary part also, the new hybrid 510 algorithm performed slightly better than the Gaussian <u>kernel</u> density clustering 511 algorithm at the four bands.

512 For BB aerosols, the median value of the real part generated using the new hybrid algorithm differed slightly from that generated by the Gaussian kernel density 513 514 clustering algorithm. Additionally, the median obtained using the Gaussian kernel 515 density clustering algorithm was closer to the baseline. Furthermore, when analyzing the imaginary part, the new hybrid algorithm performed much better than the 516 517 Gaussian kernel density clustering algorithm. Even with a 100% concordance rate 518 between the new hybrid and Gaussian kernel density clustering algorithms in identifying BB aerosols, the refractive index still differed. This result indicates that 1% 519 520 of mixed-fine aerosols classified using the Gaussian kernel density clustering 521 algorithm were correctly identified as BB aerosols by the new algorithm. Overall, 522 these results demonstrate that the new algorithm is reliable.

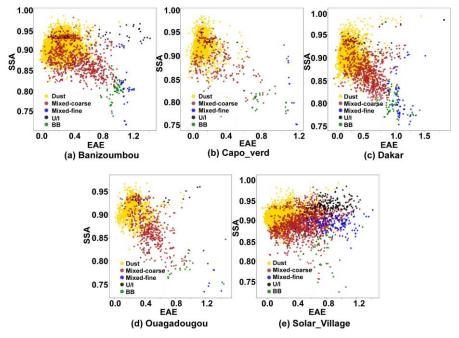
Additionally, in this study, the number of 326400 data points from optical
parameters database and 98000 observed data for calculation spans from Jan.1st,1993
to Dec.31st,2021, passing through Gaussian kernel density clustering algorithm and
new hybrid algorithm Python progresses, which is archived on the personal Windows
system computer (Intel® Core™ i7-10710U,16G DDR4 2666MHz, 512G PCIE SSD).
The computational time for the two algorithms indicates the new hybrid algorithm
runs faster than the Gaussian kernel density clustering algorithm with huge quantities

<sup>530</sup> of data and trained in advance, which can obtain aerosol type in 20 seconds, in
<sup>531</sup> contrast, it will take 30 to 40 seconds to obtain aerosol type in one site by using the
<sup>532</sup> Gaussian algorithm.

## 533 **4.2** Aerosol type determination for typical sites

## 534 **4.2.1 Dust aerosol**

535 Figure 8 shows the aerosol types obtained using the new hybrid algorithm for the five sites selected for dust aerosol identification. According to the prediction by the 536 new hybrid algorithm, the aerosols at these five sites mainly contained dust aerosols 537 538 along with a small amount of U/I, mixed-fine, and BB aerosols, and a large amount of mixed coarse aerosols. This shows that other types of aerosols invaded these areas 539 besides dust aerosol. BB aerosols may have been transferred from the southern 540 African savannah. Additionally, U/I aerosols could be from industrial cities, such as 541 542 Dakar, Abidjan, and Lagos, which are dominated by anthropogenic aerosols and are close to the AERONET sites. 543



545 Figure 8. Identification of dust aerosol at dominant aerosol sites.

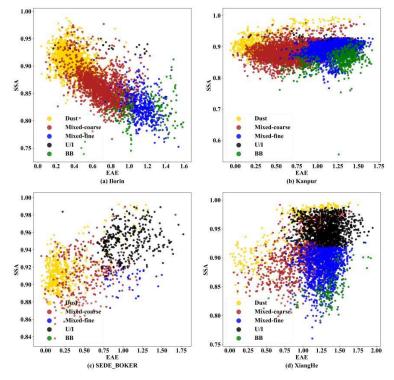
## 546 **4.2.2 Mixed aerosol**

544

547 Besides Ilorin in Africa, the mixed aerosol AERONET sites, including Kanpur,

Sede\_Boker, and XiangHe, are in Asia. The aerosol types at these four sites were 548 determined using the new hybrid algorithm (Figure 9). Mixed coarse aerosols 549 550 dominated the Kanpur, Ilorin, and Sede-Sede\_Boker sites, and mixed fine aerosols dominated XiangHe. Part of the dust in Xianghe could be due to the Takla Desert in 551 spring and the westerly winds prevailing in western China, which transported dust 552 aerosols over long distances. Additionally, the U/I aerosol in Xianghe could be a result 553 of human activities, construction emissions, and fuel burning in winter. The BB 554 aerosol was traced to the burning of a small amount of biomass in Xianghe, located in 555 a suburban area. 556

Furthermore, excluding dust aerosols, we observed BB and U/I aerosols in the Kanpur site in the Ganges Basin of India. A certain amount of U/I and dust aerosols were also observed in <u>Sede\_Sede\_Boker</u>, located in the industrial center of Israel, possibly from the Arabian desert. Lastly, Ilorin had the most dust and least BB aerosols because it is located in central Africa, often affected by the Saharan Desert and African savannah.

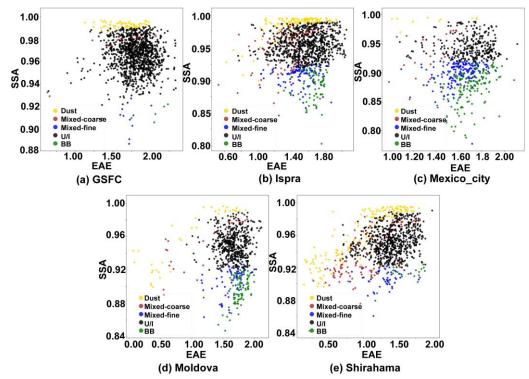


**Figure 9**. Same as Figure 8 but for Mixed aerosol.

#### 565 **4.2.3 Urban/industrial aerosol**

All the selected AERONET sites for evaluating the performance of the new hybrid algorithm in terms of U/I aerosol identification are in Europe or North America (Figure 10). GSFC is located in the densely populated and industrially developed area of Washington in the United States, explaining its complex aerosol type dominated by the U/I aerosol followed by a few mixed and BB aerosols and a small amount of dust aerosols.

Ispra is in Turin, one of Italy's largest industrial centers. However, dust-type 572 aerosols were identified, possibly transported from the Libyan desert when Italian 573 winters were controlled by southwesterly winds. Moreover, Mexico, where the 574 Mexico City site is located, is an industrialized country with modern industries and 575 agriculture, abundant oil production, and a dense population. Nevertheless, we 576 identified dust, mixed coarse, and BB aerosols in this site using the new hybrid 577 algorithm. These aerosol types could be from the Chihuahuan Desert, an inland desert 578 579 covering 12% of Mexico's area and a major source of coarse and dust aerosols. Additionally, the literature shows that Mexico City is surrounded by forested 580 mountains, which experience many wildfires during the dry period between 581 November and May; this accounts for BB aerosols in Mexico City (Yokelson et al. 582 2007). Finally, the BB aerosols identified at the Moldova site could be attributed to its 583 rich vegetation cover. 584

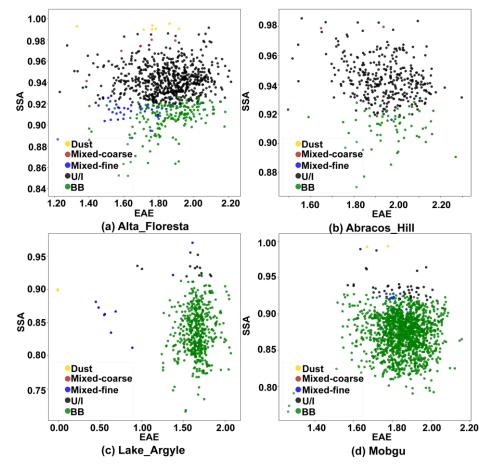


586 Figure 10. Same as Figure 9 but for urban/industrial aerosol.

# 587 4.2.4 Biomass burning aerosol

585

588 The selected sites were mainly located in the mountains and highlands. Figure 11 589 shows the aerosol types identified using the new hybrid algorithm. Large amounts of BB aerosols were identified at all sites. Additionally, a small amount of dust and 590 591 mixed-coarse aerosols were identified at the Alta Alta Floresta site, transported over 592 a long distance from the Patagonian Desert in Argentina, in southern South America. Moreover, the city where the site is located is industrially developed and has a large 593 population; therefore, more U/I aerosols were identified using the new hybrid 594 595 algorithm. The geographically close Abracos\_Hill and Alta-Alta\_Floresta sites were 596 characterized by the same aerosol type and source. Furthermore, one data point in Lake Argyle was classified as a dust aerosol. This means that, although the site is 597 located on the Kimberley Plateau, Australia has a large desert area, and coarse 598 aerosols still exist. Lastly, a few U/I and several dust-type aerosols were identified at 599 600 the Mongu site, possibly caused by aerosol emissions from nearby cities and dust transport from the Saharan Desert. 601





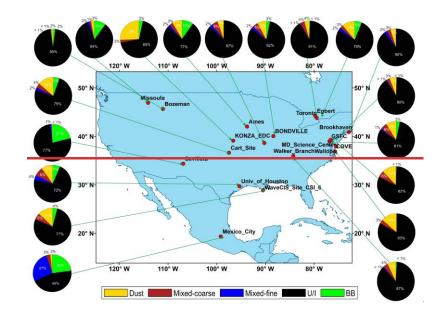
**Figure 11**. Same as Figure 10 but for BB aerosol.

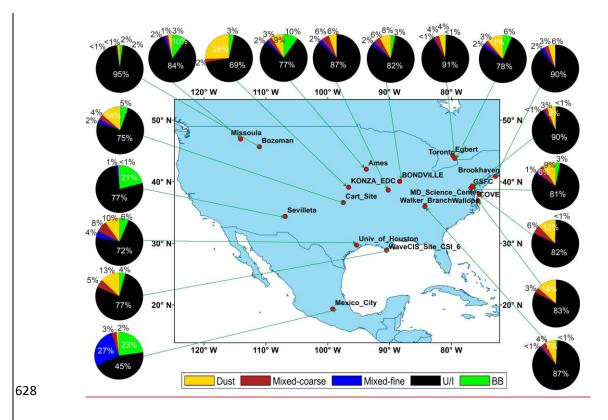
## 604 **4.3 Aerosol type distribution on a global scale**

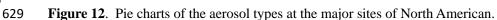
Given the advantages and accuracy of the new hybrid algorithm in identifying 605 606 aerosol types, we used it to divide the data of AERONET sites in different regions of the world to obtain global aerosol type distribution information. The aerosol types of 607 each continent are shown in Figures 12-16. Additionally, Figure 17 shows the global 608 aerosol-type distribution. Notably, the pie chart was placed on each site in the study, 609 which is a "point source" assessment of the aerosol type and does not represent the 610 entire region (the size of the pie chart is independent of the optical properties). 611 Moreover, the sites were screened, and only those with valid data of > 100 aerosol 612 types were considered; however, offshore sites and sites classified as marine aerosol-613 614 dominated by other literature were excluded.

Figure 12 shows pie charts of the aerosol types for each scanned AERONET site in North America. The U/I aerosols, particularly in most mid-eastern regions,

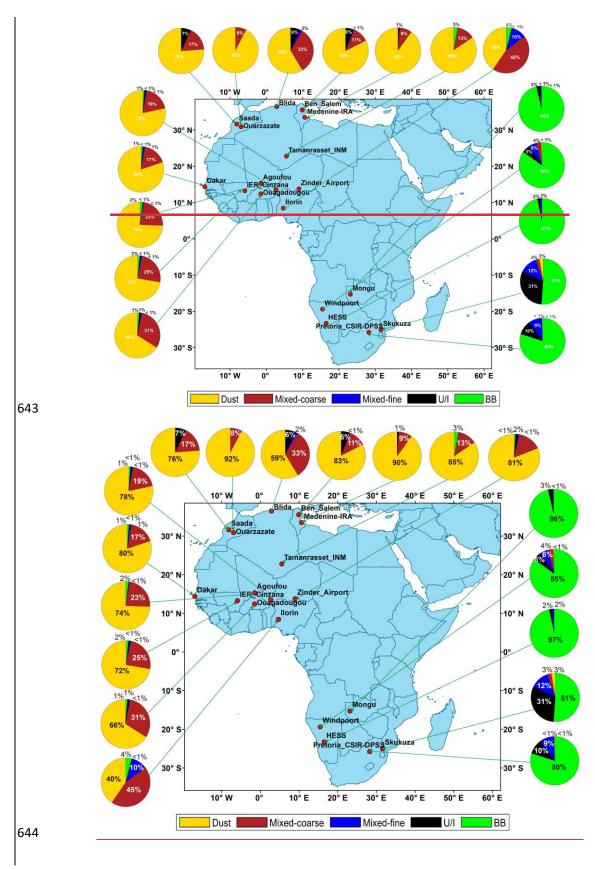
contained mixed and small amounts of biomass aerosols. Additionally, the AERONET 617 sites in large cities, such as Chicago, New York, Toronto, Ottawa, and Boston, had U/I 618 619 aerosols. Many studies have shown that dust aerosols from the Saharan Desert can cross the Atlantic Ocean to North America in summer. Moreover, there is an inland 620 desert in western North America, the Chihuahua Desert, responsible for a small 621 amount of dust and mixed aerosols at the AERONET sites in North America. 622 Additionally, wildfires in western North America and household wood burning 623 contribute to most BB aerosols yearly. The central region site is affected by the 624 environment, with an increased proportion of BB aerosols, and U/I aerosols are still 625 626 prevalent because the site is located in a large city and is densely populated.

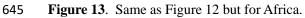






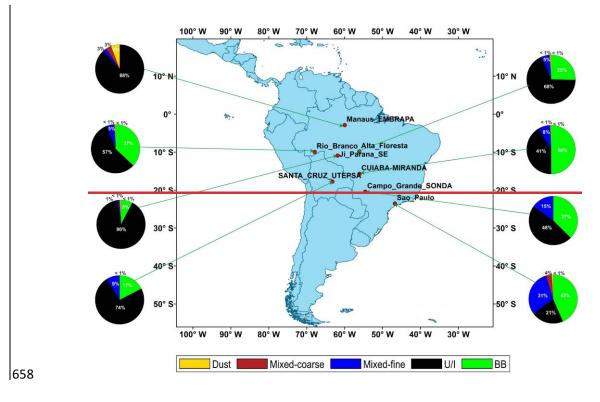
630 Figure 13 shows the aerosol types in Africa. Northern Africa has the largest desert in the world, the Saharan Desert; therefore, dust aerosols dominate north of the 631 equator in Africa. However, some AERONET sites in the Sudanese steppe were 632 primarily BB, with some U/I aerosols in nearby urban sites. The Ilorin site is a typical 633 mixed aerosol site close to the equator with a small amount of BB aerosols. Most sites 634 close to the Atlantic coast were affected by dust aerosols, even those on the islands of 635 636 CapeCapo\_Verde. The reliability of the new model in distinguishing U/I and BB aerosols is demonstrated. Sites in Southern Africa, such as Namibia, Botswana, and 637 638 Zambia, are dominated by BB aerosols. Nevertheless, studies have shown the presence of U/I aerosols at sites in the urban areas of South Africa. Although U/I and 639 BB aerosols are difficult to distinguish, the two can be identified in the context of a 640 large urban population and less biomass combustion, thus establishing the model's 641 accuracy. 642

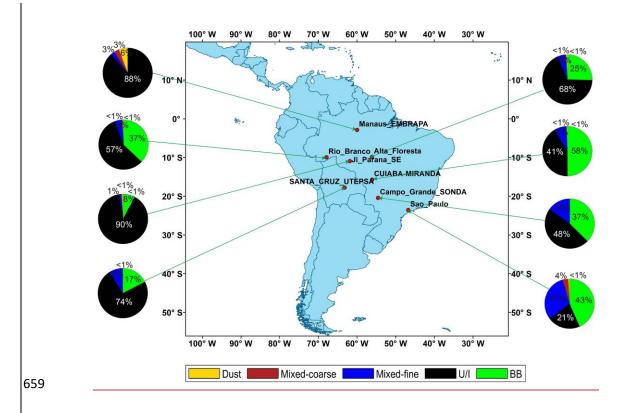




646 The aerosol types in South America are shown in Figure 14. Here, only eight sites

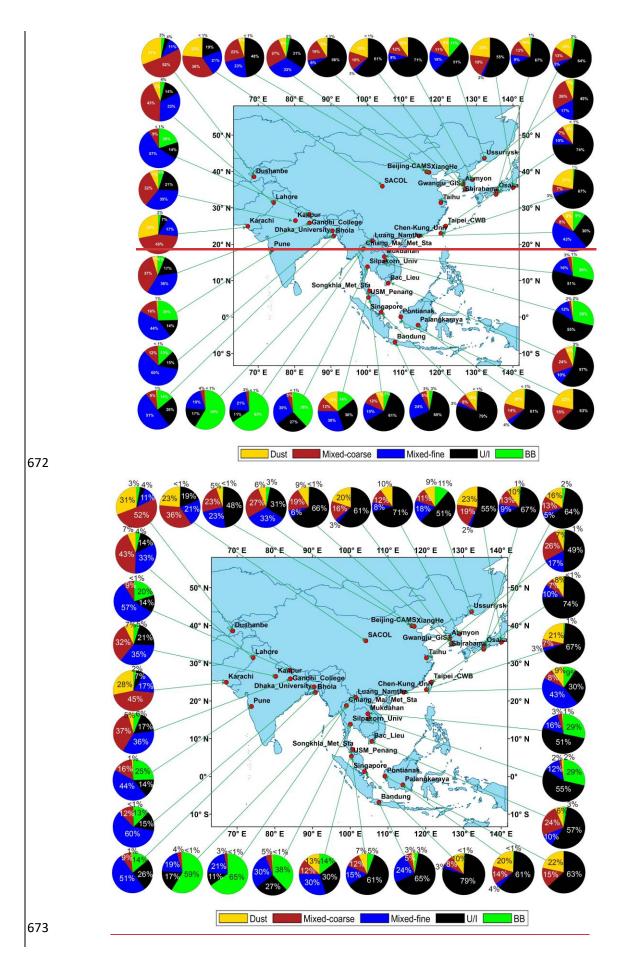
met the requirement for valid data >100 aerosol types. South America is mainly 647 dominated by mountainous plateaus, and under the influence of the Brazilian warm 648 current, many tropical rainforests are distributed in the south; therefore, the 649 background aerosols are mainly BB aerosols. As shown in Figure 14, large cities, such 650 as Rio Branco, Campo Grande, Manaus, Santa Cruz, and São Paulo, showed an 651 increased proportion of anthropogenic and mixed aerosols because of their large 652 population and developed industries. Due to the tropical rainforest climate in southern 653 South America, the proportion of BB aerosols increased, such as that at the Cuiaba 654 site near the Amazon River. Additionally, the Manaus site contained a small amount of 655 dust aerosols that were presumably transported across the Atlantic Ocean from 656 African dust at the same latitude. 657





**Figure 14**. Same as Figure 12 but for South America.

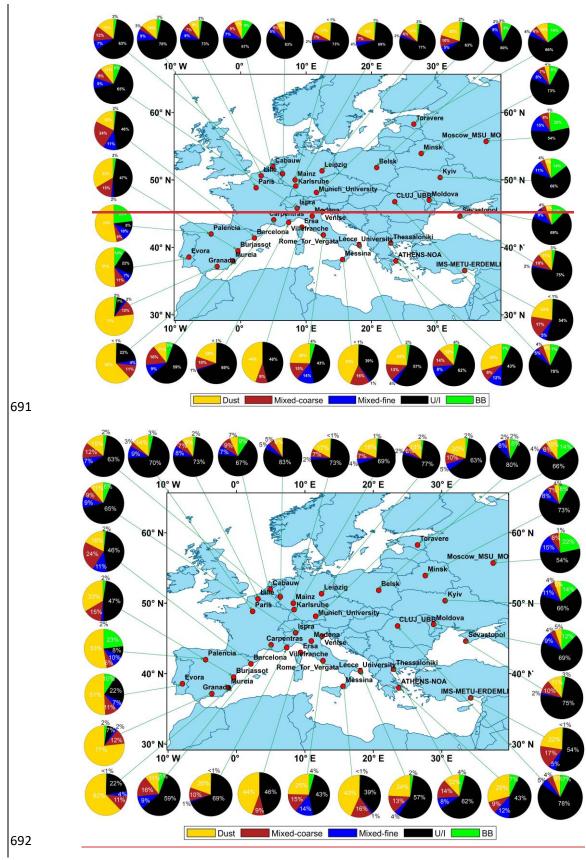
661 The aerosol types in Asia are shown in Figure 15. In western Asia, influenced by the Indian Desert, sites on the Indian Peninsula were dominated by coarse-particle 662 aerosols, including dust and mixed coarse aerosols. Kanpur and Pune are densely 663 populated cities in India, with more mixed-fine aerosols produced by human activities. 664 Additionally, in Southeast Asia, all sites contained BB aerosols, consistent with 665 Hamill (2014). This is because of the abundance of tropical rainforests in Southeast 666 Asia. Moreover, some urban sites, such as Singapore and Penang, had large numbers 667 of U/I and mixed-fine aerosols. The coastal areas of East Asia, which are densely 668 populated and industrially developed, were mainly dominated by U/I aerosols. 669 Moreover, dust aerosols appeared at these sites due to dust transported from the 670 Taklamakan Desert in East Asia. 671



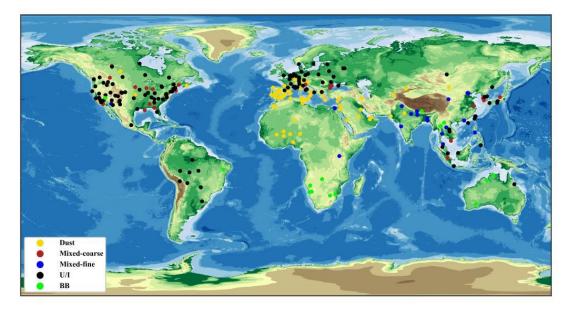
**Figure 15**. Same as Figure 12 but for Asia.

The inland areas of East Asia have a smaller population than the coastal areas; therefore, the proportion of U/I aerosols was small, and that of mixed aerosols was high. Generally, mixed aerosols are more easily overestimated than U/I aerosols; however, the new hybrid algorithm identified a larger proportion of U/I aerosols than mixed aerosols at Asian sites. Therefore, this new hybrid algorithm can be considered for improving the classification of mixed aerosols versus U/I aerosols.

Similarly, southern Europe, which is close to the Saharan and Arabian deserts, 681 was dominated by dust aerosols, with small amounts of mixed and U/I aerosols. 682 Northern European sites have many cities and a large population; therefore, the 683 aerosol type was mainly U/I aerosols, identified using the new hybrid algorithm 684 (Figure 16). Additionally, small amounts of BB aerosols were identified at most sites 685 in Europe because of olive groves in agricultural lands in the EU, which produce 91% 686 of the world's olive oil (Lopez-Pineiro et al., 2011). Papadakis et al. (2015) suggested 687 that the biomass produced from olive oil is used for heating and industry, and its 688 689 combustion produces carbonaceous aerosols, considered the major source of fine particle aerosols in Europe during winter (Puxbaum et al., 2007). 690



**Figure 16**. Same as Figure 12 but for Europe.



694 695

Figure 17. Global dominant aerosol types distribution based on AERONET sites.

The global distribution of dominant aerosols in the AERONET site is shown in 696 Figure 17. The graph does not include marine aerosols. There are more aerosol sites 697 on the global map than those on each continent because AERONET sites with > 5698 years of data were selected for the global map; however, sites with > 100 valid data 699 700 points were required for each continent. The global distribution map shows that many BB aerosols were distributed between 20°N and 20°S. This is because this region has 701 a predominantly tropical rainforest climate, with many tropical rainforests and more 702 carbon-containing aerosol emissions. This finding is consistent with those from 703 previous studies that found that global BB aerosols mainly originate from Africa 704 (approximately 52%), followed by South America (approximately 15%), equatorial 705 Asia (approximately 10%), boreal forests (approximately 9%), and Australia 706 (approximately 7%) (Van G. R. et al., 2010). Furthermore, the global distribution map 707 708 shows a clear distribution band of dust aerosols between 5°N and 35°N, originating from the Saharan Desert in Africa and the Saudi Arabian Desert in Western Asia, 709 which are transported across the ocean to other regions. 710

## 711 **5.** Conclusion

712

We developed a new hybrid algorithm to support the rapid classification of

aerosol types by building an aerosol optical database for global AERONET sites. This 713 hybrid algorithm is a complex aerosol-type processing algorithm that effectively 714 715 integrates machine learning and density clustering algorithms. Additionally, this algorithm is not limited by the amount of data and improves the accuracy of aerosol-716 type classification. On investigating the aerosol types at specific sites with dominant 717 718 aerosols, we observed that different sites contained one or more aerosol types, with the composition of some specific dominant aerosol sites being more complex than that 719 720 of others. The new algorithm showed a higher accuracy than that shown by algorithms 721 used in previous studies in identifying aerosol types at specific sites, particularly in distinguishing between U/I and mixed-fine aerosols. Finally, the recognition results of 722 723 the new hybrid algorithm were closer to the baseline CRI, confirming that the new 724 hybrid algorithm is better than the density-clustering algorithm. On investigating the aerosol types at global sites across the continents using the new algorithm, we 725 observed the dominance of different types of aerosols at different sites, and the 726 composition of these could be logically and effectively attributed to the geographical 727 728 location, energy consumption structure, meteorological conditions and activities 729 happening at the respective sites.

730 In this study, the existing aerosol type identification algorithm was improved using global ground-based AERONET optical property parameter data, and the spatial 731 distribution characteristics of global aerosol types were analyzed, which impacted 732 aerosol radiation research and optical thickness inversion accuracy. Additionally, the 733 734 presumption of spherical dust aerosols in the Mie scattering model diverges from their actual non-spherical nature in the environment, introducing potential inaccuracies. 735 The optical database's precision, therefore, necessitates further refinement. Future 736 advancements could involve adopting more potent machine learning techniques, such 737 as advanced algorithms beyond the current random forest method. Meanwhile, multi-738 739 source satellite data and reanalysis products can be incorporated into aerosol-type 740 identification. Ultimately, this study will provide support for the identification and control of air pollution sources. 741

742

In this study, the existing aerosol type identification algorithm was improved

743 using global ground-based AERONET optical property parameter data, and the spatial

- 744 distribution characteristics of global aerosol types were analyzed, which impacted
- 745 aerosol radiation research and optical thickness inversion accuracy. However, marine

## 746 Author contributions

Feng Zhang designed the study. Xiaoli Wei analyzed the results, and wrote the original draft. Qian Cui engaged in data processing, manuscript editing, and restructuring. Qian Cui collected and processed the data. Leiming Ma revised the paper and given constructive suggestions. Wenwen Li constructive comments on the

paper. **Peng Liu** revised the paper. All authors contributed to the study.

## 752 **Competing interests**

The authors declare that they have no conflict of interest.

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