

1 **The regime of aerosol asymmetry parameter over**
2 **Europe, Mediterranean and Middle East based on**
3 **MODIS satellite data: evaluation against surface**
4 **AERONET measurements**

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13

14 **Abstract**

15 Atmospheric particulates are a significant forcing agent for the radiative energy
16 budget of the Earth-atmosphere system. The particulates' interaction with radiation,
17 which defines their climate effect, is strongly dependent on their optical properties. In
18 the present work, we study one of the most important optical properties of aerosols,
19 the asymmetry parameter (g_{aer}), in the region comprising North Africa, the Arabian
20 peninsula, Europe, and the Mediterranean basin. These areas are of great interest,
21 because of the variety of aerosol types they host, both anthropogenic and natural.
22 Using satellite data from the collection 051 of MODIS (MODerate resolution Imaging
23 Spectroradiometer, Terra and Aqua), we investigate the spatio-temporal
24 characteristics of the asymmetry parameter. We generally find significant spatial
25 variability, with larger values over regions dominated by larger size particles, e.g.
26 outside the Atlantic coasts of north-western Africa, where desert-dust outflow takes
27 place. The g_{aer} values tend to decrease with increasing wavelength, especially over
28 areas dominated by small particulates. The intra-annual variability is found to be
29 small in desert-dust areas, with maximum values during summer, while in all other
30 areas larger values are reported during the cold season and smaller during the warm.
31 Significant intra-annual and inter-annual variability is observed around the Black Sea.
32 However, the inter-annual trends of g_{aer} are found to be generally small.

33 Although satellite data have the advantage of broad geographical coverage, they have
34 to be validated against reliable surface measurements. Therefore, we compare
35 satellite-measured values with g_{aer} values measured at 69 stations of the global surface
36 AERONET (Aerosol Robotic Network), located within our region of interest. This
37 way, we provide some insight on the quality and reliability of MODIS data. We report
38 generally better agreement at the wavelength of 870 nm (correlation coefficient R up
39 to 0.47), while of all wavelengths the results of the comparison were better for spring
40 and summer.

41

42 **1 Introduction**

43 Atmospheric aerosol particles interact with radiation, mainly the short wave (SW or
44 solar) part of the spectrum, modifying the energy budget of the Earth-atmosphere
45 system. The aerosol effect is either direct, through the scattering and absorption of
46 solar radiation, and thus reducing the incoming solar radiation flux at the surface,
47 indirect, through the modification of cloud properties, or semi-direct, due to the
48 absorption of solar radiation and consequent modification of the atmospheric
49 temperature profile, convection, and cloud properties (e.g. Graßl, 1979; Hansen, 1997;
50 Lohmann and Feichter, 2005).

51 The interaction of particles with the solar flux, which defines their climate role,
52 strongly depends on their optical properties (Hatzianastassiou et al., 2004; 2007),
53 which cannot be covered globally by surface in situ measurements. Besides the
54 aerosol optical depth (AOD), one of the most important optical properties of
55 atmospheric particles, which is used in radiative transfer, climate, and general
56 circulation models, is the asymmetry parameter (g_{aer}). The asymmetry parameter
57 describes the angular distribution of the scattered radiation and determines whether
58 the particles scatter radiation preferentially to the front or back. The globally available
59 satellite based AOD data are considered to a great extent as reliable and adequate, due
60 to significant developments in surface and satellite measurements during the last two
61 decades, and particularly the arrival of MODIS in 2000, which is regarded as one of
62 the most reliable datasets (Bréon et al., 2011; Nabat et al., 2013). On the other hand,
63 despite the important role of the asymmetry parameter, relevant global coverage data
64 are measured only for the few last years, or are available in long-term aerosol
65 climatologies such as Global Aerosol Data Set (GADS, Koepke et al. 1997) and Max
66 Planck Aerosol Climatology (MAC, Kinne et al., 2013). Even so, asymmetry

67 parameter data are usually examined for regions with limited geographical extent and
68 temporal coverage (Di Iorio et al, 2003), without intercomparison between alternative
69 data platforms.

70 The goal of the present work is the study of the spatiotemporal distribution of the
71 aerosol asymmetry parameter, using the most recent data from MODIS (MODerate
72 resolution Imaging Spectroradiometer, collection 051). Emphasis is given to the
73 comparison between the provided MODIS data and respective reliable surface
74 measurements of the global AERONET, in order to gain insight on the quality of the
75 former.

76 For this study we focus on the region defined by latitudes 5°N to 70°N and longitudes
77 25°W to 60°E, including North Africa, the Arabian peninsula, Europe, and the greater
78 Mediterranean basin (Fig. 1). This area is selected because it is of particular scientific
79 interest due to the simultaneous presence of a variety of particles, both natural and
80 anthropogenic (e.g. desert dust, marine, biomass burning, anthropogenic urban /
81 industrial pollution) as shown in previous studies (Lelieveld et al., 2002; Smirnov et
82 al., 2002; Sciare et al., 2003; Pace et al., 2006; Lyamani et al., 2006; Gerasopoulos et
83 al., 2006; Engelstaedter et al., 2006; Satheesh et al., 2006; Kalivitis et al., 2007; Rahul
84 et al., 2008; Kalapureddy et al., 2009; Alonso-Pérez et al., 2012; Zuluaga et al., 2012;
85 Kischka et al., 2014) which makes this area ideal for aerosol studies. The presence of a
86 variety of aerosols in the area is due to the fact that two of the largest deserts of the
87 planet are partly included in our area of interest, i.e. the Arabian desert and the
88 Sahara, while one finds also significant sources of anthropogenic pollution from urban
89 and industrial centres, mainly in the European continent. Moreover, our area of
90 interest and primarily its desert areas are characterised by a large aerosol load (large
91 optical depth, Remer et al. 2008; Ginoux et al. 2012). In addition, significant regions
92 in this area, more specifically the Mediterranean basin and North Africa, are
93 considered climatically sensitive, since they are threatened by desertification (IPCC,
94 2007; 2013). Finally, one more reason for the selection of study area is that the
95 present study complements previous ones made by our team (e.g. Papadimas et al.,
96 2008, 2012; Hatzianastassiou et al., 2009) analysing other key aerosol optical
97 properties, namely AOD, for the same region. This is the first study (to our
98 knowledge) that focuses on asymmetry parameter over a geographically extended
99 area, while at the same time compares satellite with ground-station data.

101 2 Data

102 Before presenting the data used in this study, a short introduction of the parameter
 103 studied is given here for readers more or less unfamiliar with it. The asymmetry
 104 parameter (or factor) is defined by:

105

$$106 \quad g = \frac{\bar{\omega}_1}{3} = \frac{1}{2} \int_{-1}^1 P(\cos \Theta) \cos \Theta d \cos \Theta \quad (1)$$

107 where P is the phase function, which represents the angular distribution of the
 108 scattered energy as a function of the scattering angle Θ and it is defined for molecules,
 109 cloud particles, and aerosols. The phase function can be expressed using the Legendre
 110 polynomials $\bar{\omega}_l$ (see Liou, 2002) and $\bar{\omega}_1$ in Eq. (1) stands for $l=1$. The asymmetry
 111 parameter is the first moment of the phase function and it is an important parameter in
 112 radiative transfer. For isotropic scattering, g equals zero, which is the case for
 113 Rayleigh molecular scattering. The asymmetry parameter increases as the diffraction
 114 peak of the phase function sharpens. For Lorenz-Mie type particles, namely for
 115 aerosols and cloud droplets, the asymmetry parameter takes positive values denoting a
 116 relative strength of forward scattering, with values increasing with particle size. It can
 117 also take negative values if the phase function peaks in backward directions (90-
 118 180°). The phase function along with the extinction coefficient (or equivalently the
 119 optical depth) and the single scattering albedo, constitute the fundamental parameters
 120 that drive the transfer of diffuse intensity (Joseph et al., 1976). The asymmetry
 121 parameter itself is a simple expression of the phase function (being its first moment)
 122 and it is used in many radiative transfer and climate models. Hence, the importance of
 123 aerosol asymmetry parameter is easily understood for enabling computations of
 124 aerosol radiative properties and effects (e.g. forcings).

125 Daily data of the aerosol asymmetry parameter (g_{aer}) are used for the needs of this
 126 work. In order to achieve the largest geographical coverage of the studied region, we
 127 employ satellite data from the MODIS-Terra and MODIS-Aqua datasets. These data
 128 are compared with in-situ measurements at stations of the AERONET. We provide a
 129 detailed description of the utilised data in the following sections.

130

131 2.1 Satellite MODIS Terra and Aqua data

132 MODIS is an instrument (radiometer) placed on the polar-orbiting satellites of NASA
133 (National Aeronautics and Space Administration) Terra and Aqua, 705 km from the
134 Earth, in the framework of the Earth Observing System (EOS) programme. Terra was
135 launched on 18 December 1999, while Aqua was launched on 4 May 2002. The two
136 satellites are moving on opposite directions and their equatorial crossing times are at
137 10:30 (Terra) and 13:30 (Aqua). MODIS is recording data in 36 spectral channels
138 between the visible and the thermal infrared (0.44 – 15 μm), while its swath width is
139 of the order of 2330 km, which results in almost full planetary coverage on a daily
140 basis.

141 Aerosol properties are monitored in 7 spectral channels between 0.47 and 2.13 μm
142 and final results are derived through algorithms developed for aerosol quantities both
143 over land and ocean (Kaufman et al., 1997; Tanré et al., 1997; Ichoku et al., 2002;
144 Remer et al., 2005). MODIS data are organised in “collections” and “levels”.
145 Collections comprise data produced by similar versions of the inversion algorithms,
146 with the recent collection “051” including also outputs from the “Deep Blue”
147 algorithm. Levels are characterised by data of different quality analysis and spatial
148 resolution.

149 In this study we use daily MODIS data for the asymmetry parameter (g_{aer}) provided
150 on an $1^\circ \times 1^\circ$ grid (namely 100x100 km), from Collection 051, Level 3. These data
151 were measured at wavelengths 470, 660, and 870 nm, only over oceanic regions, since
152 they were derived through the algorithm “Dark Target” over ocean. The period of
153 analysis stretches from 24-2-2000 to 22-9-2010 for MODIS-Terra and from 4-7-2002
154 to 18-9-2010 for MODIS-Aqua.

155 The MODIS C051 g_{aer} data are a derived product of the MODIS algorithm over
156 ocean. This MODIS algorithm (http://modis.gsfc.nasa.gov/data/atbd/atbd_mod02.pdf,
157 Remer et al., 2006) retrieves as primary products the AOD at 550 nm, the Fine
158 (Mode) Weighting (FW, also known as fraction of fine-model aerosol type, FMF) and
159 the Fine (f) and Coarse (c) modes used in the retrieval, along with the fitting error (ϵ)
160 of the simulated spectral reflectance. The algorithm reports additional derived
161 parameters, such as the effective radius (r_e) of the combined size distribution, the
162 spectral total, fine and coarse AODs or the columnar aerosol mass concentration.
163 Among them, g_{aer} is also derived and reported at seven (7) wavelengths, 470, 550,
164 660, 860, 1200, 1600 and 2120 nm. The derived parameters are calculated (Levy et
165 al., 2013) from information contained within the look-up table (LUT) and/or other

166 retrieved products. For example, knowing the resulting total AOD and FMF, and
167 which aerosol types were selected (or assumed), one can go back to the LUT, and
168 recover additional information about the retrieved aerosol, such as the g_{aer} . Hence, it
169 should be noted that the derived g_{aer} product is dependent on the used aerosol models
170 (modes), since the algorithm is based on a LUT approach, assuming that one fine and
171 one coarse lognormal aerosol modes can be combined with appropriate weightings to
172 represent the ambient aerosol properties over the target (spectral reflectance from the
173 LUT is compared with MODIS-measured spectral reflectance to find the “best” –
174 least squares – fit, which is the solution to the inversion). In the C051 algorithm there
175 are four fine modes and five coarse modes, for which the spectral (at the
176 aforementioned 7 wavelengths) aerosol asymmetry parameter values are given in
177 Remer et al (2006).

178 We also used Level 3 daily Ångström exponent data from MODIS-Aqua C051, and
179 also spectral aerosol optical depth data from MODIS-Aqua C006 datasets, from which
180 we computed C006 Ångström exponent. These data were used to assess the validity of
181 g_{aer} data and their temporal tendencies, as discussed in section 3.2.3.

182

183 **2.2 Ground based AERONET data**

184 AERONET (AErosolROboticNETwork) is a global network of stations focused on
185 the study of aerosol properties. AERONET currently encompasses about 970 surface
186 stations (number continuously evolving) equipped with sun photometers of type
187 CIMEL Electronique 318 A (Holben et al., 1998), which take spectral radiation flux
188 measurements.

189 The optical properties of aerosols are extracted through the application of inversion
190 algorithms (Dubovik and King, 2000). Data are provided on three levels (1.0, 1.5, and
191 2). In the present work, we use the most reliable cloud-screened and quality assured
192 Level 2 data. AERONET calculates the asymmetry parameter at wavelengths 440,
193 675, 870, and 1020 nm. We employ daily Level 2 asymmetry parameter data from 69
194 stations (Fig. 1), contained in our study area (N. Africa, Arabian peninsula, Europe).
195 We choose only coastal stations, in order to maximize the coexistence of satellite
196 marine g_{aer} data with surface data. Also, in order to compare corresponding data
197 between the satellite and station platforms, we perform comparison only for the 440,
198 675 and 870 nm.

199

200 3 Satellite based results

201 3.1 Geographical distributions

202 The spatial distribution of annual mean values of g_{aer} is given in Fig. 2 separately at
203 the wavelengths 470, 660 and 870 nm. The values are averages over the common
204 period between Terra and Aqua, namely 4 July 2002 till 18 September 2010. A
205 significant spatial variability is evident, with MODIS-Terra values varying within the
206 ranges 0.63 - 0.76, 0.57 – 0.75, and 0.55 – 0.74, at 470, 660 and 870 nm, respectively.
207 The results exhibit a decreasing tendency of g_{aer} with increasing wavelength,
208 consistent with the theory. Similar results are also obtained from MODIS-Aqua, but
209 with slightly smaller values than Terra by up to 0.02 on average. More specifically,
210 the corresponding ranges of wavelengths are 0.63 - 0.75, 0.57 – 0.73, and 0.55 – 0.73.
211 The smaller Aqua than Terra g_{aer} values could be attributed to smaller sizes of
212 aerosols in midday than morning, corresponding to passages of Aqua and Terra,
213 respectively, associated with lower relative humidity values and shrinking of aerosol
214 particles. It should be reminded that the ability of atmospheric aerosol to absorb water
215 affects the particle size (hygroscopic growth), as described by Köhler theory in the
216 early 20th century. It is also well known that relative humidity significantly affects
217 aerosol optical properties (e.g. Pilinis et al., 1996; Kondratyev, 1999), namely AOD,
218 single scattering albedo and g_{aer} , by modifying the aerosol liquid water content, size
219 and hence extinction coefficient and refractive indices. More specifically, aerosol
220 asymmetry parameter increases with increasing aerosol size, as noted in sect. 2. Such
221 diurnal variation has been also reported for AOD (Smirnov et al., 2002; Pandithurai et
222 al., 2007), but either decreasing or increasing in the day because of the influence of
223 other factors too, e.g. emissions or wind conditions, apart from aerosol
224 hygroscopicity.

225 In general, the largest g_{aer} values (deep red colors) are observed off the coasts of West
226 Africa (eastern tropical Atlantic Ocean) at all three wavelengths. High values are also
227 found over the Red and Arabian Seas. These high values are due to strong dust
228 outflows from the Saharan and Arabian deserts carrying out coarse aerosol particles
229 (Prospero et al., 2002; Alonso-Pérez et al., 2012; Miller et al., 2008) and causing
230 strong forward scattering. Nevertheless, the Persian Gulf region, which is surrounded
231 by deserts, is characterized by relatively smaller g_{aer} values. More specifically, values

232 as small as 0.69 (MODIS-Terra) and 0.67 (MODIS-Aqua) are observed in this region
233 at 470 nm, while at the longer wavelengths (660, 870 nm) the smallest values are
234 equal to 0.66 (Terra) and 0.64 (Aqua). The smaller g_{aer} values over the Persian Gulf
235 can be attributed to the presence of fine aerosols, which is corroborated by the low
236 effective radius and large fine-fraction measurements by MODIS over the Persian
237 Gulf, compared to neighbouring areas (not shown here). These fine particles originate
238 from the industrial activities in the Gulf countries related to oilfields or refineries
239 (Goloub and Arino, 2000; Smirnov et al., 2002a,b; Dubovik et al., 2002).

240 The high g_{aer} values over the northeastern tropical Atlantic Ocean as well as west of
241 the Iberian coasts are possibly related with the presence of coarse sea salt particles.
242 On the other hand, the asymmetry parameter takes clearly smaller values over the
243 Black Sea, where according to MODIS-Terra varies between 0.63 and 0.7 at 470 nm,
244 0.57 and 0.67 at 660 nm, and 0.55 and 0.66 at 870 nm, with the smallest values
245 appearing over the Crimean peninsula (corresponding maximum Aqua values are
246 smaller by 0.02). The small Black Sea g_{aer} values can be associated with industrial but
247 also biomass burning activities in nearby countries. A region of special interest is the
248 Mediterranean basin since it hosts a large variety of aerosols like anthropogenic,
249 desert dust or sea salt (e.g. Barnaba and Gobbi, 2004). The MODIS results over this
250 region show relatively small g_{aer} values, secondary to those of Black Sea,
251 characterized by an increase from north to south, which is more evident at 660 and
252 870 nm. More specifically, based on MODIS-Terra, g_{aer} over the Mediterranean takes
253 values from 0.68 to 0.74 at 470 nm, while at 670 and 870 nm it ranges from 0.64 to
254 0.73 and 0.62 to 0.72, respectively. According to MODIS-Aqua the g_{aer} values are
255 slightly smaller again. The observed low values in the northern parts of the
256 Mediterranean are probably associated with the presence of fine anthropogenic
257 aerosols transported from adjacent urban and industrial areas in the north, especially
258 in central Europe. In contrast, the higher g_{aer} values in the southern Mediterranean,
259 particularly near the North African coasts, can be explained by the proximity to the
260 Sahara desert and the frequent transport of significant amounts of coarse dust (e.g.
261 Kalivitis et al., 2006; Hatzianastassiou et al., 2009; Gkikas et al., 2009; 2014).

262 The spatial distributions of climatological monthly mean g_{aer} values from MODIS-
263 Aqua at 470 nm reveal significant differences in the range and the patterns of the
264 seasonal variability, depending on the area (Fig. 3). Thus, in tropical and sub-tropical
265 areas of Atlantic Ocean (up to about 30°N), where dust is exported from Sahara, g_{aer}

266 keeps high values throughout the year, which reach or even exceed 0.74 locally. Over
267 the regions of Arabian and Red Seas and the Gulf of Aden, which also experience
268 desert dust transport, larger g_{aer} values appear in the period from March to September,
269 with a maximum in August (locally as high as 0.75-0.76). This seasonal behavior is in
270 line with intra-annual changes of dust production over the Arabian peninsula indicated
271 by MODIS Ångström Exponent (AE) and Deep Blue aerosol optical depth data
272 (Ginoux et al., 2002), as well as over southwest Asia through in-situ data (Rashki et
273 al., 2012), aerosol index from various platforms and MODIS Deep Blue AOD data
274 (Rashki et al., 2014). Indeed, the production of dust there is relatively poor in winter,
275 increases in March and April and becomes maximum in June and July (Prospero et
276 al., 2002). Over the Arabian Sea, it is known that large amounts of desert dust are
277 carried out during spring and early summer (Prospero et al, 2002; Savoie et al., 1987;
278 Tindale and Pease, 1999; Satheesh et al., 1999). Nevertheless, according to MODIS,
279 the seasonal variability of g_{aer} remains relatively small there in line with a small
280 seasonal variability in MODIS Deep Blue AE data (results of our analysis, not shown
281 here). This can be explained by the presence of sea salt coarse particles throughout the
282 year, with which dust particles co-exist.

283 A greater seasonal variability exists over the Persian Gulf, where g_{aer} values are
284 higher during spring and in particular in summer (up to 0.74 at 470 nm according to
285 Aqua), and smaller in autumn and winter (area-minimum values smaller than 0.65).
286 This seasonal behavior can be explained taking into account the meteorological
287 conditions over the greater area of the Gulf; mainly in spring and summer dry
288 northwestern winds (Shamal) blow from northwest carrying desert dust from the arid
289 areas of Iraq (Heishman 1999; Smirnov et al. 2002a,b; Kutiel and Furman, 2003). The
290 transport of dust is gradually decreased in autumn and reaches its minimum in winter.
291 When the presence of desert dust is limited, a significant fraction of total aerosol load
292 in the region consists of fine anthropogenic particles (Smirnov et al. 2002a,b), which
293 can explain the observed relatively small g_{aer} values in autumn and winter.

294 In the Mediterranean basin g_{aer} exhibits a relatively small seasonal variation, with
295 lower values tending to appear in summer and secondarily in early and late spring, in
296 line with the stronger presence of dust in the area, transported from the Sahara desert
297 (Gkikas et al., 2013). On the contrary, over the Black Sea, a clear seasonal cycle is
298 apparent, with higher values in the cold period of the year and smaller in the warm
299 one. More specifically, according to MODIS-Aqua, the values at 470 nm drop down

300 to 0.61 in summer months whereas they reach 0.7 in January and December. This
301 seasonality is in agreement with the summer biomass burning from agricultural
302 activities and wildfires (Barnaba et al., 2011; Bovchaliuk et al., 2013), and the
303 resulting abundance of fine particles.

304 It is also interesting to look at the geographical distribution of monthly g_{aer} values in
305 latitudes higher than $50^{\circ}N$, for which annual mean values were not given in Fig. 2
306 because of unavailability of data for all months. Off shore northern France (English
307 Channel) and Germany the asymmetry parameter has small seasonally constant values
308 (note that data do not exist for January and February). In these areas, the aerosol load
309 consists mainly of anthropogenic polluted particles, which explains the small g_{aer}
310 values there.

311 In the Baltic Sea (values available from March to October) g_{aer} shows a significant
312 spatial and temporal variability. More specifically, it is small during summer whereas
313 it increases, locally up to more than 0.7, in March and October. The smaller summer
314 values can be explained by the presence of fine aerosols in the Baltic Sea originating
315 from forest fires in Europe and Russia (Zdun et al., 2011). On the contrary, in autumn
316 the local aerosol loading consists largely of coarse marine aerosols. It is also
317 important to note that the Baltic Sea hosts significant amounts of anthropogenic
318 industrial and urban aerosols throughout the year, but especially in summer (Zdun et
319 al., 2011).

320 In the higher latitudes of Atlantic Ocean, where the presence of maritime aerosols is
321 dominant, we note a remarkable month by month variation of asymmetry parameter,
322 with low values in summer (values up to 0.59) against high values (up to 0.75-0.77) in
323 spring (March, April) and autumn (October). This difference is possibly explained by
324 the seasonal variability of aerosol size in the northern Atlantic. Apart from the
325 presence of coarse sea salt throughout the year, in spring and summer small particles
326 are formed through photochemical reactions of dimethylsulphide (DMS) emitted by
327 phytoplankton decreasing the aerosol size. Moreover during summer fine
328 anthropogenic aerosols are transported in the region from North America (Yu, 2003;
329 Chubarova, 2009). These result in lower g_{aer} values between May and August.

330 Based on MODIS-Terra, the patterns of spatial distribution are generally the same
331 with Aqua, with slightly larger g_{aer} values. At larger wavelengths (660, 870 nm) a
332 decrease of g_{aer} is observed, especially for its smallest values. Further details and an

333 overall picture are given in section (3.2.1) which deals with climatological monthly
334 mean values not at the pixel but at the regional level.

335

336 **3.2 Temporal variability**

337 **3.2.1 Seasonal variability**

338 In order to provide an easier assessment of the seasonal cycle of aerosol asymmetry
339 parameter and its changes from one region to another, but also among the different
340 wavelengths (470, 660 and 870 nm), the study region was divided in 6 smaller sub-
341 regions (see Fig. 1). The average values of monthly mean climatological data of the
342 pixels found within each sub-region's geographical limits have been computed and
343 are given in Fig. 4, for every wavelength, both for Terra and Aqua. It appears that the
344 seasonal cycle differs between the sub-regions, as it has been already shown in the
345 geographical map distributions discussed in the previous section.

346 At 470 nm (Fig. 4i), the intra-annual variability of g_{aer} is greater over the Black Sea,
347 where it is as large as 0.06 according to MODIS-Terra and 0.05 according to MODIS-
348 Aqua, the north-eastern Atlantic Ocean (0.04 and 0.05 for Terra and Aqua,
349 respectively) and the seas of North Europe (0.05 for both Terra and Aqua). In these
350 regions, there is a tendency for smaller values during summer. More specifically, in
351 the Black Sea the smallest g_{aer} value (0.64) is observed in June, over the seas of North
352 Europe in July and over the north-eastern Atlantic Ocean in August. In these regions,
353 the largest values appear in the cold period of the year. Reverse seasonality with a
354 large seasonal amplitude is observed over the Persian Gulf, where the variability is as
355 large as 0.08, according to both MODIS-Terra and Aqua. The seasonal cycle of g_{aer}
356 over the Middle East exhibits a smaller range of variability (0.02 for MODIS-Terra
357 and 0.03 for Aqua) along with a reverse seasonal variation, with maximum values in
358 summer and minimum in winter. In the other two sub-regions (Mediterranean and
359 eastern Atlantic Ocean) the annual range of values is small (< 0.02). It is noteworthy
360 that in the Mediterranean Sea, there is a weak tendency of appearance of double
361 maxima in winter and spring. The spring maximum should be associated with the
362 presence of desert dust particles, which are transported from Sahara, mainly in the
363 eastern Mediterranean in this season (e.g. Fotiadi et al., 2006; Kalivitis et al., 2007;
364 Papadimas et al. 2008, Gkikas et al. 2009; Hatzianastassiou et al., 2009; Gkikas et al.,
365 2013). There is also a similar transport of Saharan dust in the central and western

366 Mediterranean during summer and autumn (e.g. Gkikas et al., 2009; 2013), but then
367 the predominance is not so clear because of the co-existence of fine anthropogenic
368 aerosols. Regardless of the annual cycle, smaller g_{aer} values are clearly distinguished
369 over the Black Sea and North Europe seas throughout the whole year.

370 At 660 nm, the g_{aer} values are lower than at 470 nm, in particular over Black Sea,
371 North Europe and North-East Atlantic, whereas the intra-annual variability (range of
372 g_{aer} values) increases up to 0.10 (Terra) and 0.08 (Aqua) over the Black Sea. This
373 increase is mainly attributed to the reduction of summer values due to the strong
374 appearance of fine aerosols in this season. Also, at 660 nm, there is a clearer double
375 annual variation of g_{aer} over the Mediterranean Sea than at 470 nm. At 870 nm the
376 general picture is similar to that of 660 nm though a further increase of month by
377 month variability is noticeable.

378 In general, our results indicate that over the regions characterized by a strong presence
379 of desert dust particles (eastern Atlantic and the Middle East and Mediterranean Seas)
380 the annual range of variability of g_{aer} is smaller than in the other regions. An
381 additional feature above regions with desert dust is the smaller decrease of g_{aer} values
382 with increasing wavelengths. This is attributed to the lower g_{aer} spectral dependence
383 of coarse compared to fine particles (e.g. Dubovik et al, 2002; J. Bi et al., 2011).

384 We should note that the MODIS-Terra and Aqua g_{aer} seasonal cycles are about similar
385 but with generally greater larger Terra than Aqua values.

386 **3.2.2 Inter-annual variability and changes**

387 Figure 5 displays the geographical distribution of the slope of inter-annual trend of
388 g_{aer} over the study region, as computed from the application of the Mann-Kendall test
389 to time series of deseasonalized monthly anomalies of g_{aer} at 470 nm. Results are
390 shown in units decade⁻¹ for both Terra and Aqua over their common time period,
391 namely 2002 – 2010, only if the trend is statistically significant at the 95% confidence
392 level. We also performed the same analysis for the 660 and 870 nm (not shown), with
393 similar results to the 470 nm wavelength.

394 In general, the estimated changes are relatively small. Terra produces widely
395 statistically significant positive trends, showing that during the period of interest, the
396 asymmetry parameter increased over the examined area, with very few exceptions.
397 The results from Aqua are statistically significant at considerably fewer cells, but also
398 give a few points with decreasing g_{aer} . Based on Terra data, the stronger increases are

399 observed in the eastern and southern Black Sea, as well as over the Baltic and Barents
400 Seas. According to MODIS-Aqua, negative trends are found over few Atlantic Ocean
401 cells. Both Aqua and Terra report increases of g_{aer} over the Persian Gulf, the Red Sea,
402 South Black Sea, East Mediterranean, the coast of the Iberian Peninsula, and some
403 coastal areas of West Africa. The differences encountered between the Terra and
404 Aqua g_{aer} trends may be attributed to the different time of passage of each satellite
405 platform carrying the same MODIS instrument, given that everything else is the same.
406 Nevertheless, they may more probably be the result of calibration differences between
407 the two MODIS sensors. It is known that there is a degradation of MODIS sensor
408 (Levy et al., 2010; Lyapustin et al., 2014) impacting time series of MODIS products.
409 More specifically, it is also known that Terra suffers more than Aqua from optical
410 sensor degradation. These calibration issues are known to affect MODIS AOD
411 retrievals, producing an offset between Terra and Aqua, and they are also expected to
412 affect aerosol asymmetry parameter, which is probably more sensitive to such
413 calibration uncertainties than AOD. In this sense, the results of Fig. 5 shown here are
414 not to be taken as truth but rather they are given as a diagnostic of a problematic
415 situation with MODIS aerosol asymmetry parameter inter-annual changes. Such
416 calibration issues are expected to be addressed, at least partly, in the new Collection
417 006 products. Nevertheless, a preliminary comparison between MODIS Aqua C051
418 and C006 Ångström exponent (AE), which is another common aerosol parameter
419 strongly dependent on size, using data for the 550-865 pair of wavelengths spanning
420 the period 2002-2010, does not reveal significant modifications in geographical
421 patterns of AE inter-annual changes. This puts some confidence on the C051 g_{aer}
422 results given in the present study. The results of this analysis are presented in detail in
423 the next sub-section (3.2.3).

424 The overall g_{aer} changes of Fig. 5 may hide smaller timescale variations of g_{aer} , which
425 are obtained by the time-series shown in Fig. 6. Results are given for the 7 sub-
426 regions defined previously, at the three different wavelengths and for Terra and Aqua
427 separately. A general pattern is the decrease of g_{aer} values with increasing wavelength,
428 in particular from 470 to 660 nm. The largest month to month and year to year
429 variation is for Black Sea (Fig. 6i). Relatively large variability is also found in the
430 sub-regions of NE Atlantic (6v), North Europe (6vi) and the Persian Gulf (6vii). On
431 the contrary, small variability is noticed in the eastern Atlantic, where systematic dust
432 outflows from Sahara take place leading to consistently high values of g_{aer} . There are
433 also some other interesting patterns, like the significant drop of g_{aer} with wavelength

434 in areas characterized by the presence of fine aerosols, namely the Black Sea, North
 435 Europe and the Persian Gulf (Figs, 6i,vi,vii, respectively). The specific patterns of
 436 inter-annual changes of g_{aer} are suggested by both Terra and Aqua, though a slight
 437 overestimation by Terra is again apparent in this figure. The obtained results of our
 438 analysis are meaningful and in accordance with the theory, underlining the ability of
 439 satellite observations to reasonably capture the g_{aer} regime over the studied regions.

440 **3.2.3 Possible uncertainties of MODIS aerosol asymmetry parameter**

441 The MODIS aerosol asymmetry parameter is not a direct retrieval product of the
 442 MODIS retrieval algorithm, but it is rather a derived by-product. Since this parameter
 443 is dependent on aerosol modes used and relative weights, it is understood that there
 444 can be uncertainties associated with it. Therefore questions may arise about the
 445 validity of g_{aer} and their spatial and temporal patterns presented in the previous sub-
 446 sections. Given that, as already mentioned, it is an aerosol optical parameter that is
 447 valuable and highly required by radiative transfer and climate models, it is worth
 448 assessing it through comparison against another more common aerosol size parameter,
 449 namely the C051 MODIS Ångström exponent at the 550-865 nm wavelength pair
 450 ($AE_{550-865}$) over ocean, which is an evaluated MODIS aerosol size product (Levy et
 451 al., 2010) that is extensively used in literature. Figure 7a, displays the geographical
 452 distribution of long-term average AE for the whole study period, i.e. 2002-2010. In
 453 this figure, the northernmost areas are blank with respect to g_{aer} (Fig. 7a) because
 454 there are no data during winter and a long-term average would be biased. The main
 455 geographical patterns in Fig.7a are in line with those of asymmetry parameter (Fig. 2).
 456 For example, note the high AE values in the Black Sea (between about 1.3 and 1.8,
 457 yellowish-reddish colors), indicative of fine aerosols, the relatively high values in the
 458 Mediterranean Sea (between about 0.7 and 1.2, greenish-yellowish colors) and the
 459 low values (0.1-0.4, deep bluish colors) off the western African coasts corresponding
 460 to exported Saharan dust. Over the same areas, g_{aer} takes inverse low and high values,
 461 for example smaller than 0.65 over the Black Sea and larger than 0.7-0.75 off the
 462 western African coasts (Figs 2ii-b and 2iii-b), indicating the predominance of fine and
 463 coarse aerosols respectively, in accordance with AE. The consistency between g_{aer} and
 464 AE data is shown by the strong anti-correlation between the MODIS $AE_{550-865}$ and g_{aer}
 465 data at 660 and 870 nm, shown in Figures 7b and 7c, respectively. It should be noted
 466 that correlation coefficients are computed from any available data pairs, i.e. available
 467 data for both g_{aer} and $AE_{550-865}$ at a given pixel and day. Note that there are no blank

468 areas in Figs 7b and 7c, opposite to Fig. 7a, because as there are both AE and g_{aer} data
469 for all but winter months, correlation coefficients can be calculated for these regions
470 (Figs. 7b, 7c). Strong negative correlation coefficients, larger than 0.7 and 0.8 in Figs
471 7b and 7c, respectively, relate inversely high g_{aer} values with low AE ones and vice-
472 versa, over the same areas. In both cases (Figs 7b and 7c), the correlation is slightly
473 higher over sea areas characterized by the presence of fine aerosols (e.g. Black Sea or
474 Persian Gulf) and lower over seas undergoing frequent transport of coarse dust
475 particles (e.g. southern Mediterranean Sea, Arabian Sea or Atlantic Ocean off the
476 western African coasts). The overall computed correlation coefficient between g_{aer} and
477 AE is equal to -0.95 over the Black Sea, -0.89 over the Mediterranean Sea, -0.87 and -
478 0.94 over the Arabian Sea and Persian Gulf, respectively and -0.89 off the western
479 African coasts (values given for $\text{AE}_{550-865}$ and g_{aer} data at 870 nm). These results
480 indicate that the spatial patterns of MODIS C051 g_{aer} product are reasonable as
481 compared to the C051 Ångström exponent data. This shows that the use of g_{aer} in
482 modeling studies can be considered as reasonably reliable with regards to the
483 consideration of fine and coarse aerosols over the examined study area, with slightly
484 more confidence over areas characterized by the presence of fine particles, such as the
485 Black Sea or Persian Gulf.

486 Since questions may also arise about possible uncertainties regarding the long-term
487 variability of MODIS C051 aerosol size products, due to the calibration issues
488 discussed in the previous section, the corresponding MODIS C006 AE product is
489 displayed in Fig. 8a. Figs. 8a and 7a are similar in the main geographical patterns of
490 the two collections' AE product. The similarity between C051 and C006 AE data is
491 also depicted in the computed correlation coefficients (Fig. 8b), exceeding 0.8, and
492 biases (in absolute and relative percentage terms, Figs 8c and 8d, respectively). For
493 the Mediterranean Sea, the Arabian Sea and Persian Gulf, biases are smaller than 0.1
494 or 10% in most areas and 0.2 or 20% almost everywhere. Relative biases larger than
495 30% are only observed over the open Atlantic Ocean). The overall computed
496 correlation coefficient for the entire study region is 0.88 (0.86, 0.89, 0.95 and 0.84 for
497 Mediterranean, Arabian, Persian and Atlantic sea surfaces off the western African
498 coasts). The corresponding overall relative percent bias is equal to 15.6% (9.1, 6.7,
499 6.1 and 15.7 for the same sub-areas as above). Our results indicate that the uncertainty
500 related to the use of C051 AE data is small, especially over the Mediterranean Sea, the
501 Arabian Sea, the Persian Gulf and the Atlantic Ocean areas not far from the European,
502 African and Asian coastlines. Our AE results are in line with those of Levy et al.

503 (2013, Fig. 15) which refer, however, only to year 2008 (ours are for 2002-2010). In
504 addition, a comparison is attempted in Figs 8e and 8f between the computed trends of
505 C051 and C006 AE data over the common period 2002-2010, in order to assess
506 whether changes are detected, which could be an indication of possible changes in
507 corresponding asymmetry parameter trends. Figures 8e and 8f show the computed
508 deseasonalized trends of slope values for both C051 and C006 AE. The results reveal
509 similar patterns between C051 and C006. Small trends are found in both of them, in
510 agreement with the small trends of asymmetry parameter reported in Fig. 5. We find
511 that the sign of AE trends basically does not change from C051 to C006. This might
512 be a signal that no changes of aerosol asymmetry parameter are expected in C006 and
513 puts confidence on the C051 results given in the present study.

514

515 **4 Evaluation against AERONET data**

516 In this section we compare the satellite-measured aerosol asymmetry parameter with
517 measurements from the global network of surface stations of AERONET, which is
518 considered as the reference dataset (Holben et al., 1998). For this purpose, we
519 identified the AERONET stations inside our area of interest and finally utilised only
520 the coastal ones, so that both satellite and surface data be available. The total number
521 of these stations is 69, and their locations are shown in Fig. 1 (open and full circles).

522 Table 1 contains the comparison statistical metrics for all wavelengths (Pearson
523 correlation coefficient, bias, root mean square error (RMSE), slope, intercept) of the
524 comparison between surface daily mean data from AERONET and satellite data from
525 MODIS-Terra and MODIS-Aqua, which correspond to the $1^\circ \times 1^\circ$ cell wherein each
526 station is located. For this analysis, we use all cells and days with common data
527 between Terra-AERONET and Aqua-AERONET. The mean differences are
528 calculated as $g_{\text{aer}}(\text{AERONET}) - g_{\text{aer}}(\text{Aqua})$ and $g_{\text{aer}}(\text{AERONET}) - g_{\text{aer}}(\text{Terra})$.

529 In general, we may note that on an annual level, the MODIS-Terra and Aqua
530 asymmetry parameter values at 470 nm are not in very good agreement with the
531 respective data from AERONET at 440 nm, while the results at the largest
532 wavelengths are more reassuring, though not being very satisfactory (increasing R and
533 decreasing relative bias and RMSE values at 675/660 nm and 870 nm). At 870 nm
534 (Table 1 and Fig. 9), correlation coefficients are found to be the largest and equal to
535 0.47 (AERONET-Terra) and 0.46 (AERONET-Aqua), while satellite data are slightly

536 overestimated compared to the surface data (bias -0.035 or 5.54% and -0.015 or -
537 2.43%, respectively).

538 It is important to note that the agreement of satellite and surface data is better in
539 spring and summer, for all studied wavelengths. Specifically, in case of MODIS-Aqua
540 g_{aer} , the correlation coefficients increase up to 0.35, 0.50 and 0.54 at 440/470 nm,
541 660/675 nm and 870 nm, respectively, while the bias decreases down to 0.0005
542 (0.07%), 0.003 (0.46%) and 0.007 (1.11%), respectively.

543 Moreover, we find that for all seasons g_{aer} values at 870 nm and 660 nm, both from
544 MODIS-Terra and MODIS-Aqua, are overestimated compared to g_{aer} (AERONET) at the
545 corresponding wavelengths (stronger overestimation at 870 nm and by Terra). Finally
546 we note an underestimation of g_{aer} at 470 nm from MODIS-Aqua, relative to the data
547 by AERONET at 440 nm, while very small biases (<0.5 %) are found between Terra
548 and AERONET at the same wavelengths.

549 In Fig. 9 we present a scatterplot comparison between MODIS and AERONET g_{aer}
550 data pairs. There is bias towards larger g_{aer} values from both Aqua and Terra
551 compared to AERONET, with Terra overpredicting more than Aqua. The root mean
552 square error to the fit between MODIS and AERONET is very similar between Aqua
553 and Terra. There are concerns on the application of ordinary least squares regression,
554 arising from the assumption that as the assigned independent variable, AERONET
555 values should be free from error. We cannot guarantee the validity of this assumption,
556 so we recognize that the reported R and slope values from Fig. 9 and Table 1, if
557 viewed as metrics of agreement between MODIS g_{aer} and real g , may be subject to the
558 effect of regression dilution and consequently biased low. This possible bias for R and
559 slope could be neglected only if AERONET errors can also be considered negligible.
560 With the above caveat in mind, the applied least-squares fit line to the scatterplot
561 comparison between matched MODIS-AERONET data pairs (Fig. 9) indicates that
562 MODIS overestimates g_{aer} more in the smaller than larger values, i.e. more for fine
563 than coarse particles.

564 We present the frequency distributions of asymmetry parameter daily values (Fig. 10)
565 on the days when data from all three databases (MODIS-Terra, MODIS-Aqua and
566 AERONET) were provided. Fig. 10a corresponds to the whole area of interest, while
567 Figs. 10b and c correspond to two broad sub-regions with basic differences in the
568 aerosol source, namely Europe with great anthropogenic sources, and Africa, Middle
569 East and Arabian peninsula, with predominant natural sources and mainly desert dust.

570 There is an apparent skew in the MODIS-Terra and MODIS-Aqua g_{aer} distributions,
571 while the AERONET distributions are more symmetrical. Moreover, the satellite data
572 distributions show larger values and smaller standard deviations compared to
573 AERONET, with the Terra overestimation being more exaggerated. The disagreement
574 is more pronounced in the sub-region of Europe, while in the sub-region of North
575 Africa / Arabian peninsula, the distributions of satellite and surface data agree more
576 thus confirming the finding of Fig. 9 based on the slope of applied linear regression
577 fit. Values over Europe are generally smaller than over North Africa / Arabian
578 peninsula (Fig. 3), which can be attributed to the presence of larger size particles of
579 desert origin in the latter sub-region, in contrast to Europe, where due to industrial
580 activity and frequent biomass burning the presence of smaller size particles is
581 important. Therefore, the smaller g_{aer} values (<0.6) in the frequency distributions of
582 the whole area, are overwhelmingly contributed by the European sub-region,
583 contrasting with larger values (0.7-0.75) being contributed by both sub-regions and
584 even more by N. Africa/Arabian peninsula at larger g_{aer} .

585

586 The overall comparison between satellite and surface g_{aer} data performed in the
587 scatterplot of Fig. 9 and Table 1 does not allow one to have an insight to how the
588 comparison behaves spatially, namely how it differs from one region to another. This
589 is addressed in Fig. 11, showing the comparison of satellite and surface data at the
590 wavelength of 870 nm separately between MODIS-Terra - AERONET and MODIS-
591 Aqua – AERONET. For this comparison, we selected AERONET stations for which
592 there is satisfactory overlap between the time series from AERONET and the time
593 series from MODIS, namely the number of common days between AERONET-Terra
594 and AERONET-Aqua is larger than 100. This criterion is satisfied by 36 stations for
595 AERONET-Terra and by 34 for AERONET-Aqua shown in Fig. 11. For each
596 AERONET station we compute the Pearson correlation coefficient between the
597 station data and the corresponding MODIS-Terra or Aqua data at 870 nm, for the
598 $1^{\circ} \times 1^{\circ}$ cell containing the station. Moreover, there is the information if the trends
599 between AERONET and either MODIS-Terra or Aqua have the same sign (blue
600 color) or not (red color).

601 In the case of the $g_{aer}(\text{AERONET}) - g_{aer}(\text{Terra})$ comparison, at 5 stations, (i.e. in 14% of
602 total 36 stations), the correlation coefficient R is larger than 0.5 (largest R found is
603 0.64 at station “Bahrain”), while at 13 stations (36 %) and 26 stations (72%) R is

604 larger than 0.4 and 0.3, respectively.. With respect to the agreement on the sign of the
605 trends, at 24 out of 36 stations (67%) there is a trend sign match and at 12 stations
606 (33%) a mismatch. Nevertheless, it should be noted that no systematic spatial
607 behaviour, i.e. homogeneous spatial patterns, is found concerning the performance of
608 MODIS-Terra g_{aer} against AERONET in terms of either the magnitude of correlation
609 or the agreement of trends between the satellite and ground datasets. A similar picture
610 emerges for the comparison $g_{aer} (AERONET) - g_{aer} (Aqua)$. In this case, there are again 5
611 stations (15% of total 34 stations) with $R > 0.5$ (maximum value $R = 0.61$ again at
612 “Bahrain”), while at 13 stations (38%) and 24 stations (71%) R is larger than 0.4 and
613 0.3, respectively. Also, we see that at 22 stations (65%) there is a trend sign match
614 and at 12 (35%) there is a mismatch.

615

616 **5 Summary and Conclusions**

617 Using satellite data from collection (051) of MODIS-Terra and Aqua data, we
618 examine the spatiotemporal variations of the aerosol asymmetry parameter (g_{aer}) over
619 North Africa, the Arabian peninsula and Europe. To our knowledge, this is the first
620 time that a satellite (MODIS) based dataset of g_{aer} , assessed and evaluated (against
621 AERONET data), is used for the study region. This is important, since such an
622 evaluated satellite dataset is very useful for many applications, like radiative transfer
623 and climate modelling as well as for remote sensing. The advantages of MODIS g_{aer}
624 data are that:

- 625 (i) They ensure complete spatial coverage over sea surfaces surrounding Europe,
626 Mediterranean and Middle East, which is essential for investigating and
627 understanding physical processes related to aerosols. These processes are
628 strongly dependent on the aerosol radiative and optical properties, g_{aer} being one
629 of the three key ones (the other two being aerosol optical thickness and single
630 scattering albedo). Such a complete spatial coverage is especially required by
631 radiative transfer and climate models.
- 632 (ii) They provide with spectral g_{aer} values, at 7 wavelengths from 470 to 2130 nm,
633 which are of essential importance for radiative transfer models. Such spectrally
634 resolved aerosol optical properties can induce significant differences in model
635 computations of aerosol radiative effects (Hatzianastassiou et al., 2007).

- 636 (iii) They provide a relatively long temporal coverage, i.e. 8 years, which is
637 significant for examining seasonal and inter-annual cycles and changes of this
638 aerosol optical property, especially combined with the complete spatial coverage.
639 This is also important since it provides a reasonable statistical bed for attempting
640 evaluations through comparison against other g_{aer} data like the AERONET.
- 641 (iv) They constitute the first to know so far satellite based g_{aer} dataset; until now, the
642 utilized g_{aer} data in modelling or other analyses were taken from in-situ
643 measurements or aerosol models, which both have their own deficiencies, namely
644 limited spatial coverage or pure theoretical basis.

645 According to the obtained results, generally, the largest values of the asymmetry
646 parameter, indicating the strongest forward scattering of radiation by atmospheric
647 aerosols, are found over areas with aerosol load being dominated by large size
648 particles of desert dust (tropical Atlantic, Arabian and Red Seas),. On the contrary,
649 smaller g_{aer} values are seen where a significant fraction of aerosol load comes from
650 small size particles of anthropogenic origin, e.g. over the Black Sea. The results are
651 consistent with the theory and thus prove a good performance of the MODIS retrieval
652 of aerosol asymmetry parameter. Depending on the area of interest, the seasonal cycle
653 of the asymmetry parameter varies markedly. More specifically, in areas with
654 abundance of desert dust particles, the range of intra-annual variation is small, with
655 the largest values during summer, while in other areas the seasonality is reversed, with
656 the largest values during the cold season and the smallest during the warm season.
657 The asymmetry parameter decreases with wavelength, especially when one examines
658 its spatially minimum values, while this decrease is weaker for the larger g_{aer} values,
659 corresponding to the presence of coarser particles.

660 The seasonal fluctuation is more pronounced with increasing wavelength in the
661 examined regions, which is attributed to the different spectral behaviour of the
662 asymmetry parameter for small and large particles. With respect to the inter-annual
663 variability of the asymmetry parameter, we did not discern very important either
664 increasing or decreasing tendencies, with absolute changes smaller than 0.04 in any
665 case. On the other hand, we found opposing tendencies for the two satellite datasets.
666 MODIS-Terra observes mostly increasing tendencies, while Aqua gives also a few
667 regions with decreasing tendencies. Generally, the largest intra-annual and inter-
668 annual variations are seen over the Black Sea, while the smallest over the tropical
669 Atlantic. However, some strong trends (especially from Terra) may be due to

670 calibration drift errors, which may be addressed in collection 006. Along these lines
671 we performed some preliminary comparisons between 051 and 006 Ångström
672 Exponent trends from Aqua, which ensured that AE and g_{aer} are very closely anti-
673 correlated. These preliminary results, show that 051 Aqua AE trends resemble very
674 closely the 006 trends, supporting that the g_{aer} trends from collection 051 (at least for
675 Aqua) reported in this study are credible.

676 The 051 MODIS g_{aer} data is not a retrieved but a derived MODIS parameter. Given
677 that the retrieval is strongly dependent on the assumptions made, namely on the
678 aerosol modes used, uncertainties can be associated with its use in radiative transfer
679 modeling. In order to examine these uncertainties, the g_{aer} data were compared with
680 051 AE data for the same period. The results from the comparison showed a strong
681 anti-correlation (coefficient higher than 0.7-0.8) proving the consistency and
682 reasonably safe use of g_{aer} data in modeling studies, at least to the same degree with
683 MODIS AE data in modeling and other analyses. The correlation is even higher over
684 sea areas characterized by stronger presence of fine aerosols, like the Black Sea, the
685 Persian Gulf or the North Sea. This confidence is further strengthened by the small
686 identified uncertainties related with the use of collection 051 instead of 006 MODIS
687 g_{aer} data reported in the previous paragraph. This was obtained indirectly based on the
688 use of AE data of both collections since g_{aer} data are not yet available in collection
689 006.

690 We compare satellite data with surface data from the AERONET, in order to further
691 validate the reliability of the former. Through the examination of frequency
692 distributions of daily g_{aer} , a shift of satellite data towards larger values relative to
693 surface data becomes apparent. This finding is more pronounced for g_{aer} over Europe,
694 while the North African, Arabian peninsula values are more in agreement. Moreover,
695 the smallest g_{aer} values originate from particles from Europe, because of the
696 generation of smaller size particles by industrial activities and biomass burning.

697 We present scatter plots of daily g_{aer} values between MODIS-Terra, MODIS-Aqua,
698 and AERONET, which show moderate agreement between satellite data at 470 nm
699 and surface data at 440 nm, with small correlation coefficients ($R < 0.3$) and a slight
700 underestimation by MODIS. Slightly better agreement was noted at larger
701 wavelengths, but still without reaching very satisfactory levels ($R < 0.47$).
702 Nevertheless, during spring and summer, satellite and surface measurements tend to
703 agree more. Finally, for the comparisons at 660/675 and 870 nm, we report an
704 overestimation of g_{aer} by MODIS compared to AERONET,.

705 When examined at the local scale, i.e. station by station, the MODIS g_{aer} data agree
706 reasonably and for some stations better than in overall, but still not very well, with
707 those of AERONET. This analysis, based on 36 and 34 AERONET stations ensuring
708 at least 100 common days with MODIS-Terra and Aqua, respectively, shows that in
709 36 and 38% of stations, respectively, the MODIS data have correlation coefficients
710 larger than 0.4 (reaching values up to 0.64), while in about 65% of stations the trends
711 of g_{aer} from MODIS and AERONET have the same sign. Nevertheless, the magnitude
712 of correlation coefficients or the agreement between trends of g_{aer} from the satellite
713 and ground datasets do not exhibit a systematic (homogeneous) spatial pattern.

714 Our results offer an interesting way to assess the uncertainty induced by the use of
715 such satellite g_{aer} data in climate and radiative transfer models that compute aerosol
716 radiative and climate effects. Based on an overall assessment of satellite MODIS g_{aer}
717 through detailed comparisons against ground AERONET data, it appears that in
718 overall MODIS performs satisfactorily in terms of magnitude of g_{aer} values. This is
719 indicated by the computed biases, which are smaller than 5% with respect to MODIS
720 values, with better performance at smaller wavelengths. The root mean squared errors
721 vary within the range 5-10% again being smaller for smaller wavelengths. These
722 results indicate an uncertainty of MODIS g_{aer} data over the study region up to of 10%
723 at maximum. Previous analyses and sensitivity studies for the same study region
724 (Papadimas et al., 2012) have shown that such g_{aer} uncertainties can induce
725 modifications of aerosol direct radiative effects (DREs) which are equal to 30% at the
726 top-of-atmosphere (TOA) and 1% in the atmosphere and 10% at the surface, at
727 maximum. Therefore, the uncertainty associated with the use of MODIS g_{aer} is larger
728 as to any aerosol related physical process taking place at TOA, namely planetary
729 cooling or warming and its magnitude, smaller for processes at the Earth's surface,
730 e.g. surface cooling and very small for aerosol processes and feedbacks in the
731 atmosphere, like the aerosol semi-direct effect and its implications. Results from the
732 same previous analysis (Papadimas et al., 2012) proved that the exact magnitude of
733 MODIS g_{aer} DRE uncertainty can be estimated by simple linear equations relating
734 DREs and g_{aer} , separately given for TOA, atmosphere and surface.

735 The results of the present analysis are useful since they assess for the first time the
736 performance of satellite based products of aerosol asymmetry parameter over broad
737 regions of special climatic interest. The obtained results are relatively satisfactory
738 given the difficulties encountered by satellite retrieval algorithms due to the different

739 assumptions they made. Nevertheless, our results and identified weaknesses remind
740 that users should be aware of the g_{aer} uncertainties and their consequences. The
741 identified weaknesses may provide an opportunity to improve such satellite retrievals
742 of aerosol asymmetry parameter in forthcoming data products like those of MODIS
743 C006. The increased temporal coverage of g_{aer} data, combined with the continued
744 operation of MODIS, is expected to make possible the building of the first real
745 satellite climatology of this important aerosol optical property.

746

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756

757 **References**

- 758 Alonso-Pérez S., Cuevas E., Querol X., Guerra J. C., and Pérez C. (2012). African dust source
759 regions for observed dust outbreaks over the Subtropical Eastern North Atlantic region,
760 above 25°N, *J. Arid Environ.*, 79, 100-109, [doi:10.1016/j.jaridenv.2011.11.013](https://doi.org/10.1016/j.jaridenv.2011.11.013).
- 761 Barnaba F. and G. P. Gobbi: Aerosol seasonal variability over the Mediterranean region and
762 relative impact of maritime, continental and Saharan dust particles over the basin from
763 MODIS data in the year 2001, *Atmospheric Chemistry and Physics*, 4, 4285 - 4337, SRef-
764 ID: 1680-7375/acpd/2004-4-4285 SRef-ID: 1680-7375/acpd/2004-4-4285, 2004.
- 765 Barnaba F., Angelini F., Curci G., and Gobbi G. P. (2011), An important fingerprint of
766 wildfires on the European aerosol load, *Atmos. Chem. Phys.*, 11, 10,487–10,501.
- 767 Bovchaliuk A., Milinevsky G., Danylevsky V., Goloub P., Dubovik O., Holdak A., and
768 Sosonkin, M. (2013). Variability of aerosol properties over Eastern Europe observed from
769 ground and satellites in the period from 2003 to 2011. *Atmos. Chem. Phys.*, 13, 6587-6602
- 770 Bréon F.-M., Vermeulen A., and Descloitres J. (2011). An evaluation of satellite aerosol
771 products against sunphotometers measurements, *Remote Sens. Environ.*, 115, 3102–3111.
- 772 Chubarova, N. Y.: Seasonal distribution of aerosol properties over Europe and their impact on
773 UV irradiance, *Atmos. Meas. Tech.*, 2, 593–608, doi:10.5194/amt-2-593-2009, 2009.
- 774 Di Iorio, T., A. di Sarra, W. Junkermann, M. Cacciani, G. Fiocco, and D. Fua, Tropospheric
775 aerosols in the Mediterranean: 1. Microphysical and optical properties, *J. Geophys. Res.*,
776 108(D10), 4316, doi:10.1029/2002JD002815, 2003
- 777 Dubovik, Oleg, Brent Holben, Thomas F. Eck, Alexander Smirnov, Yoram J. Kaufman,
778 Michael D. King, Didier Tanré, Ilya Slutsker, 2002: Variability of Absorption and Optical
779 Properties of Key Aerosol Types Observed in Worldwide Locations. *J. Atmos. Sci.*, **59**,
780 590–608.
- 781 Dubovik, O. and M. D. King, 2000: A flexible inversion algorithm for retrieval of aerosol
782 optical properties from Sun and sky radiance measurements," *J. Geophys. Res.*, 105, 20
783 673-20 696.
- 784 Engelstaedter S., Tegen I., and Washington R. (2006). North African dust emissions and
785 transport, *Earth-Sci. Revi.*, 79(1-2), 73-100.
- 786 Fotiadi, A., E. Drakakis, N. Hatzianastassiou, C. Matsoukas, K. G. Pavlakis, D.
787 Hatzidimitriou, E. Gerasopoulos, N. Mihalopoulos, and I. Vardavas (2006), Aerosol
788 physical and optical properties in the eastern Mediterranean Basin, Crete, from Aerosol
789 Robotic Network data, *Atmos. Chem. Phys.*, 6, 5399– 5413.
- 790 Gerasopoulos, E., Kouvarakis, G., Babasakalis, P., Vrekoussis, M., Putaud, J. P., and
791 Mihalopoulos, N.: Origin and variability of particulate matter (PM10) mass concentrations
792 over the Eastern Mediterranean, *Atmos. Environ.*, 40, 4679–4690, 2006.

- 793 Ginoux P, Prospero J. M., Gill T. E., Hsu C. N., Zhao M., (2012). Global-scale attribution of
794 anthropogenic and natural dust sources and their emission rates based on MODIS Deep
795 Blue aerosol products. *Rev. Geophys.* 50, RG3005. doi:10.1029/2012RG000388.
- 796 Gkikas, A., Hatzianastassiou, N., and Mihalopoulos, N.: Study and characterization of aerosol
797 episodes in the Mediterranean basin for the 7-year period 2000–2007 based on MODIS
798 data, European Aerosol Conference, Greece, Thessaloniki, 24–29 August 2008.
- 799 Gkikas, A., Hatzianastassiou, N., and Mihalopoulos, N.: Aerosol events in the broader
800 Mediterranean basin based on 7-year (2000–2007) MODIS C005 data, *Ann. Geophys.*, 27,
801 3509–3522, doi:10.5194/angeo-27-3509-2009, 2009.
- 802 Gkikas, A., Houssos, E., Hatzianastassiou, N., Papadimas, C. and Bartzokas, A. (2011),
803 Synoptic conditions favouring the occurrence of aerosol episodes over the broader
804 Mediterranean basin. *Q.J.R. Meteorol. Soc.* doi: 10.1002/qj.978.
- 805 Gkikas, A., Hatzianastassiou, N., Mihalopoulos, N., Katsoulis, V., Kazadzis, S., Pey, J., and
806 Torres, O. (2013). The regime of intense desert dust episodes in the Mediterranean based
807 on contemporary satellite observations and ground measurements. *Atmos. Chem. Phys.*,
808 13, 12135-12154.
- 809 Gkikas A., Houssos E. E., Lolis C. J., Bartzokas A., Mihalopoulos N., and Hatzianastassiou
810 N. (2014), Atmospheric circulation evolution related to desert-dust episodes over the
811 Mediterranean. *Quart. J. Roy. Meteorol. Soc.*, 690, 1634-1645.
- 812 Goloub, P., and O. Arino, 2000: Verification of the consistency of POLDER aerosol index
813 over land with ATSR-2 fire product. *Geophys. Res. Lett.*, 27, 899–902.
- 814 Graßl, H.: Possible changes of planetary albedo due to aerosol particles, in *Man's Impact on*
815 *Climate*, edited by: W. Bach, J. Pankrath, and W. Kellogg, Elsevier, New York, 1979.
- 816 Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, *J. Geophys.*
817 *Res.*, 102, 6831–6864, 1997.
- 818 Hatzianastassiou, N., B. Katsoulis, I. Vardavas: Sensitivity analysis of aerosol direct radiative
819 forcing in ultraviolet - visible wavelengths and consequences for the heat budget, *Tellus*,
820 56b, 368 - 381, 2004.
- 821 Hatzianastassiou, N., A. Gkikas, N. Mihalopoulos, O. Torres, and B. D. Katsoulis: Natural
822 versus anthropogenic aerosols in the eastern Mediterranean basin derived from multiyear
823 TOMS and MODIS satellite data, *J. Geophys. Res.*, 114, D24202,
824 doi:10.1029/2009JD011982, 2009.
- 825 Hatzianastassiou, N., Matsoukas, C., Drakakis, E., Stackhouse Jr., P. W., Koepke, P.,
826 Fotiadi, A., Pavlakis, K. G., and Vardavas, I.: The direct effect of aerosols on solar
827 radiation based on satellite observations, reanalysis datasets, and spectral aerosol optical
828 properties from Global Aerosol Data Set (GADS), *Atmos. Chem. Phys.*, 7, 2585-2599,
829 doi:10.5194/acp-7-2585-2007, 2007.

- 830 Haywood, J.M., and O. Boucher, 2000: Estimates of the direct and indirect radiative forcing
831 due to tropospheric aerosols: A review. *Rev. Geophys.*, 38, 513–543.
- 832 Heishman, J. (1999), Commanding Officer, Forecaster's Handbook, U.S. Navy Cent.
833 Meteorol. and Oceanogr. Cent., Manama, Bahrain.
- 834 Holben B.N., T.F. Eck, I. Slutsker, D. Tanré, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan,
835 Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, 1998: AERONET -
836 A federated instrument network and data archive for aerosol characterization, *Rem. Sens.*
837 *Environ.*, 66, 1-16.
- 838 Ichoku C., D. Allen Chu, Shana Mattoo, Yoram J. Kaufman, Lorraine A. Remer, Didier Tanré,
839 Ilya Slutsker, and Brent N. Holben: A spatio-temporal approach for global validation and
840 analysis of MODIS aerosol products. *GEOPHYSICAL RESEARCH LETTERS*, VOL. 29,
841 NO. 12, 10.1029/2001GL013206, 2002.
- 842 IPCC, 2007: *Climate Change 2007: The Physical Science Basis*. Contribution of Working
843 Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate
844 Change [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor
845 and H.L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and
846 New York, NY, USA.
- 847 IPCC, 2013: *Climate Change 2013: The Physical Science Basis*. Contribution of Working
848 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change
849 [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y.
850 Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United
851 Kingdom and New York, NY, USA, 1535 pp.
- 852 Joseph J. H., Wiscombe W. J., and Weinman, J. A. (1976). The delta-Eddington
853 approximation for radiative flux transfer. *J. Atmos. Sci.*, 33, 2452–2459.
- 854 Jianrong Bi, Jianping Huang, Qiang Fu, XinWanga, Jinsen Shi , Wu Zhang , Zhongwei
855 Huang, Beidou Zhang: Toward characterization of the aerosol optical properties over
856 Loess Plateau of Northwestern China. *Journal of Quantitative Spectroscopy & Radiative*
857 *Transfer* 112 (2011) 346–360.
- 858 Kalapureddy, M. C. R., D. G. Kaskaoutis, P. Ernest Raj, P. C. S. Devara, H. D. Kambezidis,
859 P. G. Kosmopoulos, and P. T. Nastos (2009), Identification of aerosol type over the
860 Arabian Sea in the premonsoon season during the Integrated Campaign for Aerosols,
861 Gases and Radiation Budget (ICARB), *J. Geophys. Res.*, 114, D17203,
862 doi:10.1029/2009JD011826.
- 863 Kalivitis, N., Gerasopoulos, E., Vrekoussis, M., Kouvarakis, G., Kubilay, N.,
864 Hatzianastassiou, N., Vardavas, I., and Mihalopoulos, N.: Dust transport over the eastern
865 Mediterranean derived from TOMS, AERONET and surface measurements, *J. Geophys.*
866 *Res.*, 112, D03202, doi:10.1029/2006JD007510, 2007.

- 867 Kaufman, Y. J., D. Tanré, L. A. Remer, E. F. Vermote, A. Chu, and B. N. Holben:
868 Operational remote sensing of tropospheric aerosol over land from EOS moderate
869 resolution imaging spectroradiometer, *J. Geophys. Res.*, 102, 17,051– 17,067, 1997.
- 870 Kinne, S., D. O'Donnell, P. Stier, S. Kloster, K. Zhang, H. Schmidt, S. Rast, M. Giorgetta, T.
871 F. Eck, and B. Stevens (2013), MAC-v1: A new global aerosol climatology for climate
872 studies, *J. Adv. Model. Earth Syst.*, 5, 704–740, doi:10.1002/jame.20035.
- 873 Kishcha, P., A. M. da Silva, B. Starobinets, C. N. Long, O. Kalashnikova, and P. Alpert,
874 Meridional distribution of aerosol optical thickness over the tropical Atlantic Ocean,
875 *Atmos. Chem. Phys. Discuss.*, 14, 23309-23339, 2014.
- 876 Koepke, P., M. Hess, I. Schult, and E. P. Shettle: Global aerosol data set, Rep. No. 243, Max-
877 Planck Institut fuer Meteorologie, Hamburg, Germany, 44 pp., 1997.
- 878 Kondratyev, K. Y. 1999. Climatic effects of aerosols and clouds. Springer, New York.
- 879 Kutiel H., Furman, H., (2003). Dust storms in the Middle East: sources of origin and their
880 temporal characteristics. *Indoor Built Environ.* 12, 419–426.
- 881 Lelieveld, J., et al. (2002), Global air pollution crossroads over the Mediterranean, *Science*,
882 298, 794– 799, doi:10.1126/science.1075457.
- 883 Levy, R. C., Remer, L. A., Kleidman, R. G., Mattoo, S., Ichoku, C., Kahn, R., and Eck, T. F.:
884 Global evaluation of the Collection 5 MODIS dark-target aerosol products over land,
885 *Atmos. Chem. Phys.*, 10, 10399-10420, doi:10.5194/acp-10-10399-2010, 2010.
- 886 Lohmann U., Feichter, J. [2005] Global indirect aerosol effects: A review . *Atmos. Chem.*
887 *Phys.*, 5, 715-737.
- 888 Miller S. D., Kuciauskas A. P., Liu M., Ji Q., Reid J. S., Breed D. W. Walker A. L., and
889 Mandoos A. A.. (2008). Haboob dust storms of the southern Arabian Peninsula, *J.*
890 *Geophys. Res.*, 113, D01202, doi:[10.1029/2007JD008550](https://doi.org/10.1029/2007JD008550).
- 891 Nabat P., Somot S., Malle, M., Chiapello I., Morcrette J. J., Solmon F., Szopa S., Dulac F.,
892 Collins W., Ghan S., Horowitz L. W., Lamarque J. F., Lee Y. H., Naik V., Nagashima T.,
893 Shindell D., and Skeie R. (2013), A 4-D climatology (1979–2009) of the monthly
894 tropospheric aerosol optical depth distribution over the Mediterranean region from a
895 comparative evaluation and blending of remote sensing and model products. *Atmos. Meas.*
896 *Tech.*, 6, 1287–1314.
- 897 Lyamani, H., F. J. Olmo, A. Alcañtara, and L. Alados-Arboledas (2006), Atmospheric
898 aerosols during the 2003 heat wave in southeastern Spain. I: Spectral optical depth, *Atmos.*
899 *Environ.*, 40, 6453 – 6464, doi:10.1016/j.atmosenv.2006.04.048.
- 900 Papadimas, C. D., N. Hatzianastassiou, N. Mihalopoulos, X. Querol, and I.
901 Vardavas (2008), Spatial and temporal variability in aerosol properties over the
902 Mediterranean basin based on 6-year (2000–2006) MODIS data, *J. Geophys. Res.*, 113,
903 D11205, doi:10.1029/2007JD009189.

- 904 Papadimas C. D., Hatzianastassiou N., Matsoukas C., Kanakidou M., Mihalopoulos N., and
905 Vardavas, I. (2012). The direct effect of aerosols on solar radiation over the broader
906 Mediterranean basin. *Atmos. Chem. Phys.*, 12, 7165-7185.
- 907 Pace, G., A. di Sarra, Meloni, D., Piacentino, S., and Chamard, P.: Aerosol optical properties
908 at Lampedusa (Central Mediterranean). 1. Influence of transport and identification of
909 different aerosol types, *Atmos. Chem. Phys.*, 6, 697–713, 2006, [www.atmos-chem-](http://www.atmos-chem-phys.net/6/697/2006/)
910 [phys.net/6/697/2006/](http://www.atmos-chem-phys.net/6/697/2006/).
- 911 Pandithurai, R. T. Pinker, P. C. S. Devara, T. Takamura, K. K. Dani, Seasonal asymmetry in
912 diurnal variation of aerosol optical characteristics over Pune, western India, *J. Geophys.*
913 *Res.*, 112, D8, DOI: 10.1029/2006JD007803, 2007.
- 914 Pilinis, Ch., Pandis, S. N. and Seinfeld, J. H. 1996. Sensitivity of direct climate forcing by
915 anthropogenic aerosols to aerosol size distribution and composition. *J. Geophys. Res.* 100,
916 18 739–18 754.
- 917 Prospero, J., P. Ginoux, O. Torres, and S. E. Nicholson (2002), Environmental
918 Characterization of Global sources of atmospheric soil dust derived from the NIMBUS-7
919 TOMS absorbing aerosol product, *Rev. Geophys.*, 40(1), 1002,
920 doi:10.1029/20000GR000095.
- 921 Rahul, PRC and Salvekar, PS and Devara, PCS (2008) Aerosol optical depth variability over
922 Arabian Sea during drought and normal years of Indian monsoon. *Geophysical Research*
923 *Letters*, 35 (22).
- 924 Rashki A., Kaskaoutis D. G., de W. Rautenbach C. J., Eriksson P. G., Qiang M, and Gupta
925 P., (2012). Dust storms and their horizontal dust loading in the Sistan region, Iran. *Aeolian*
926 *Res* 5: 51–62.
- 927 Rashki A., Kaskaoutis D. G., Eriksson P. G., de W. Rautenbach C. J., Flamant C., Abdi
928 Vishkaee F., (2014). Spatio-temporal variability of dust aerosols over the Sistan region in
929 Iran based on satellite observations. *Nat. Hazards*, doi: 10.1007/s11069-013-0927-0.
- 930 Remer L.A., Tanre D., Kaufman Y.J., Levy R, and Matoo S.: Algorithm for Remote Sensing
931 of Tropospheric Aerosol from MODIS: Collection 005, 2006,
932 http://modis.gsfc.nasa.gov/data/atbd/atbd_mod02.pdf.
- 933 Remer L.A., Kaufman Y.J., Tanre D. and co-authors: The MODIS aerosol algorithm,
934 products, and validation, *J. Atmos. Sci.*, 62: 947-973, 2005.
- 935 Remer LA, Kleidman RG, Levy RC, Kaufman YJ, Tanre D, Mattoo S, Martins JV, Ichoku
936 C, Koren I, Yu H, Holben BN. 2008. Global aerosol climatology from the MODIS satellite
937 sensors. *Journal of Geophysical Research* 113: D14S07, DOI: 10.1029/2007JD009661.
- 938 Satheesh, S. K., V. Ramanathan, X. Li-Jones, J. M. Lobert, I. A. Podgorny, J. M. Prospero, B.
939 N. Holben, and N. G. Loeb, A model for the natural and anthropogenic aerosols over the
940 tropical Indian Ocean derived from Indian Ocean Experiment data, *J. Geophys. Res.*, 104,
941 27,421–27,440, 1999.

- 942 Satheesh, S. K., K. Krishna Moorthy, Y. J. Kaufman, and T. Takemura, Aerosol optical depth,
943 physical properties and radiative forcing over the Arabian Sea, *Meteorology and*
944 *Atmospheric Physics*, Volume 91, Issue 1, pp 45-62, 2006.
- 945 Savoie, D. L., J. M. Prospero, and R. T. Nees, Nitrate, nonsea-salt sulfate, and mineral
946 aerosol over the northwestern Indian Ocean, *J. Geophys. Res.*, 92, 933–942, 1987.
- 947 Sciare, J., H. Bardouki, C. Moulin, and N. Mihalopoulos (2003), Aerosol sources and their
948 contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea
949 during summertime, *Atmos. Chem. Phys.*, 3, 291–302, SRef-ID:1680 – 7324/acp/2003–3-
950 291.
- 951 Smirnov A., Holben B. N., Dubovik O., O'Neill N. T., Eck T. F., Westphal D. L., Goroch A.
952 K., Pietras C., and Slutsker I, 2002a: Atmospheric Aerosol Optical Properties in the
953 Persian Gulf. *J. Atmos. Sci.*, **59**, 620–634, doi: [http://dx.doi.org/10.1175/1520-
954 0469\(2002\)059<0620:AAOPIT>2.0.CO;2](http://dx.doi.org/10.1175/1520-0469(2002)059<0620:AAOPIT>2.0.CO;2).
- 955 Smirnov, A., B. N. Holben, Y. J. Kaufman, O. Dubovik, T. F. Eck, I. Slutsker, C. Pietras, and
956 R. N. Halthore, 2002b: Optical properties of atmospheric aerosol in maritime
957 environments. *J. Atmos. Sci.*, **59**, 501–523.
- 958 Smirnov, A., B. N. Holben, T. F. Eck, I. Slutsker, B. Chatenet, and R. T. Pinker, 2002c:
959 Diurnal variability of aerosol optical depth observed at AERONET (Aerosol Robotic
960 Network) sites, *Geophys. Res. Lett.*, **29**, 23, 2115, doi:10.1029/2002GL016305.
- 961 Tanré, D., Y. J. Kaufman, M. Herman, and S. Mattoo: Remote sensing of aerosol properties
962 over oceans using the MODIS/EOS spectral radiances, *J. Geophys. Res.*, 102, 16,971–
963 16,988, 1997.
- 964 Tindale, N. W., and P. P. Pease, Aerosols over the Arabian Sea: Atmospheric transport
965 pathways and concentrations of dust and sea salt, *Deep Sea Res.*, 46, 1577–1595, 1999.
- 966 Yu, H., Dickinson, R. E., Chin, M., Kaufman, Y. J., Holben, B. N. Geogdzhayev, I. V., and
967 Mishchenko, M. I.: Annual cycle of global distributions of aerosol optical depth from
968 integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, 108,
969 4128, doi:10.1029/2002JD002717, 2003.
- 970 Zdun, A., A. Rozwadowska and S. Kratzer, 2011. Seasonal variability in the optical
971 properties of Baltic aerosols, *Oceanologia*, 53(1), 7-34.
- 972 Zuluaga, M. D., P. J. Webster, and C. D. Hoyos (2012), Variability of aerosols in the tropical
973 Atlantic Ocean relative to African Easterly Waves and their relationship with atmospheric
974 and oceanic environments, *J. Geophys. Res.*, 117, D16207, doi:10.1029/2011JD017181.
- 975

976 **Table 1.** Correlation coefficients (R), mean bias, root mean squared error (RMSE)
 977 and the slope and intercept values of applied linear regression fits between MODIS
 978 and AERONET g_{aer} data. The statistical parameters are given separately for the pairs
 979 of wavelengths: (i) 470 nm (MODIS) and 440 nm (AERONET), (ii) 660 nm
 980 (MODIS) and 675nm (AERONET) and (iii) 870 nm (MODIS and AERONET). The
 981 statistical parameters are also given separately for winter, spring, summer and
 982 autumn. ^a

983

984 *MODIS-Terra*

985

		R	Bias*	RMSE	Slope	Intercept
year	470-440	0.25	2×10^{-4}	0.045	0.36	0.45
	660-675	0.41	-0.028	0.060	0.55	0.32
	870	0.47	-0.035	0.070	0.60	0.29
W i n t e r	470-440	0.20	4.5×10^{-4}	0.046	0.26	0.53
	660-675	0.35	-0.033	0.056	0.41	0.42
	870	0.41	-0.053	0.057	0.40	0.43
Sp r i n g	470-440	0.27	-5×10^{-4}	0.046	0.40	0.43
	660-675	0.44	-0.023	0.060	0.63	0.27
	870	0.50	-0.026	0.071	0.67	0.24
Su m m e	470-440	0.33	-0.002	0.044	0.51	0.35
	660-675	0.48	-0.031	0.061	0.71	0.22
	870	0.54	-0.030	0.077	0.79	0.16
Au t u m n	470-440	0.21	0.003	0.044	0.30	0.50
	660-675	0.33	-0.027	0.059	0.45	0.38
	870	0.41	-0.035	0.068	0.53	0.34

986

987

988 *MODIS-Aqua*

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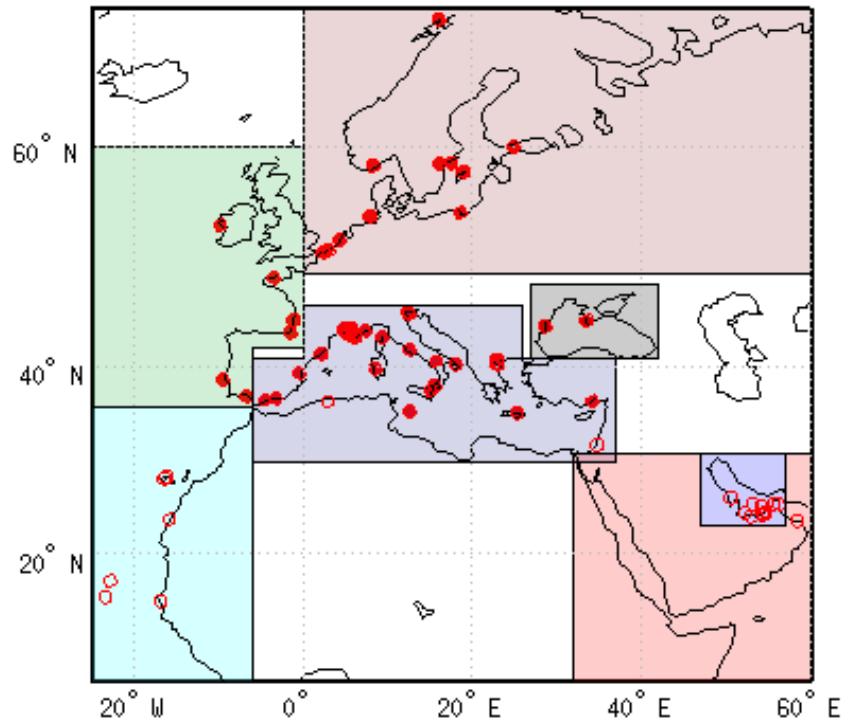
		R	Bias*	RMSE	Slope	Intercept
	470-440	0.27	0.018	0.047	0.41	0.40
	660-675	0.42	-0.005	0.062	0.61	0.26
	870	0.46	-0.015	0.072	0.61	0.26
W						

^aThe reported correlation coefficients and slopes may be biased low, because we did not include in our analysis the unknown AERONET errors.

i n t e r	470-440	0.25	0.024	0.049	0.36	0.43
	660-675	0.39	-0.001	0.062	0.55	0.30
	870	0.43	-0.021	0.068	0.51	0.33
Sp rin g	470-440	0.29	0.015	0.048	0.45	0.38
	660-675	0.45	-0.003	0.064	0.70	0.20
	870	0.50	-0.007	0.076	0.71	0.19
Su m me	470-440	0.35	0.014	0.045	0.55	0.30
	660-675	0.50	-0.012	0.060	0.72	0.19
	870	0.53	-0.018	0.074	0.73	0.19
Au t u m n	470-440	0.20	0.021	0.047	0.30	0.47
	660-675	0.32	-0.003	0.061	0.46	0.36
	870	0.37	-0.014	0.069	0.48	0.34

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991
992

* $g_{aer}(AERONET) - g_{aer}(MODIS)$



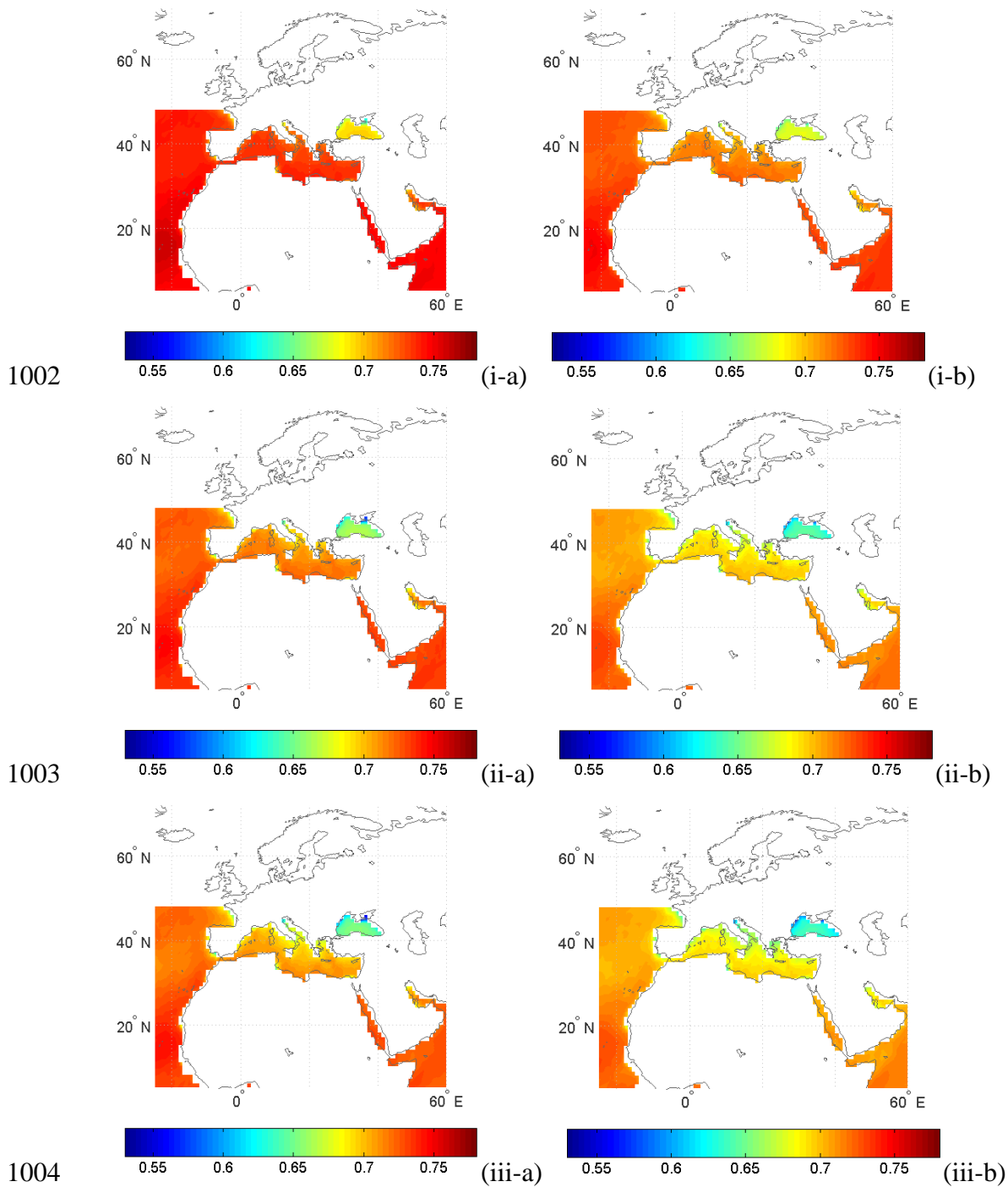
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994

995 **Figure 1.** The study region (5°N–70°N, 25°W–60°E) and the location of 69
 996 AERONET stations used for validation of MODIS satellite aerosol asymmetry
 997 parameter (g_{aer}) data. Solid red circles denote stations located in Europe and hollow
 998 red circles are stations in Africa, Middle East and the Arabian peninsula. Also shown
 999 are seven sub-regions selected for studying the seasonal variation of g_{aer} .

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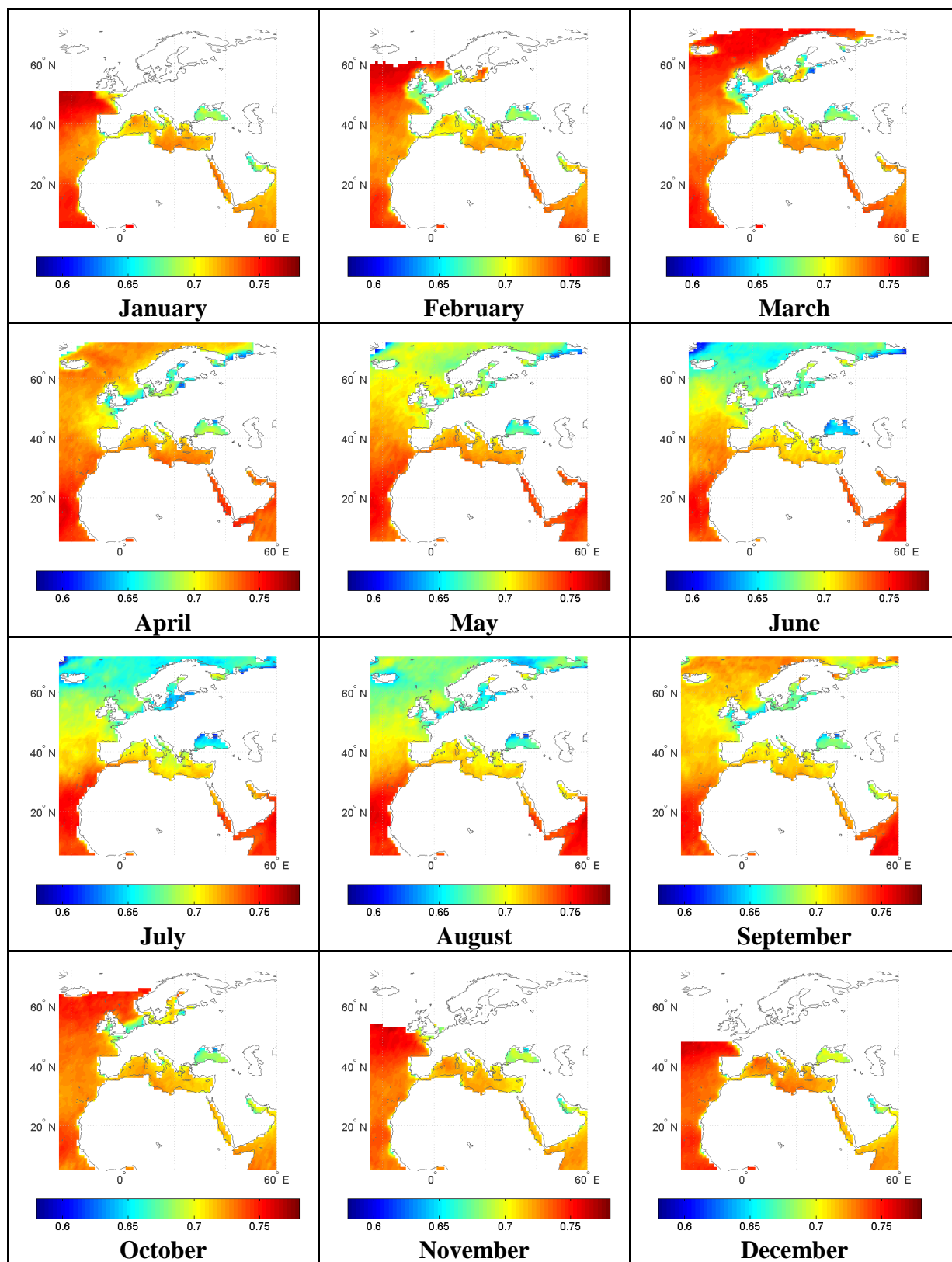
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1005

1006 **Figure 2.** Geographical distribution of MODIS-Terra (-a, left column) and MODIS-
 1007 Aqua (-b, right column) g_{aer} values averaged over 2002-2010, at the wavelengths of:
 1008 470 nm (i-, top row), 660 nm (ii-, middle row) and 870 nm (iii-, bottom row).

1009

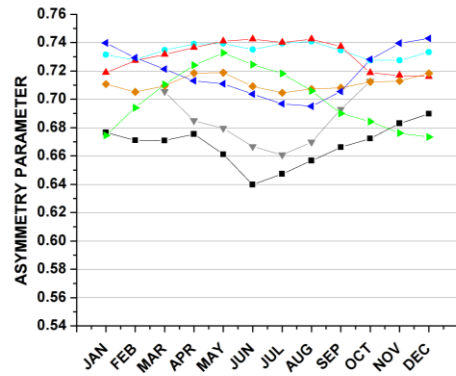
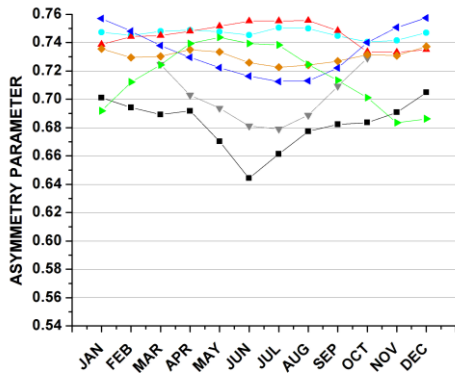


1011

1012 **Figure 3.** Month by month variation of MODIS-Aqua g_{aer} values at 470 nm averaged

1013 over the period 2002-2010.

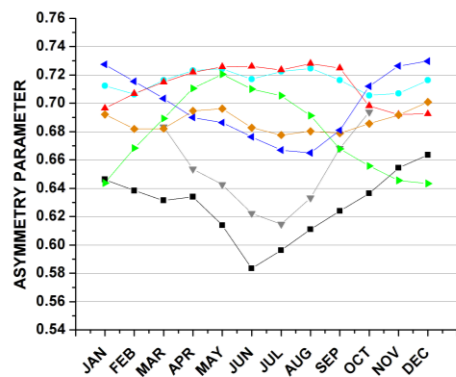
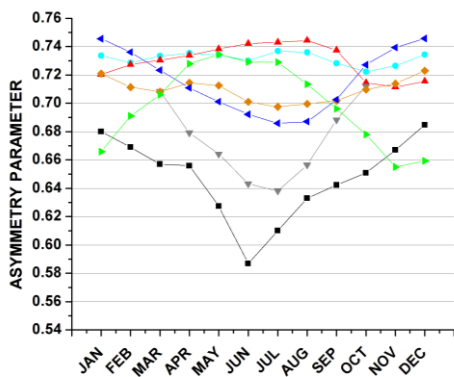
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(i-a)

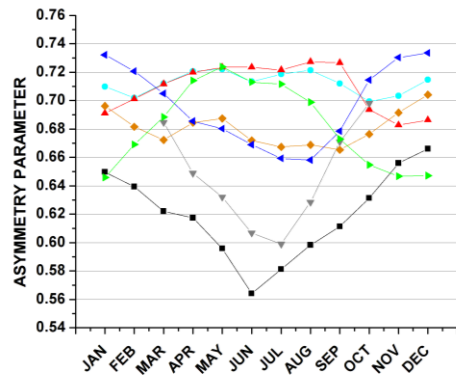
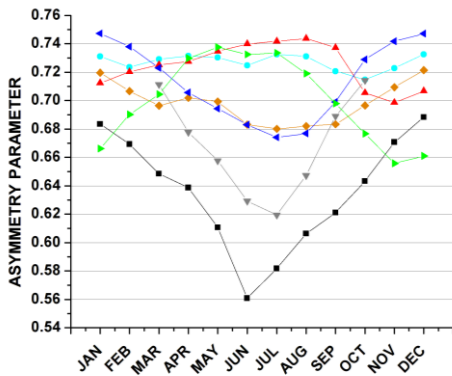
(i-b)



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(ii-a)

(ii-b)



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(iii-a)

(iii-b)



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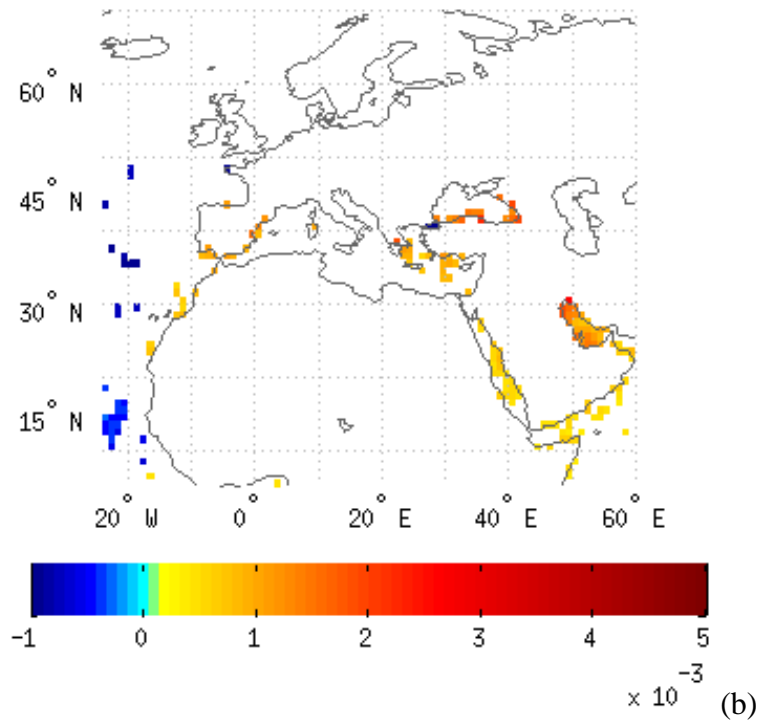
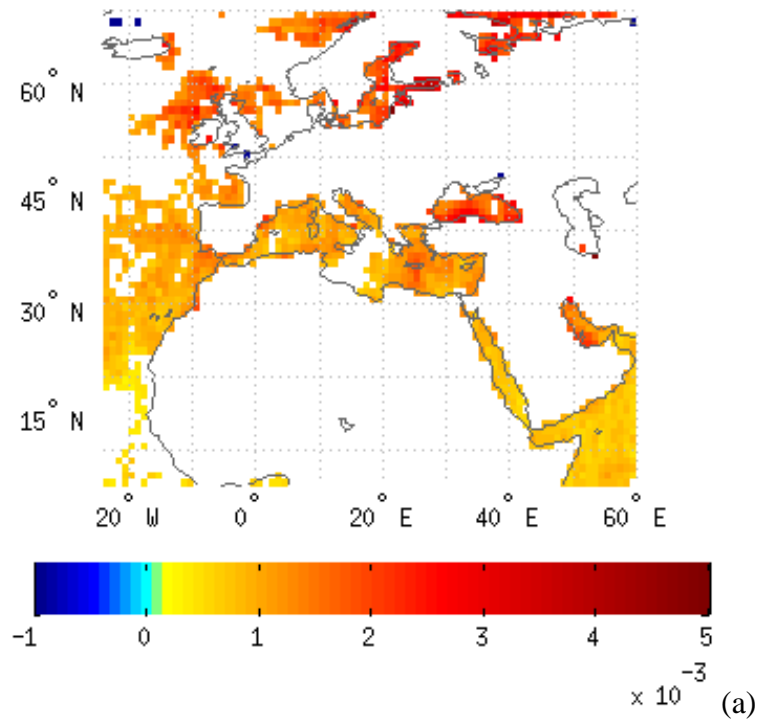
1019 **Figure 4.** Intra-annual variation of MODIS Terra (-a, left column) and Aqua (-b, right column) g_{aer} values averaged over seven selected sub-regions (Fig. 1). Results are

1020 given for g_{aer} values at: 470 nm (i-, top row), 660 nm (ii-, middle row) and 870 nm

1021 (iii-, bottom row), averaged over the period 2002-2010, respectively.

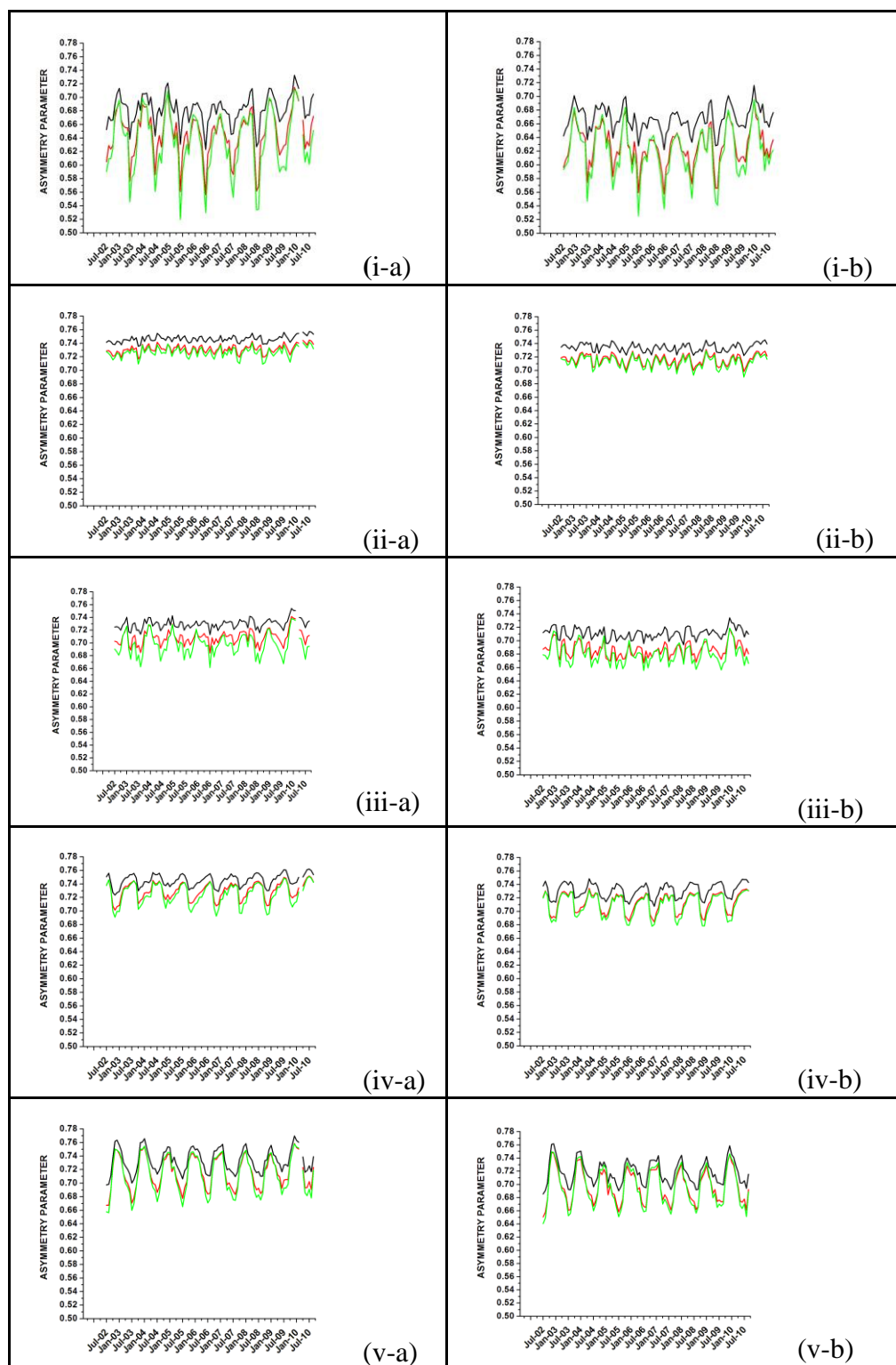
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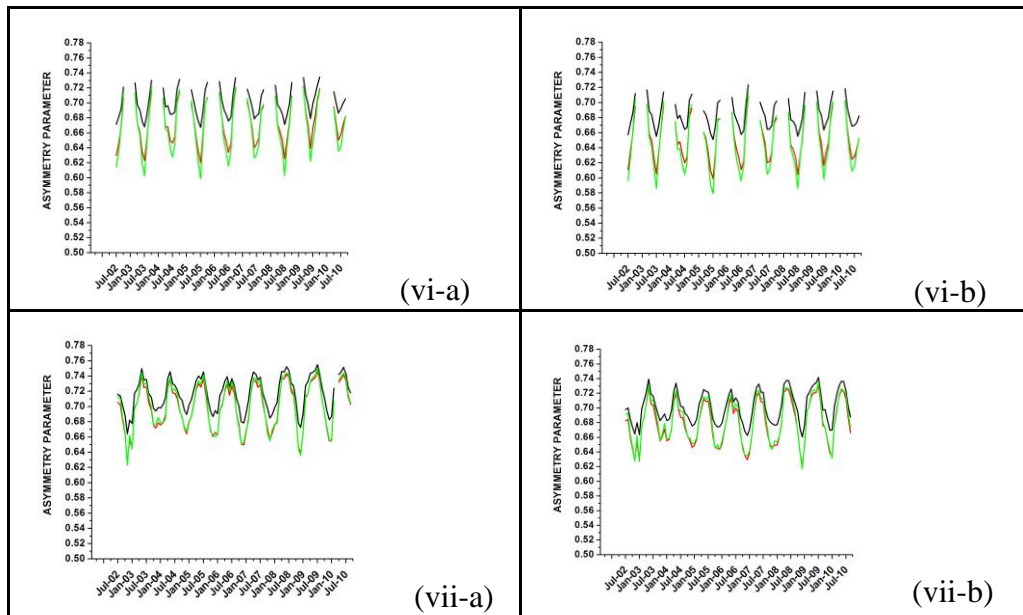
1027 **Figure 5.** Slope (in units decade⁻¹) of MODIS g_{aer} deseasonalized anomalies over the
 1028 period 2002-2010 from MODIS-Terra (-a, top) and MODIS-Aqua (-b, bottom), for
 1029 the wavelengths of 470 nm. Results are shown only if the trend is statistically
 1030 significant at the 95% confidence level.

1031



1032

1033 **Figure 6.** Inter-annual (2002-2010) variation of monthly mean g_{aer} values at 470 nm
 1034 averaged over the sub-regions of: (i) Black Sea, (ii) Eastern Atlantic Ocean, (iii)
 1035 Mediterranean Sea, (iv) Middle East, (v) North-eastern Atlantic Ocean, (vi) North
 1036 Europe and (vii) Persian Gulf. Results are given based on MODIS-Terra (-a, left
 1037 column) and MODIS-Aqua (-b, right column).

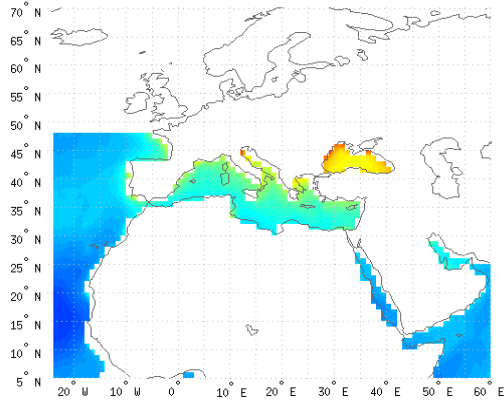


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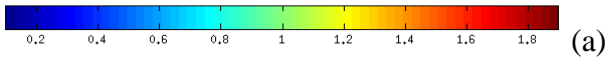
1040 **Figure 6 (continued).** Inter-annual (2002-2010) variation of monthly mean g_{aer} values
 1041 at 470 nm averaged over the sub-regions of: (i) Black Sea, (ii) Eastern Atlantic Ocean,
 1042 (iii) Mediterranean Sea, (iv) Middle East, (v) North-eastern Atlantic Ocean, (vi) North
 1043 Europe and (vii) Persian Gulf. Results are given based on MODIS-Terra (-a, left
 1044 column) and MODIS-Aqua (-b, right column).

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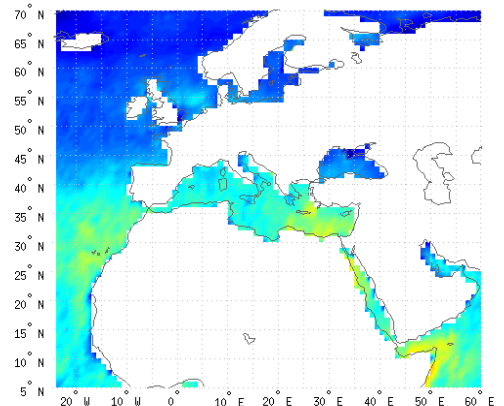
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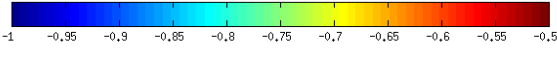
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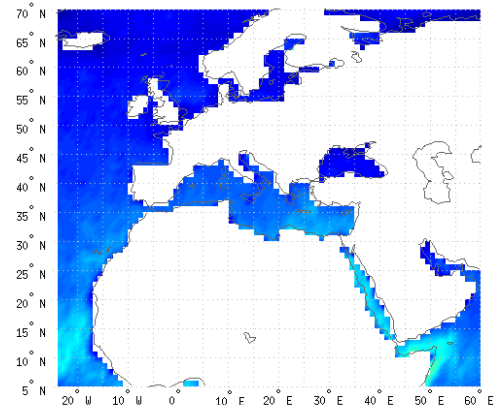
(a)



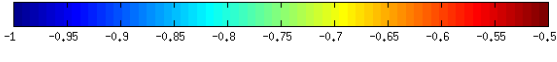
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(b)

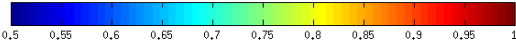
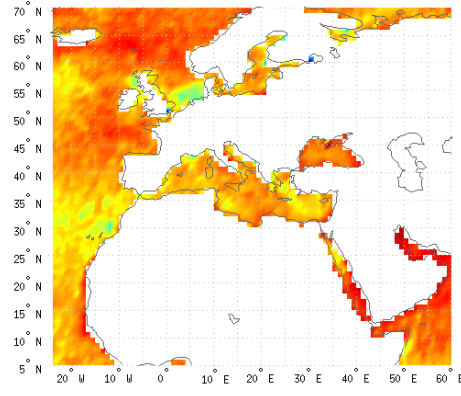
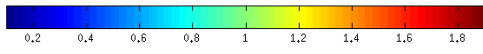
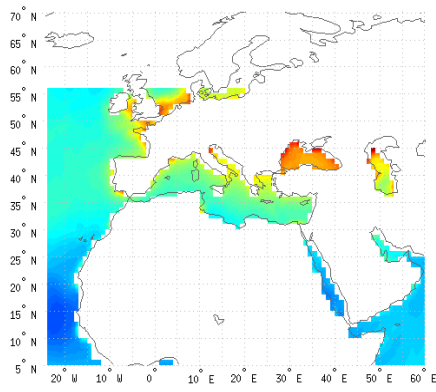


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(c)

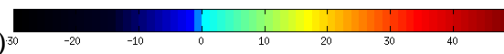
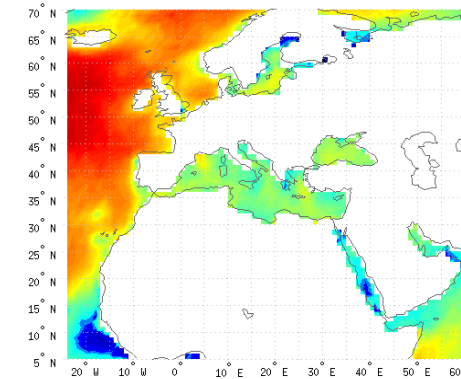
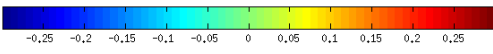
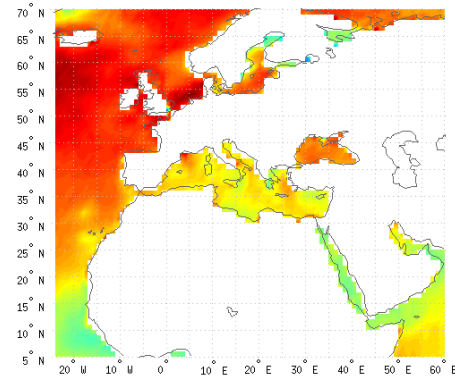
1050 **Figure 7.** (a) Geographical distribution of MODIS-Aqua 051 Ångström exponent
 1051 ($AE_{550-865}$) values averaged over 2002-2010, at the wavelength pair of 550-865nm.
 1052 Winter AE data are missing from the northernmost areas and therefore the long-term
 1053 averages in (a) are left blank. The correlation coefficients between $AE_{550-865}$ and g_{aer}
 1054 data at 660 and 870 nm are given in (b) and (c), respectively.



1055

(a)

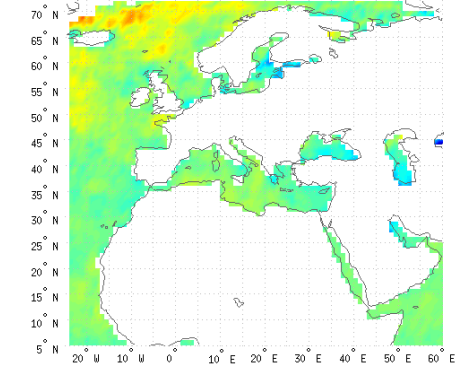
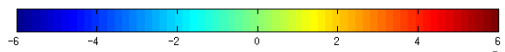
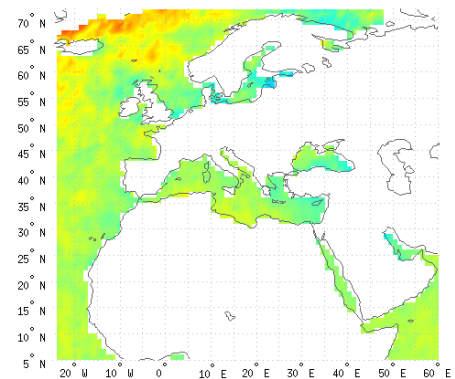
(b)



1056

(c)

(d)



1057

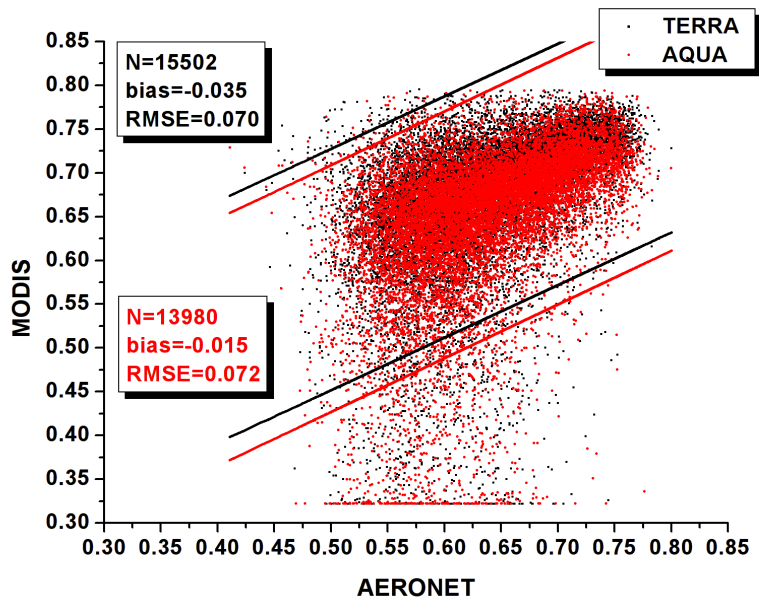
(e)

(f)

1058

1059 **Figure 8.** (a) Geographical distribution of MODIS-Aqua C006 Angström exponent
 1060 (AE₅₅₀₋₈₆₅) values averaged over 2002-2010, at the wavelength pair of 550-865nm.
 1061 Winter AE data are missing from the northernmost areas and therefore the long-term
 1062 averages in (a) are left blank. In (b), (c) and (d) are given the correlation coefficients,
 1063 the absolute biases and the relative percent biases, respectively, between the C006 and
 1064 corresponding 051 AE₅₅₀₋₈₆₅ data. In (e) and (f) are given the computed

1065 deseasonalized trends of MODIS Aqua 051 and C006 AE₅₅₀₋₈₆₅) slope values for years
1066 2002-2010, respectively.
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1070 **Figure 9.** Scatterplot comparison between g_{aer} values at 870 nm from MODIS Terra

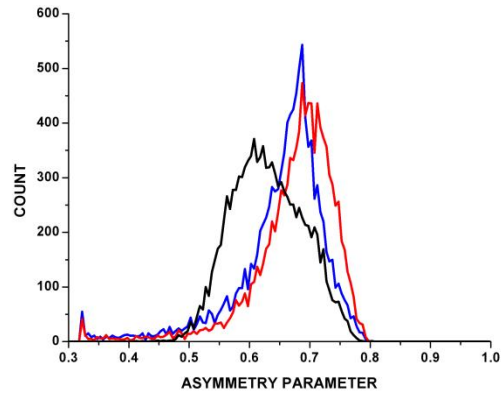
1071 (black color) and Aqua (red color) and corresponding values from AERONET stations

1072 (blue squares, Fig. 1). The 95% prediction bands as well as the mean bias (AERONET

1073 minus MODIS) and root mean squared error are given.

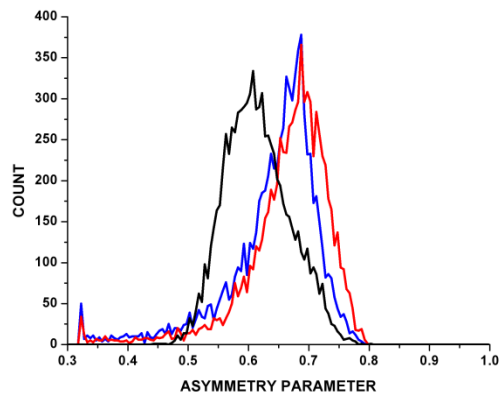
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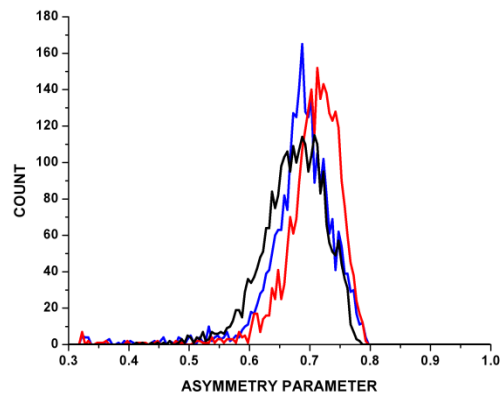
(a)

1076



(b)

1077



(c)

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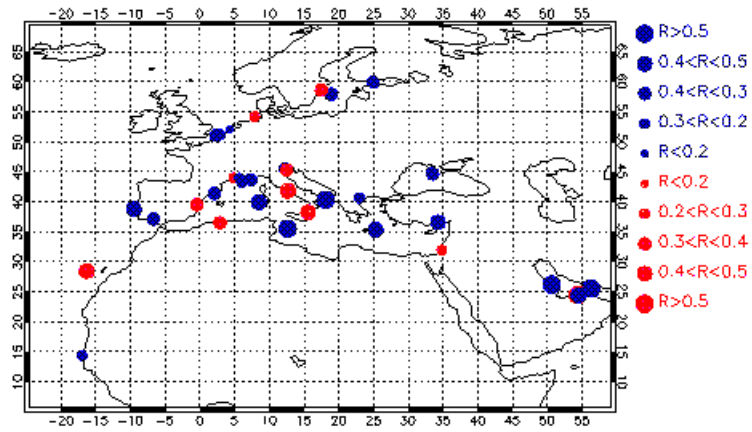
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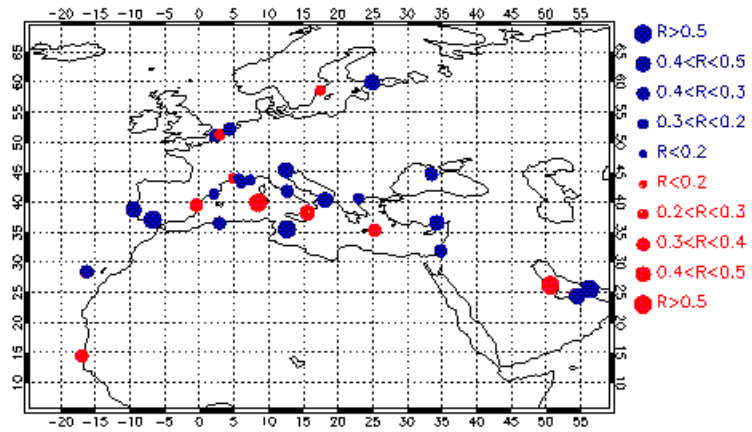
Figure 10. Frequency distribution histograms for MODIS-Terra (red colored lines) MODIS-Aqua (blue-colored lines) and AERONET (black lines) g_{aer} values at 870 nm. The histograms are given separately for: (a) the entire study region, (b) Europe and (c) Africa, Middle East and Arabian peninsula.

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(a)

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(b)

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Figure 11. Map distribution of correlation coefficients between: (i) MODIS-Terra and AERONET g_{aer} values at 870 nm (left column) and (ii) MODIS-Aqua and AERONET g_{aer} values at 870 nm (right column). The size of circles corresponds to the magnitude of correlation coefficients, while blue and red colors are used for stations for which MODIS and AERONET indicate same and opposite tendency of g_{aer} , respectively.