

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 of 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 is
27 taking place. The g_{aer} values tend to decrease with increasing wavelength, especially
28 over 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), which provides a good measure of the aerosol load over
55 an area, one of the most important optical properties of atmospheric particles, which is
56 used in radiative transfer, climate, and general circulation models, is the asymmetry
57 parameter (g_{aer}). The asymmetry parameter describes the angular distribution of the
58 scattered radiation and determines whether the particles scatter radiation preferentially
59 to the front or back. The globally available satellite based AOD data are considered to
60 a great extent reliable and adequate, due to significant developments in surface and
61 satellite measurements during the last two decades, and particularly after 2000. On the
62 other hand, despite of the important role of the asymmetry parameter, relevant global
63 coverage data are measured only for the few last years, or are available in long-term
64 aerosol climatologies such as Global Aerosol Data Set (GADS, Koepke et al. 1997)
65 and Max Planck Aerosol Climatology (MAC, Kinne et al., 2013). Even so,
66 asymmetry parameter data are usually examined for regions with limited geographical

67 extent and temporal coverage (Di Iorio et al, 2003), without intercomparison between
68 alternative data platforms.

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

75 For this study we focus on the region defined by latitudes 5°N to 70°N and longitudes
76 25°W to 60°E, including North Africa, the Arabian peninsula, Europe, and the greater
77 Mediterranean basin (Fig. 1). This area is selected because of the simultaneous
78 presence of a variety of particles, both natural and anthropogenic (e.g. desert dust,
79 marine, biomass burning, anthropogenic urban / industrial pollution) as shown in
80 previous studies (Lelieveld et al., 2002; Sciare et al., 2003; Pace et al., 2006; Lyamani
81 et al., 2006; Gerasopoulos et al., 2006; Kalivitis et al., 2007). This is due to the fact
82 that two of the largest deserts of the planet are partly included in our area of interest,
83 i.e. the Arabian desert and the Sahara, while one finds also significant sources of
84 anthropogenic pollution, mainly in the European continent, with urban and industrial
85 centres. Moreover, our area of interest and primarily its desert areas, are characterised
86 by a large aerosol load (large optical depth, Remer et al. 2008). Finally, significant
87 regions in this area, more specifically the Mediterranean basin and North Africa, are
88 considered climatically sensitive, since they are threatened by desertification (IPCC,
89 2007; 2013). This is the first study (to our knowledge) that focuses on asymmetry
90 parameter over a geographically extended area, while at the same time compares
91 satellite with ground-station data.

92

93 **2 Data**

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

97

$$98 \quad g = \frac{\overline{\omega_1}}{3} = \frac{1}{2} \int_{-1}^1 P(\cos\Theta) \cos\Theta d \cos\Theta \quad (1)$$

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

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

122

123 **2.1 Satellite MODIS Terra and Aqua data**

124 MODIS is an instrument (radiometer) placed on the polar-orbiting satellites of NASA
125 (National Aeronautics and Space Administration) Terra and Aqua, 705 km from the
126 Earth, in the framework of the Earth Observing System (EOS) programme. Terra was
127 launched on 18 December 1999, while Aqua was launched on 4 May 2002. The two
128 satellites are moving on opposite directions and their equatorial crossing times are at
129 10:30 (Terra) and 13:30 (Aqua). MODIS is recording data in 36 spectral channels
130 between the visible and the thermal infrared ($0.44 - 15 \mu\text{m}$), while its swath width is
131 of the order of 2330 km, which results in almost full planetary coverage on a daily

132 basis. The global MODIS database is generally considered as one of the most reliable
133 at present.

134 Aerosol properties are monitored in 7 spectral channels between 0.47 and 2.13 μm
135 and final results are derived through algorithms developed for aerosol quantities both
136 over land and ocean (Kaufman et al., 1997; Tanré et al., 1997; Ichoku et al., 2002;
137 Remer et al., 2005). MODIS data are organised in “collections” and “levels”.
138 Collections comprise data produced by similar versions of the inversion algorithms,
139 with the most recent being “051”, which includes also outputs from the “Deep Blue”
140 algorithm. Levels are characterised by data of different quality analysis and spatial
141 resolution.

142 In this study we use daily MODIS data for the asymmetry parameter (g_{aer}) provided
143 on an $1^\circ \times 1^\circ$ grid (namely 100x100 km), from the most recent Collection 051, Level
144 3. These data were measured at wavelengths 470, 660, and 870 nm, only over oceanic
145 regions, since they were derived through the algorithm for aerosol properties over the
146 ocean. The period of analysis stretches from 24-2-2000 to 22-9-2010 for MODIS-
147 Terra and from 4-7-2002 to 18-9-2010 for MODIS-Aqua. We also used Level 3 daily
148 Angström exponent data from MODIS-Aqua C005, and also spectral aerosol optical
149 depth data from MODIS-Aqua C006 datasets, from which we computed C006
150 Angström exponent. These data were used to assess the validity of g_{aer} data and their
151 changes, as discussed in section 3.2.3.

152

153 **2.2 Ground based AERONET data**

154 AERONET (AErosol RObotic NETwork) is a global network of stations focused on
155 the study of aerosol properties. AERONET currently encompasses about 970 surface
156 stations (number continuously evolving) equipped with sun photometers of type
157 CIMEL Electronique 318 A (Holben et al., 1998), which take spectral radiation flux
158 measurements.

159 The optical properties of aerosols are extracted through the application of inversion
160 algorithms (Dubovik and King, 2000). Data are provided on three levels (1.0, 1.5, and
161 2). In the present work, we use the most reliable Level 2 data, due to their being
162 cloud-screened and quality-assured. AERONET calculates the asymmetry parameter
163 at wavelengths 440, 675, 870, and 1020 nm. We employ daily Level 2 asymmetry
164 parameter data from 69 stations (Fig. 1), contained in our study area (N. Africa,

165 Arabian peninsula, Europe). We choose only coastal stations, in order to maximize the
166 coexistence of satellite marine g_{aer} data with surface data. Also, in order to compare
167 corresponding data between the satellite and station platforms, we perform
168 comparison only for the 440, 675 and 870 nm.

169

170 **3 Satellite based results**

171 **3.1 Geographical distributions**

172 The spatial distribution of annual mean values of g_{aer} is given in Fig. 2 separately at
173 the wavelengths 470, 660 and 870 nm. The values are averages over the common
174 period between Terra and Aqua, namely 4 July 2002 till 18 September 2010. A
175 significant spatial variability is evident, with MODIS-Terra values varying within the
176 ranges 0.63 - 0.76, 0.57 – 0.75, and 0.55 – 0.74, at 470, 660 and 870 nm, respectively.
177 The results exhibit a decreasing tendency of g_{aer} with increasing wavelength,
178 consistent with the theory. Similar results are also obtained from MODIS-Aqua, but
179 with slightly smaller values than Terra by up to 0.02 on average. More specifically,
180 the corresponding ranges of wavelengths are 0.63 - 0.75, 0.57 – 0.73, and 0.55 – 0.73.
181 The smaller Aqua than Terra g_{aer} values could be attributed to smaller sizes of
182 aerosols in midday than morning, corresponding to passages of Aqua and Terra,
183 respectively, associated with lower relative humidity values and shrinking of aerosol
184 particles. Such diurnal variation has been also reported for AOD (Smirnov et al.,
185 2002; Pandithurai et al., 2007), but either decreasing or increasing in the day because
186 of the influence of other factors too, e.g. emissions or wind conditions, apart from
187 aerosol hygroscopicity.

188 In general, the largest g_{aer} values (deep red colors) are observed off the coasts of West
189 Africa (eastern tropical Atlantic Ocean) at all three wavelengths. High values are also
190 found over the Red and Arabian Seas. These high values are due to strong dust
191 outflows from the Saharan and Arabian deserts carrying out coarse aerosol particles
192 causing strong forward scattering. Nevertheless, the Persian Gulf region, which is
193 surrounded by deserts, is characterized by relatively smaller g_{aer} values. More
194 specifically, values as small as 0.69 (MODIS-Terra) and 0.67 (MODIS-Aqua) are
195 observed in this region at 470 nm, while at the longer wavelengths (660, 870 nm) the
196 smallest values are equal to 0.66 (Terra) and 0.64 (Aqua). The smaller g_{aer} values over
197 the Persian Gulf can be attributed to the presence of fine aerosols, which is

198 corroborated by the low effective radius and large fine-fraction measurements by
199 MODIS over the Persian Gulf, compared to neighbouring areas (not shown here).
200 These fine particles originate from the industrial activities in the Gulf countries
201 related to oilfields or refineries (Goloub and Arino, 2000; Smirnov et al., 2002a,b;
202 Dubovik et al., 2002).

203 The high g_{aer} values over the northeastern tropical Atlantic Ocean as well as west of
204 the Iberian coasts are possibly related with the presence of coarse sea salt particles.
205 On the other hand, the asymmetry parameter takes clearly smaller values over the
206 Black Sea, where according to MODIS-Terra varies between 0.63 and 0.7 at 470 nm,
207 0.57 and 0.67 at 660nm, and 0.55 and 0.66 at 870 nm, with the smallest values
208 appearing in the Crimean peninsula (corresponding maximum Aqua values are
209 smaller by 0.02). The small Black Sea g_{aer} values can be associated with the vicinity
210 of industrial but also biomass burning activities in nearby countries. A region of
211 special interest is the Mediterranean basin since it hosts a large variety of aerosols like
212 anthropogenic, desert dust or sea salt (e.g. Barnaba and Gobbi, 2004). The MODIS
213 results over this region show relatively small g_{aer} values, secondary to those of Black
214 Sea, characterized by an increase from north to south, which is more evident at 660
215 and 870 nm. More specifically, based on MODIS-Terra, g_{aer} over the Mediterranean
216 takes values from 0.68 to 0.74 at 470 nm, while at 670 and 870 nm it ranges from 0.64
217 to 0.73 and 0.62 to 0.72, respectively. According to MODIS-Aqua the g_{aer} values are
218 slightly smaller again. The observed low values in the northern parts of the
219 Mediterranean are probably associated with the presence of fine anthropogenic
220 aerosols transported from adjacent urban and industrial areas in the north, especially
221 in central Europe. In contrast, the higher g_{aer} values in the southern Mediterranean,
222 particularly near the North African coasts, can be explained by the proximity to the
223 Sahara desert and the frequent transport of significant amounts of coarse dust (e.g.
224 Kalivitis et al., 2006; Hatzianastassiou et al., 2009; Gkikas et al., 2009; 2014).

225 The spatial distributions of climatological monthly mean g_{aer} values from MODIS-
226 Aqua at 470 nm reveal significant differences either as to the range or to the patterns
227 of the seasonal variability, depending on the area (Fig. 3). Thus, in tropical and sub-
228 tropical areas of Atlantic Ocean (up to about 30°N), where dust is exported from
229 Sahara, g_{aer} keeps high values throughout the year, which reach or even exceed 0.74
230 locally. Over the regions of Arabian and Red Seas and the Gulf of Aden, which also
231 experience desert dust transport, larger g_{aer} values appear in the period from March to

232 September, with a maximum on August (locally as high as 0.75-0.76). This seasonal
233 behavior is in line with intra-annual changes of dust production over the Arabian
234 peninsula indicated primarily by MODIS Angström Exponent (AE) and secondarily
235 by Deep Blue aerosol optical depth data and reported in the literature (Prospero et al.,
236 2002). Indeed, the production of dust there is relatively poor in winter, increases in
237 March and April and becomes maximum in June and July (Prospero et al., 2002).
238 Over the Arabian Sea, it is known that large amounts of desert dust are carried out
239 during spring and early summer (Prospero et al, 2002; Savoie et al., 1987; Tindale and
240 Pease, 1999; Satheesh et al., 1999). Nevertheless, according to MODIS, the seasonal
241 variability of g_{aer} remains relatively small there in line with a small seasonal
242 variability in MODIS Deep Blue AE data. This can be explained by the presence of
243 sea salt coarse particles throughout the year, with which dust particles co-exist.

244 A greater seasonal variability exists over the Persian Gulf, where g_{aer} values are
245 higher during spring and in particular in summer (up to 0.74 at 470 nm according to
246 Aqua), and smaller in autumn and winter (area-minimum values smaller than 0.65).
247 This seasonal behavior can be explained taking into account the meteorological
248 conditions over the greater area of the Gulf; from June to September dry northwestern
249 winds (Shamal) blow from northwest carrying desert dust from the arid areas of Iraq
250 (Heishman 1999; Smirnov et al. 2002a,b). The transport of dust is gradually decreased
251 in autumn, minimizes in winter and increases again in spring. When the presence of
252 desert dust is limited, a significant fraction of total aerosol load in the region is
253 consisted of fine anthropogenic particles (Smirnov et al. 2002a,b), which can explain
254 the observed relatively small g_{aer} values in autumn and winter.

255 In the Mediterranean basin, g_{aer} exhibits a relatively small seasonal variation, though
256 lower values tend to appear in summer and secondarily in early and late spring, in line
257 with the stronger presence of dust in the area, transported from the Sahara desert
258 (Gkikas et al., 2013). On the contrary, over the Black Sea, a clear seasonal cycle is
259 apparent, with higher values in the cold period of the year and smaller in the warm
260 one. More specifically, according to MODIS-Aqua, the values at 470 nm drop down
261 to 0.61 in summer months whereas they reach 0.7 in January and December.

262 It is also interesting to look at the geographical distribution of monthly g_{aer} values in
263 latitudes higher than 50°N, for which annual mean values were not given in Fig. 2
264 because of unavailability of data for all months. Off the coasts of northern France
265 (English Channel) and Germany the asymmetry parameter has small values, with a

266 non-significant annual course (note that values do not exist for January and February).
267 In these areas, the aerosol load consists mainly of anthropogenic polluted particles,
268 which explains the small g_{aer} values there.

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

278 In the higher latitudes of Atlantic Ocean, where the presence of maritime aerosols is
279 dominant, it is observed a remarkable month by month variation of asymmetry
280 parameter, with low values in summer (values up to 0.59) against high values (up to
281 0.75-0.77) in spring (March, April) and autumn (October). This difference is possibly
282 explained by the seasonal variability of aerosol size in the northern Atlantic. Apart
283 from the presence of coarse sea salt throughout the year, in spring and summer small
284 particles are formed through photochemical reactions of dimethylsulphide (DMS)
285 emitted by phytoplankton decreasing the aerosol size. Moreover during summer fine
286 anthropogenic aerosols are transported in the region from North America (Yu, 2003;
287 Chubarova, 2009). These result in lower g_{aer} values between May and August.

288 Based on MODIS-Terra, the patterns of spatial distribution are generally the same
289 with Aqua, with slightly larger g_{aer} values. At larger wavelengths (660, 870 nm) it is
290 observed a decrease of g_{aer} , in particular of its smallest values. Further details and also
291 an overall picture will be given later on, in the section (3.2.1) which deals with
292 climatological monthly mean values not at the pixel but regional level.

293

294 **3.2 Temporal variability**

295 **3.2.1 Seasonal variability**

296 In order to provide an easier assessment of the seasonal cycle of aerosol asymmetry
297 parameter and its changes from a region to another, but also among the different
298 wavelengths (470, 660 and 870 nm), the study region was divided in 6 smaller sub-

299 regions (see Fig. 1). For each sub-region, the average values of monthly mean
300 climatological data of the pixels found within each sub-region's geographical limits
301 have been computed and are given in Fig. 4, for every wavelength and for Terra and
302 Aqua. It appears that the seasonal cycle differs between the sub-regions, as it has been
303 already shown in the geographical map distributions discussed in the previous section.

304 At 470 nm (Fig. 4i), the intra-annual variability of g_{aer} is greater over the Black Sea,
305 where it is as large as 0.06 according to MODIS-Terra and 0.05 according to MODIS-
306 Aqua, the north-eastern Atlantic Ocean (0.04 and 0.05 for Terra and Aqua,
307 respectively) and the seas of North Europe (0.05 for both Terra and Aqua). In these
308 regions, there is a tendency for smaller values during summer. More specifically, in
309 the Black Sea the smallest g_{aer} value (0.64) is observed in June, over the seas of North
310 Europe in July and over the north-eastern Atlantic Ocean in August. In these regions,
311 the largest values appear in the cold period of the year. Reverse seasonality with a
312 large seasonal amplitude is observed over the Persian Gulf, where the variability is as
313 large as 0.08, according to both MODIS-Terra and Aqua. The seasonal cycle of g_{aer}
314 over the Middle East exhibits a smaller range of variability (0.02 for MODIS-Terra
315 and 0.03 for Aqua) along with a reverse seasonal variation, with maximum values in
316 summer and minimum in winter. In the other two sub-regions (Mediterranean and
317 eastern Atlantic Ocean) the annual range of values is small (< 0.02). It is noteworthy
318 that in the Mediterranean Sea, there is a weak tendency of appearance of double
319 maxima in winter and spring. The spring maximum should be associated with the
320 presence of desert dust particles, which are transported from Sahara, mainly in the
321 eastern Mediterranean in this season (e.g. Fotiadi et al., 2006; Kalivitis et al., 2007;
322 Papadimas et al. 2008, Gkikas et al. 2009; Hatzianastassiou et al., 2009; Gkikas et al.,
323 2013). There is also a similar transport of Saharan dust in the central and western
324 Mediterranean during summer and autumn (e.g. Gkikas et al., 2009; 2013), but then
325 the predominance is not so clear because of the co-existence of fine anthropogenic
326 aerosols. Regardless of the annual cycle, smaller g_{aer} values are clearly distinguished
327 over the Black Sea and North Europe seas throughout the whole year.

328 At 660 nm, the g_{aer} values are lower than at 470 nm, in particular over Black Sea,
329 North Europe and North-East Atlantic, whereas the intra-annual variability (range of
330 g_{aer} values) increases up to 0.1 (Terra) and 0.08 (Aqua) over the Black Sea. This
331 increase is mainly attributed to the reduction of summer values due to the strong
332 appearance of fine aerosols in this season. Also, at 660 nm, there is a clearer double

333 annual variation of g_{aer} over the Mediterranean Sea than at 470 nm. At 870 nm the
334 general picture is similar to that of 660 nm though a further increase of month by
335 month variability is noticeable.

336 In general, our results indicate that over the regions characterized by a strong presence
337 of desert dust particles (eastern Atlantic and the Middle East and Mediterranean Seas)
338 the annual range of variability of g_{aer} is smaller than in the other regions. An
339 additional feature above regions with desert dust is the smaller decrease of g_{aer} values
340 with increasing wavelengths, which can be attributed to the different spectral behavior
341 of solar radiation scattering by fine and coarse aerosols (e.g. Dubovik et al, 2002; J. Bi
342 et al, 2011).

343 It should be noted here that according to our results, using MODIS-Terra and Aqua
344 data, the g_{aer} seasonal cycle is about similar but with generally greater larger Terra
345 than Aqua values.

346 **3.2.2 Inter-annual variability and changes**

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

354 In general, the estimated changes are relatively small. Terra produces widely
355 statistically significant positive trends, showing that during the period of interest, the
356 asymmetry parameter increased over the examined area, with very few exceptions.
357 The results from Aqua are statistically significant at considerably fewer cells, but also
358 give a few points with decreasing g_{aer} . Based on Terra data, the stronger increases are
359 observed in the eastern and southern Black Sea, as well as over the Baltic and Barents
360 Seas. According to MODIS-Aqua, negative changes are found over few Atlantic
361 Ocean cells. Both Aqua and Terra report increases of g_{aer} over the Persian Gulf, the
362 Red Sea, South Black Sea, East Mediterranean, the coast of the Iberian Peninsula, and
363 some coastal areas of West Africa. The differences encountered between the Terra
364 and Aqua g_{aer} trends may be attributed to the different time of passage of each satellite
365 platform carrying the same MODIS instrument, given that everything else is the same.

366 Nevertheless, most probably, they may be the result of calibration differences between
367 the two MODIS sensors. It is known that there is a degradation of MODIS sensor
368 (Levy et al., 2010; Lyapustin et al., 2014) impacting time series of MODIS products.
369 More specifically, it is also known that Terra suffers more than Aqua from optical
370 sensor degradation. These calibration issues are known to affect MODIS AOD
371 retrievals, producing an offset between Terra and Aqua, and they are also expected to
372 affect aerosol asymmetry parameter, which is probably more sensitive to such
373 calibration uncertainties than AOD. In this sense, the results of Fig. 5 shown here are
374 not to be taken as truth but rather they are given as a diagnostic of a problematic
375 situation with MODIS aerosol asymmetry parameter inter-annual changes. Such
376 calibration issues are expected to be addressed, at least partly, in the new Collection
377 006 products. Nevertheless, a preliminary comparison between MODIS Aqua C005
378 and C006 Angström exponent (AE), which is a common aerosol size parameter, using
379 AE data for the 550-865 pair of wavelengths spanning the period 2002-2010, does not
380 reveal significant modifications in geographical patterns of AE inter-annual changes.
381 This puts some confidence on the C005 g_{aer} results given in the present study. The
382 results of this analysis are presented in detail in the next sub-section (3.2.3).

383 The overall g_{aer} changes of Fig. 5 may hide smaller timescale variations of g_{aer} , which
384 are obtained by the time-series shown in Fig. 6. Results are given for the 7 sub-
385 regions defined previously, at the three different wavelengths and for Terra and Aqua
386 separately. A general pattern is the decrease of g_{aer} values with increasing wavelength,
387 in particular from 470 to 660 nm. The largest month to month and year to year
388 variation is for Black Sea (Fig. 6i). Relatively large variability is also found in the
389 sub-regions of NE Atlantic (6v), North Europe (6vi) and the Persian Gulf (6vii). On
390 the contrary, small variability is noticed in the eastern Atlantic, where systematic dust
391 outflows from Sahara take place leading to consistently high values of g_{aer} . There are
392 also some other interesting patterns, like the significant drop of g_{aer} with wavelength
393 in areas characterized by the presence of fine aerosols, namely the Black Sea, North
394 Europe and the Persian Gulf (Figs, 6i,vi,vii, respectively). The specific patterns of
395 inter-annual changes of g_{aer} are suggested by both Terra and Aqua, though a slight
396 overestimation by Terra is again apparent in this figure. The obtained results of our
397 analysis are meaningful and in accordance with the theory, underlining the ability of
398 satellite observations to reasonably capture the g_{aer} regime over the studied regions.

399 **3.2.3 Possible uncertainties of MODIS aerosol asymmetry parameter**

400 The MODIS aerosol asymmetry parameter is not a direct retrieval product of the
401 MODIS retrieval algorithm, but it is rather a derived by product. Since this parameter
402 is dependent on aerosol modes used and relative weights, it is understood that there
403 can be uncertainties associated with it. Therefore questions may arise about the
404 validity of g_{aer} and their spatial and temporal patterns presented in the previous sub-
405 sections. Given that, as it was already mentioned, it is an aerosol optical parameter
406 that is valuable and highly required by radiative transfer and climate models, it is
407 worth assessing it through comparison against another more common aerosol size
408 parameter, namely the C005 MODIS Angström exponent at the 550-865 nm
409 wavelength pair ($AE_{550-865}$) over ocean, which is an evaluated MODIS aerosol size
410 product (Levy et al., 2010). Figure 7a, displays the geographical distribution of AE for
411 the study period, i.e. 2002-2010. The main geographical patterns in Fig. 7a are in line
412 with those of asymmetry parameter (Fig. 2). For example, note the high AE values in
413 the Black Sea (yellowish-reddish colors), indicative of fine aerosols, the relatively
414 high values in the Mediterranean Sea (greenish-yellowish colors) and the low values
415 (deep bluish colors) off the western African coasts corresponding to exported Saharan
416 dust. The consistency between g_{aer} and AE data is shown by the strong anti-
417 correlation between the MODIS $AE_{550-865}$ and g_{aer} data at 660 and 870 nm, shown in
418 Figures 7b and 7c, respectively. Strong negative correlation coefficients, larger than
419 0.7 and 0.8 in Figs 7b and 7c, respectively, relate inversely high/low g_{aer} values with
420 low/high AE ones over the same areas. These results indicate that the spatial patterns
421 of MODIS C005 g_{aer} product are reasonable as compared to the C005 Angström
422 exponent data.

423 Since questions arise about possible uncertainties regarding the long-term variability
424 of MODIS C005 aerosol size products, due to the calibration issues discussed in the
425 previous section, the corresponding MODIS C006 AE product is displayed in Fig. 8a.
426 From Figs. 8a and 7a a similarity is apparent in the main geographical patterns of the
427 two collections' AE product. The similarity between C005 and C006 AE data is also
428 depicted in the computed correlation coefficients (Fig. 8b), exceeding 0.8, and biases
429 (in absolute and relative percentage terms, Figs 8c and 8d, respectively) which are
430 smaller than 0.1 or 10% in most areas of the study region and 0.2 or 20% almost
431 everywhere. It should be noticed that our AE results are in line with those of Levy et
432 al. (2013, Fig. 15) which refer, however, only to year 2008 (ours are for 2002-2010).
433 In addition, a comparison is attempted in Figs 8e and 8f between the computed trends
434 of C005 and C006 AE data over the common period 2002-2010, in order to assess

435 whether changes are detected, which could be an indication of possible changes in
 436 corresponding asymmetry parameter trends. Figures 8e and 8f show the computed
 437 deseasonalized trends of slope values for both C005 and C006 AE. The results reveal
 438 similar patterns between C005 and C006. Small trends are found in both of them, in
 439 agreement with the small trends of asymmetry parameter reported in Fig. 5. It is found
 440 that the sign of AE trends mainly does not change from C005 to C006. This might be
 441 a signal that no changes of aerosol asymmetry parameter are expected in C006 and
 442 puts some confidence on the C005 results given in the present study.

443

444 **4 Evaluation against AERONET data**

445 In this section, we compare the satellite-measured aerosol asymmetry parameter with
 446 measurements from the global network of surface stations of AERONET, which is
 447 considered as the reference dataset (Holben et al., 1998). For this purpose, we
 448 identified the AERONET stations inside our area of interest and finally utilised only
 449 the coastal ones, so that both satellite and surface data be available. The total number
 450 of these stations is 69, and their locations are shown in Fig. 1 (blue squares).

451 Table 1 contains the comparison statistical metrics for all wavelengths (Pearson
 452 correlation coefficient, bias, standard deviation, slope, intercept) of the comparison
 453 between surface data from AERONET and satellite data from MODIS-Terra and
 454 MODIS-Aqua, which correspond to the $1^\circ \times 1^\circ$ cell wherein each station is located. For
 455 this analysis, we use all cells and days with common data between Terra-AERONET
 456 and Aqua-AERONET. The mean differences are calculated as $g_{\text{aer}}(\text{AERONET}) -$
 457 $g_{\text{aer}}(\text{Aqua})$ and $g_{\text{aer}}(\text{AERONET}) - g_{\text{aer}}(\text{Terra})$.

458 In general, we may note that on an annual level, the MODIS-Terra and Aqua
 459 asymmetry parameter values at 470 nm are not in very good agreement with the
 460 respective data from AERONET at 440 nm, while the results at the largest
 461 wavelengths are more reassuring, though not being very satisfactory (increasing R and
 462 decreasing relative bias and RMSE values at 675/660 nm and 870 nm). At 870 nm
 463 (Table 1 and Fig. 9), correlation coefficients are found to be the largest and equal to
 464 0.47 (AERONET-Terra) and 0.46 (AERONET-Aqua), while satellite data are slightly
 465 overestimated compared to the surface data (bias -0.035 or 5.54% and -0.015 or -
 466 2.43%, respectively).

467 It is important to note that the agreement of satellite and surface data is better in
 468 spring and summer, for all studied wavelengths. Specifically, the correlation

469 coefficients increase up to 0.35, 0.50 and 0.54 at 440/470 nm, 660/675 nm and 870
470 nm, respectively, while the bias decreases down to 0.0005 (0.07%), 0.003 (0.46%)
471 and 0.007 (1.11%), respectively.

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

478 In Fig. 9 we present a scatterplot comparison between MODIS and AERONET g data
479 pairs. There is bias towards larger g values from both Aqua and Terra compared to
480 AERONET, with Terra overpredicting more than Aqua. The root mean square error to
481 the fit between MODIS and AERONET is very similar between Aqua and Terra.
482 There are concerns on the application of ordinary least squares regression, arising
483 from the assumption that as the assigned independent variable, AERONET values
484 should be free from error. We cannot guarantee the validity of this assumption, so we
485 recognize that the reported R and slope values from Fig. 9 and Table 1, if viewed as
486 metrics of agreement between MODIS g and real g , may be subject to the effect of
487 regression dilution and consequently biased low. This possible bias for R and slope
488 could be neglected only if AERONET errors can also be considered negligible. With
489 the above caveat in mind, the applied least-squares fit line to the scatterplot
490 comparison between matched MODIS-AERONET data pairs (Fig. 9) indicates that
491 MODIS overestimates g_{aer} more in the smaller than larger values, i.e. more for fine
492 than coarse particles.

493 We present the frequency distributions of asymmetry parameter daily values (Fig. 10)
494 on the days when data from all three databases (MODIS-Terra, MODIS-Aqua and
495 AERONET) were provided. Fig. 10a corresponds to the whole area of interest, while
496 Figs. 10b and c correspond to two broad sub-regions with basic differences in the
497 aerosol source, namely Europe with great anthropogenic sources, and Africa, Middle
498 East and Arabian peninsula, with predominant natural sources and mainly desert dust.
499 There is an apparent skew in the MODIS-Terra and MODIS-Aqua g_{aer} distributions,
500 while the AERONET distributions are more symmetrical. Moreover, the satellite data
501 distributions show larger values and smaller standard deviations compared to
502 AERONET, with the Terra overestimation being more exaggerated. The disagreement

503 is more pronounced in the sub-region of Europe, while in the sub-region of North
504 Africa / Arabian peninsula, the distributions of satellite and surface data agree more
505 thus confirming the finding of Fig. 9 based on the slope of applied linear regression
506 fit. Values over Europe are generally smaller than over North Africa / Arabian
507 peninsula (Fig. 3), which can be attributed to the presence of larger size particles of
508 desert origin in the latter sub-region, in contrast to Europe, where due to industrial
509 activity and frequent biomass burning the presence of smaller size particles is
510 important. Therefore, the smaller g_{aer} values (<0.6) in the frequency distributions of
511 the whole area, are overwhelmingly contributed by the European sub-region,
512 contrasting with larger values (0.7-0.75) being contributed by both sub-regions and
513 even more by N. Africa/Arabian peninsula at larger g_{aer} .

514 Potentially useful results may be derived by the comparison of the temporal trends
515 from satellite and surface data. We show in Fig. 11 the absolute and relative changes
516 of the asymmetry factor, calculated through regression on monthly time series of g_{aer}
517 at 9 AERONET stations with satisfactory temporal coverage of data, selected to have
518 recorded at least 40 monthly values. In the same figure, these variations are compared
519 with corresponding data from MODIS-Terra and MODIS-Aqua, from the 1x1 degree
520 cells containing the locations of the 9 selected stations. We note that we only perform
521 this analysis in a month only if all three datasets give data for the specific month. It
522 should be noted that the g_{aer} changes for these stations do not refer to the same period
523 but they all ensure a complete enough time period enabling thus the derivation of safe
524 conclusions on how MODIS and AERONET changes compare to each other. At five
525 out of the nine stations (“Barcelona”, “Dhadnah”, “Lecce University”, “Rome Tor
526 Vergata” and “Villefranche”) the temporal tendencies have the same sign for all three
527 databases, with AERONET showing larger trends. Moreover, the trends are
528 statistically significant at the 95% confidence level for “Barcelona” station.

529 The overall comparison between satellite and surface g_{aer} data performed in the
530 scatterplot of Fig. 9 and Table 1 does not allow one to have an insight to how the
531 comparison behaves spatially, namely how it differs from a region to another. This is
532 addressed in Fig. 12, showing the comparison of satellite and surface data at the
533 wavelength of 870 nm separately between MODIS-Terra - AERONET and MODIS-
534 Aqua – AERONET. For this comparison, we selected AERONET stations for which
535 there is satisfactory overlap between the time series from AERONET and the time
536 series from MODIS, namely the number of common days between AERONET-Terra

537 and AERONET-Aqua is larger than 100. This intentionally selected less strict
538 criterion that the one used in Fig. 11 is satisfied by 36 stations for AERONET-Terra
539 and by 34 for AERONET-Aqua. For each AERONET station we compute the Pearson
540 correlation coefficient between the station data and the corresponding MODIS-Terra
541 or Aqua data at 870 nm, for the $1^\circ \times 1^\circ$ cell containing the station. Moreover, there is
542 the information if the trends between AERONET and either MODIS-Terra or Aqua
543 have the same sign.

544 In the case of the $g_{\text{aer}}(\text{AERONET}) - g_{\text{aer}}(\text{Terra})$ comparison, at 5 stations the correlation
545 coefficient R is larger than 0.5, while at 21 stations $0.3 < R < 0.5$. The largest R found is
546 0.64 at station “Bahrain”. With respect to the agreement on the sign of the trends, at
547 24 out of 36 stations (67%) there is a trend sign match and at 12 stations (33%) a
548 mismatch. A similar picture emerges for the comparison $g_{\text{aer}}(\text{AERONET}) - g_{\text{aer}}(\text{Aqua})$. In
549 this case, there are again 5 stations with $R > 0.5$ (maximum value $R = 0.61$ again at
550 “Bahrain”), while at 19 stations $0.30 < R < 0.50$. Also, we see that at 22 stations there is
551 a trend sign match and at 12 there is a mismatch (respective percentages equal to 65%
552 and 35%).

553

554 **5 Summary and Conclusions**

555 Using satellite data from the latest available collection (051) of MODIS-Terra and
556 Aqua data, we examine the spatiotemporal variations of the aerosol asymmetry
557 parameter over North Africa, the Arabian peninsula and Europe. Generally, the largest
558 values of the asymmetry parameter, indicating the strongest forward scattering of
559 radiation by atmospheric aerosols, are found over areas with aerosol load being
560 dominated by large size particles of desert dust (tropical Atlantic, Arabian and Red
561 Seas). On the contrary, smaller g_{aer} values are seen where a significant fraction of
562 aerosol load comes from small size particles of anthropogenic origin, e.g. over the
563 Black Sea. The results are consistent with the theory and thus prove a good
564 performance of the MODIS retrieval of aerosol asymmetry parameter. Depending on
565 the area of interest, the seasonal cycle of the asymmetry parameter varies markedly.
566 More specifically, in areas with abundance of desert dust particles, the range of intra-
567 annual variation is small, with the largest values during summer, while in other areas
568 the seasonality is reversed, with the largest values during the cold season and the
569 smallest during the warm season. The asymmetry parameter decreases with
570 wavelength, especially when one examines its spatially minimum values, while this

571 decrease is weaker for the larger g_{aer} values, corresponding to the presence of coarser
572 particles.

573 The seasonal fluctuation is more pronounced with increasing wavelength in the
574 examined regions, which is attributed to the different spectral behaviour of the
575 asymmetry parameter for small and large particles. With respect to the inter-annual
576 variability of the asymmetry parameter, we did not discern very important either
577 increasing or decreasing tendencies, with absolute changes smaller than 0.04 in any
578 case. On the other hand, we found opposing tendencies for the two satellite datasets.
579 MODIS-Terra observes mostly increasing tendencies, while Aqua gives extensive
580 regions with decreasing tendencies. Generally, the largest intra-annual and inter-
581 annual variations are seen over the Black Sea, while the smallest over the tropical
582 Atlantic. However, some strong trends (especially from Terra) may be due to
583 calibration drift errors, which may be addressed in collection 006. Along these lines
584 we performed some preliminary comparisons between 051 and 006 Angstrom
585 Exponent trends from Aqua, which ensured that AE and g are very closely anti-
586 correlated. These preliminary results, show that 051 Aqua AE trends resemble very
587 closely the 006 trends, supporting that the g trends from collection 051 (at least for
588 Aqua) reported in this study are credible.

589 We compare satellite data with surface data from the AERONET, in order to validate
590 the reliability of the former. The quantitative comparison is very useful, since satellite
591 data provide broad geographical coverage and are very important in any study related
592 to aerosols and their climate impact. The disagreement with surface stations can give
593 insights in the resulting errors. Through the examination of frequency distributions of
594 daily g_{aer} , a shift of satellite data towards larger values relative to surface data
595 becomes apparent. This finding is more pronounced for g_{aer} over Europe, while the
596 North African, Arabian peninsula values are more in agreement. Moreover, the
597 smallest g_{aer} values originate from particles from Europe, because of the generation of
598 smaller size particles by industrial activities and biomass burning.

599 In this work we present scatter plots of daily g_{aer} values between MODIS-Terra,
600 MODIS-Aqua, and AERONET, which show moderate agreement between satellite
601 data at 470 nm and surface data at 440 nm, with small correlation coefficients
602 ($R < 0.3$). Slightly better agreement was noted at larger wavelengths, but still without
603 reaching very satisfactory levels ($R < 0.47$). Nevertheless, during spring and summer,
604 satellite and surface measurements tend to agree more. Finally, for the comparisons at
605 660/675 and 870 nm, we report an overestimation of g_{aer} by MODIS compared to

606 AERONET, as expected because of the less steep decrease of g_{aer} with wavelength of
607 MODIS.

608 We extract pairs of daily Terra-AERONET and Aqua-AERONET values at stations
609 with at least 100 common days. At 21 of 36 stations (Terra-AERONET comparison)
610 and at 19 of 34 stations (Aqua-AERONET comparison) we derive $0.3 < R < 0.5$, while
611 at 5 stations in both cases, the correlation coefficients are larger than 0.5. Finally, as
612 far as the signs of temporal trends are concerned, we determine agreement in 67%
613 (Terra-AERONET comparison) and in 65% of stations (Aqua-AERONET
614 comparison).

615 The results of the present analysis are useful since they assess for the first time the
616 performance of satellite based products of aerosol asymmetry parameter over broad
617 regions of special climatic interest. Our results can offer an interesting way to assess
618 the uncertainty induced by the use of such satellite g_{aer} data in climate and radiative
619 transfer models that compute aerosol radiative and climate effects. The obtained
620 results are relatively satisfactory given the difficulties encountered by satellite
621 retrieval algorithms due to the different assumptions they made. The identified
622 weaknesses may provide an opportunity to improve such satellite retrievals of aerosol
623 asymmetry parameter in forthcoming data products like those of MODIS C006.

624

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634

635 **References**

- 636 Barnaba F. and G. P. Gobbi: Aerosol seasonal variability over the Mediterranean region and
637 relative impact of maritime, continental and Saharan dust particles over the basin from
638 MODIS data in the year 2001, *Atmospheric Chemistry and Physics*, 4, 4285 - 4337, SRef-
639 ID: 1680-7375/acpd/2004-4-4285SRef-ID: 1680-7375/acpd/2004-4-4285, 2004.
- 640 Chubarova, N. Y.: Seasonal distribution of aerosol properties over Europe and their impact on
641 UV irradiance, *Atmos. Meas. Tech.*, 2, 593–608, doi:10.5194/amt-2-593-2009, 2009.
- 642 Di Iorio, T., A. di Sarra, W. Junkermann, M. Cacciani, G. Fiocco, and D. Fua, Tropospheric
643 aerosols in the Mediterranean: 1. Microphysical and optical properties, *J. Geophys. Res.*,
644 108(D10), 4316, doi:10.1029/2002JD002815, 2003
- 645 Dubovik, Oleg, Brent Holben, Thomas F. Eck, Alexander Smirnov, Yoram J. Kaufman,
646 Michael D. King, Didier Tanré, Ilya Slutsker, 2002: Variability of Absorption and Optical
647 Properties of Key Aerosol Types Observed in Worldwide Locations. *J. Atmos. Sci.*, **59**,
648 590–608.
- 649 Dubovik, O. and M. D. King, 2000: A flexible inversion algorithm for retrieval of aerosol
650 optical properties from Sun and sky radiance measurements," *J. Geophys. Res.*, 105, 20
651 673-20 696.
- 652 Fotiadi, A., E. Drakakis, N. Hatzianastassiou, C. Matsoukas, K. G. Pavlakis, D.
653 Hatzidimitriou, E. Gerasopoulos, N. Mihalopoulos, and I. Vardavas (2006), Aerosol
654 physical and optical properties in the eastern Mediterranean Basin, Crete, from Aerosol
655 Robotic Network data, *Atmos. Chem. Phys.*, 6, 5399– 5413.
- 656 Gerasopoulos, E., Kouvarakis, G., Babasakalis, P., Vrekoussis, M., Putaud, J. P., and
657 Mihalopoulos, N.: Origin and variability of particulate matter (PM10) mass concentrations
658 over the Eastern Mediterranean, *Atmos. Environ.*, 40, 4679–4690, 2006.
- 659 Gkikas, A., Hatzianastassiou, N., and Mihalopoulos, N.: Study and characterization of aerosol
660 episodes in the Mediterranean basin for the 7-year period 2000–2007 based on MODIS
661 data, European Aerosol Conference, Greece, Thessaloniki, 24–29 August 2008.
- 662 Gkikas, A., Hatzianastassiou, N., and Mihalopoulos, N.: Aerosol events in the broader
663 Mediterranean basin based on 7-year (2000–2007) MODIS C005 data, *Ann. Geophys.*, 27,
664 3509–3522, doi:10.5194/angeo-27-3509-2009, 2009.
- 665 Gkikas, A., Houssos, E., Hatzianastassiou, N., Papadimas, C. and Bartzokas, A. (2011),
666 Synoptic conditions favouring the occurrence of aerosol episodes over the broader
667 Mediterranean basin. *Q.J.R. Meteorol. Soc.*.. doi: 10.1002/qj.978
- 668 Goloub, P., and O. Arino, 2000: Verification of the consistency of POLDER aerosol index
669 over land with ATSR-2 fire product. *Geophys. Res. Lett.*, 27, 899–902.
- 670 Graßl, H.: Possible changes of planetary albedo due to aerosol particles, in *Man's Impact on*
671 *Climate*, edited by: W. Bach, J. Pankrath, and W. Kellogg, Elsevier, New York, 1979.

- 672 Hansen, J., Sato, M., and Ruedy, R.: Radiative forcing and climate response, *J. Geophys.*
673 *Res.*, 102, 6831–6864, 1997.
- 674 Hatzianastassiou, N., B. Katsoulis, I. Vardavas: Sensitivity analysis of aerosol direct radiative
675 forcing in ultraviolet - visible wavelengths and consequences for the heat budget, *Tellus*,
676 56b, 368 - 381, 2004.
- 677 Hatzianastassiou, N., A. Gkikas, N. Mihalopoulos, O. Torres, and B. D. Katsoulis: Natural
678 versus anthropogenic aerosols in the eastern Mediterranean basin derived from multiyear
679 TOMS and MODIS satellite data, *J. Geophys. Res.*, 114, D24202,
680 doi:10.1029/2009JD011982, 2009.
- 681 Hatzianastassiou, N., Matsoukas, C., Drakakis, E., Stackhouse Jr., P. W., Koepke, P.,
682 Fotiadi, A., Pavlakis, K. G., and Vardavas, I.: The direct effect of aerosols on solar
683 radiation based on satellite observations, reanalysis datasets, and spectral aerosol optical
684 properties from Global Aerosol Data Set (GADS), *Atmos. Chem. Phys.*, 7, 2585-2599,
685 doi:10.5194/acp-7-2585-2007, 2007.
- 686 Haywood, J.M., and O. Boucher, 2000: Estimates of the direct and indirect radiative forcing
687 due to tropospheric aerosols: A review. *Rev. Geophys.*, 38, 513–543.
- 688 Heishman, J. (1999), Commanding Officer, Forecaster's Handbook, U.S. Navy Cent.
689 Meteorol. and Oceanogr. Cent., Manama, Bahrain.
- 690 Holben B.N., T.F. Eck, I. Slutsker, D. Tanré, J.P. Buis, A. Setzer, E. Vermote, J.A. Reagan,
691 Y. Kaufman, T. Nakajima, F. Lavenu, I. Jankowiak, and A. Smirnov, 1998: AERONET -
692 A federated instrument network and data archive for aerosol characterization, *Rem. Sens.*
693 *Environ.*, 66, 1-16.
- 694 Ichoku C., D. Allen Chu, Shana Mattoo, Yoram J. Kaufman, Lorraine A. Remer, Didier
695 Tanre', Ilya Slutsker, and Brent N. Holben: A spatio-temporal approach for global
696 validation and analysis of MODIS aerosol products. *GEOPHYSICAL RESEARCH*
697 *LETTERS*, VOL. 29, NO. 12, 10.1029/2001GL013206, 2002.
- 698 IPCC, 2007: *Climate Change 2007: The Physical Science Basis*. Contribution of Working
699 Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate
700 Change [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor
701 and H.L. Miller (eds.)]. Cambridge University Press, Cambridge, United Kingdom and
702 New York, NY, USA.
- 703 IPCC, 2013: *Climate Change 2013: The Physical Science Basis*. Contribution of Working
704 Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change
705 [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y.
706 Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United
707 Kingdom and New York, NY, USA, 1535 pp.
- 708 Jianrong Bi, Jianping Huang, Qiang Fu, Xin Wang, Jinsen Shi, Wu Zhang, Zhongwei
709 Huang, Beidou Zhang: Toward characterization of the aerosol optical properties over

- 710 Loess Plateau of Northwestern China. *Journal of Quantitative Spectroscopy & Radiative*
711 *Transfer* 112 (2011) 346–360
- 712 Kalivitis, N., Gerasopoulos, E., Vrekoussis, M., Kouvarakis, G., Kubilay, N.,
713 Hatzianastassiou, N., Vardavas, I., and Mihalopoulos, N.: Dust transport over the eastern
714 Mediterranean derived from TOMS, AERONET and surface measurements, *J. Geophys.*
715 *Res.*, 112, D03202, doi:10.1029/2006JD007510, 2007.
- 716 Kaufman, Y. J., D. Tanré, L. A. Remer, E. F. Vermote, A. Chu, and B. N. Holben:
717 Operational remote sensing of tropospheric aerosol over land from EOS moderate
718 resolution imaging spectroradiometer, *J. Geophys. Res.*, 102, 17,051– 17,067, 1997.
- 719 Kinne, S., D. O’Donnell, P. Stier, S. Kloster, K. Zhang, H. Schmidt, S. Rast, M. Giorgetta, T.
720 F. Eck, and B. Stevens (2013), MAC-v1: A new global aerosol climatology for climate
721 studies, *J. Adv. Model. Earth Syst.*, 5, 704–740, doi:10.1002/jame.20035.
- 722 Koepke, P., M. Hess, I. Schult, and E. P. Shettle: Global aerosol data set, Rep. No. 243, Max-
723 Planck Institut fuer Meteorologie, Hamburg, Germany, 44 pp., 1997.
- 724 Lelieveld, J., et al. (2002), Global air pollution crossroads over the Mediterranean, *Science*,
725 298, 794– 799, doi:10.1126/science.1075457.
- 726 Lohmann U., Feichter, J. [2005] Global indirect aerosol effects: A review . *Atmos. Chem.*
727 *Phys.*, 5, 715-737
- 728 Lyamani, H., F. J. Olmo, A. Alca´ntara, and L. Alados-Arboledas (2006), Atmospheric
729 aerosols during the 2003 heat wave in southeastern Spain. I: Spectral optical depth, *Atmos.*
730 *Environ.*, 40, 6453 – 6464, doi:10.1016/ j.atmosenv.2006.04.048.
- 731 Papadimas, C. D., N. Hatzianastassiou, N. Mihalopoulos, X. Querol, and I.
732 Vardavas (2008), Spatial and temporal variability in aerosol properties over the
733 Mediterranean basin based on 6-year (2000–2006) MODIS data, *J. Geophys. Res.*, 113,
734 D11205, doi:10.1029/2007JD009189.
- 735 Pace, G., A. di Sarra, Meloni, D., Piacentino, S., and Chamard, P.: Aerosol optical properties
736 at Lampedusa (Central Mediterranean). 1. Influence of transport and identification of
737 different aerosol types, *Atmos. Chem. Phys.*, 6, 697–713, 2006, www.atmos-chem-
738 phys.net/6/697/2006/.
- 739 Pandithurai, R. T. Pinker, P. C. S. Devara, T. Takamura, K. K. Dani, Seasonal asymmetry in
740 diurnal variation of aerosol optical characteristics over Pune, western India, *J. Geophys.*
741 *Res.*, 112, D8, DOI: 10.1029/2006JD007803, 2007.
- 742 Prospero, J., P. Ginoux, O. Torres, and S. E. Nicholson (2002), Environmental
743 Characterization of Global sources of atmospheric soil dust derived from the NIMBUS-7
744 TOMS absorbing aerosol product, *Rev. Geophys.*, 40(1), 1002,
745 doi:10.1029/20000GR000095.
- 746 Remer L.A., Kaufman Y.J., Tanre D. and co-authors: The MODIS aerosol algorithm,
747 products, and validation, *J. Atmos. Sci.*, 62: 947-973, 2005.

- 748 Remer LA, Kleidman RG, Levy RC, Kaufman YJ, Tanre D, Mattoo S, Martins JV, Ichoku
749 C, Koren I, Yu H, Holben BN. 2008. Global aerosol climatology from the MODIS satellite
750 sensors. *Journal of Geophysical Research* 113: D14S07, DOI: 10.1029/2007JD009661.
- 751 Satheesh, S. K., V. Ramanathan, X. Li-Jones, J. M. Lobert, I. A. Podgorny, J. M. Prospero, B.
752 N. Holben, and N. G. Loeb, A model for the natural and anthropogenic aerosols over the
753 tropical Indian Ocean derived from Indian Ocean Experiment data, *J. Geophys. Res.*, 104,
754 27,421–27,440, 1999.
- 755 Savoie, D. L., J. M. Prospero, and R. T. Nees, Nitrate, nonsea-salt sulfate, and mineral
756 aerosol over the northwestern Indian Ocean, *J. Geophys. Res.*, 92, 933–942, 1987.
- 757 Sciare, J., H. Bardouki, C. Moulin, and N. Mihalopoulos (2003), Aerosol sources and their
758 contribution to the chemical composition of aerosols in the Eastern Mediterranean Sea
759 during summertime, *Atmos. Chem. Phys.*, 3, 291–302, SRef-ID:1680 – 7324/acp/2003–3-
760 291.
- 761 Smirnov, A., B. N. Holben, Y. J. Kaufman, O. Dubovik, T. F. Eck, I. Slutsker, C. Pietras, and
762 R. N. Halthore, 2002b: Optical properties of atmospheric aerosol in maritime
763 environments. *J. Atmos. Sci.*, **59**, 501–523.
- 764 Smirnov, and Coauthors, 2002a: Atmospheric aerosol optical properties in the Persian Gulf. *J.*
765 *Atmos. Sci.*, 59, 620–634.
- 766 Smirnov, A., B. N. Holben, T. F. Eck, I. Slutsker, B. Chatenet, and R. T. Pinker, 2002c:
767 Diurnal variability of aerosol optical depth observed at AERONET (Aerosol Robotic
768 Network) sites, *Geophys. Res. Lett.*, **29**, 23, 2115, doi:10.1029/2002GL016305.
- 769 Tanré, D., Y. J. Kaufman, M. Herman, and S. Mattoo: Remote sensing of aerosol properties
770 over oceans using the MODIS/EOS spectral radiances, *J. Geophys. Res.*, 102, 16,971–
771 16,988, 1997.
- 772 Tindale, N. W., and P. P. Pease, Aerosols over the Arabian Sea: Atmospheric transport
773 pathways and concentrations of dust and sea salt, *Deep Sea Res.*, 46, 1577–1595, 1999.
- 774 Yu, H., Dickinson, R. E., Chin, M., Kaufman, Y. J., Holben, B. N. Geogdzhayev, I. V., and
775 Mishchenko, M. I.: Annual cycle of global distributions of aerosol optical depth from
776 integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, 108,
777 4128, doi:10.1029/2002JD002717, 2003.
- 778 Zdun, A., A. Rozwadowska and S. Kratzer, 2011. Seasonal variability in the optical
779 properties of Baltic aerosols, *Oceanologia*, 53(1), 7-34.
- 780
781

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

789

790 *MODIS-Terra*

791

		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

792

793

794 *MODIS-Aqua*

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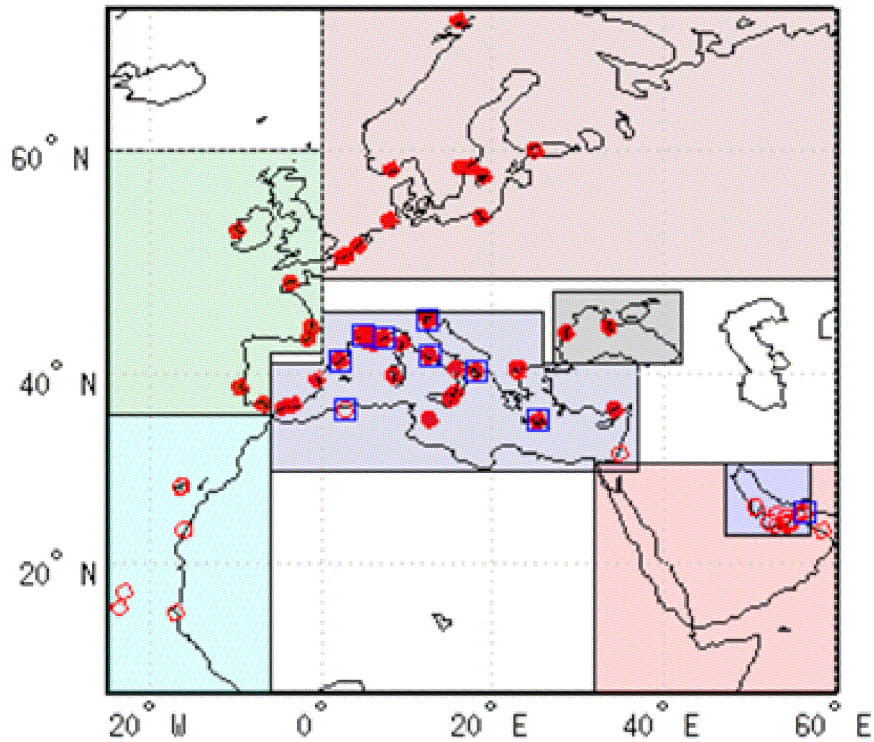
		R	Bias*	RMSE	Slope	Intercept
	470-440	0.27	0.018	0.047	0.41	0.40
	660-675	0.42	-0.005	0.062	0.61	0.26
	870	0.46	-0.015	0.072	0.61	0.26
W i n	470-440	0.25	0.024	0.049	0.36	0.43

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

t e r						
	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 tu mn	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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* $g_{aer}(AERONET) - g_{aer}(MODIS)$



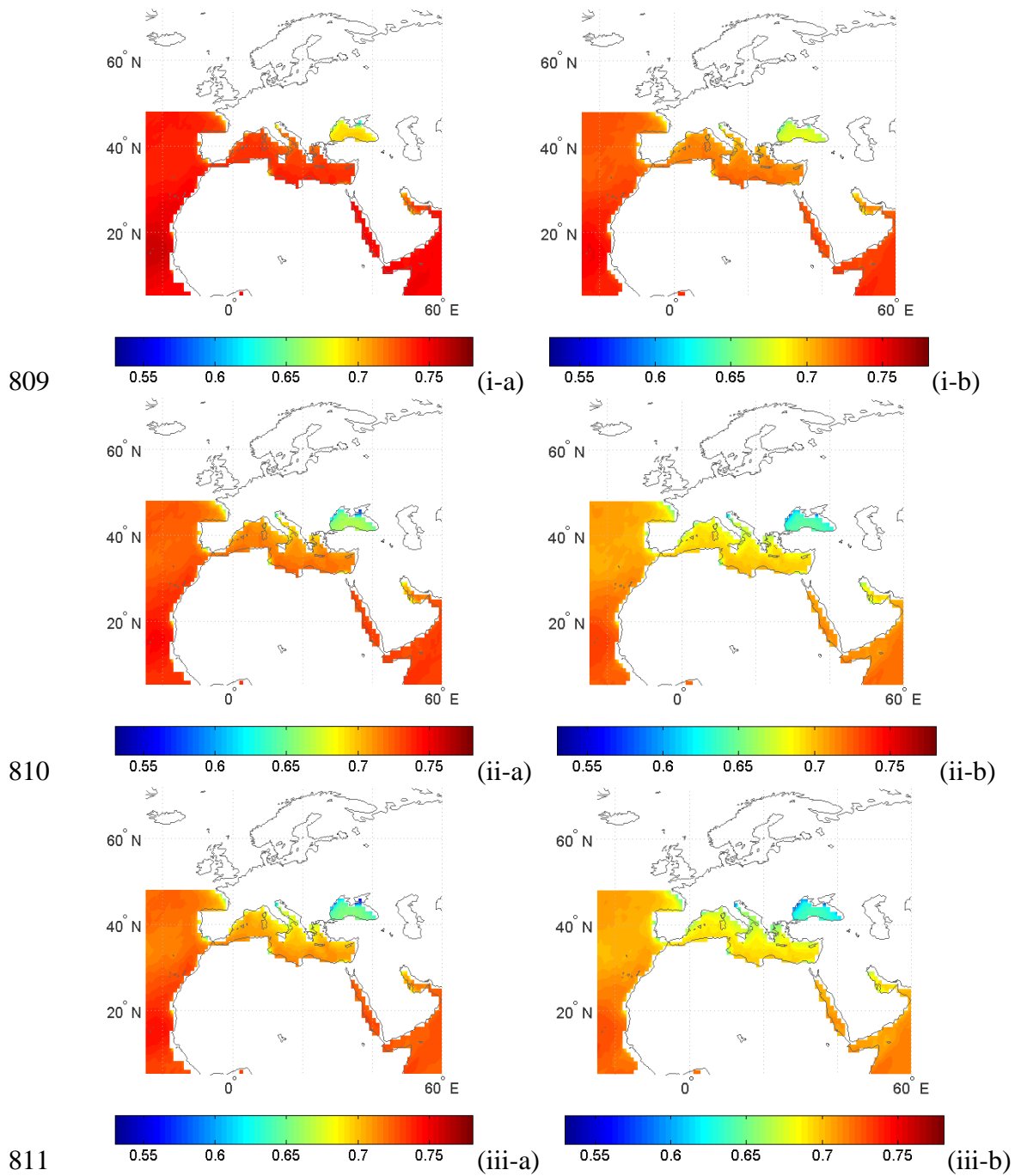
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800

801 **Figure 1.** Comparison between HAC and MODIS total aerosol optical depth at
 802 550nm. Global seasonal distribution of relative percentage differences ((HAC-
 803 MODIS)/MODIS -%) for: (a) winter (December-January-February), (b) spring
 804 (March-April-May), (c) summer (June-July-August) and (d) autumn (September-
 805 October-November). White shaded areas correspond to cases for which MODIS AOD
 806 values are missing or do not qualify for the averaging threshold.

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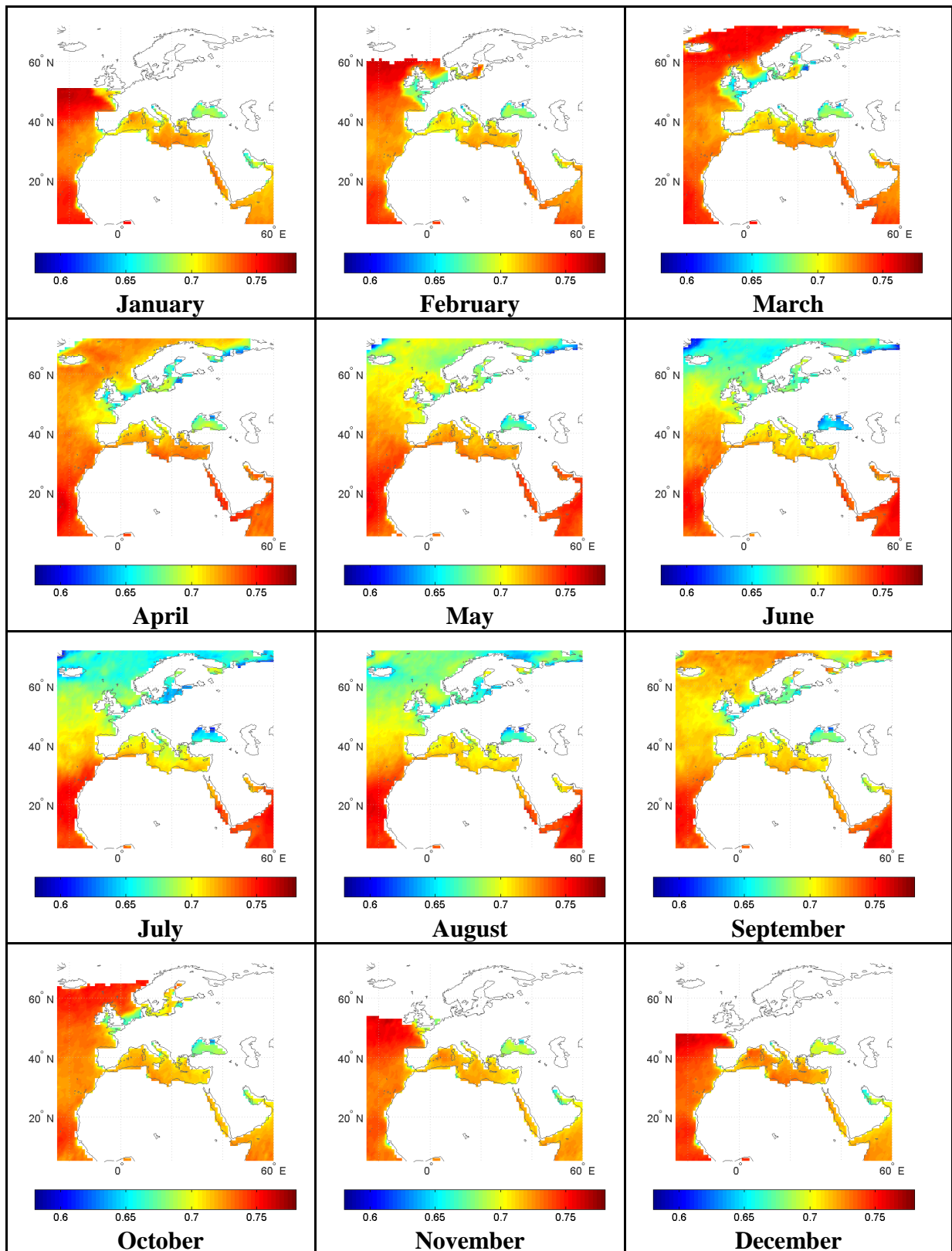
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812

813 **Figure 2.** Geographical distribution of MODIS-Terra (-a, left column) and MODIS-
 814 Aqua (-b, right column) g_{aer} values averaged over 2002-2010, at the wavelengths of:
 815 470 nm (i-, top row), 660 nm (ii-, middle row) and 870 nm (iii-, bottom row).

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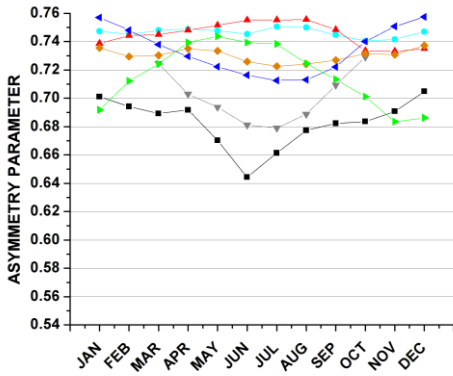


817

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

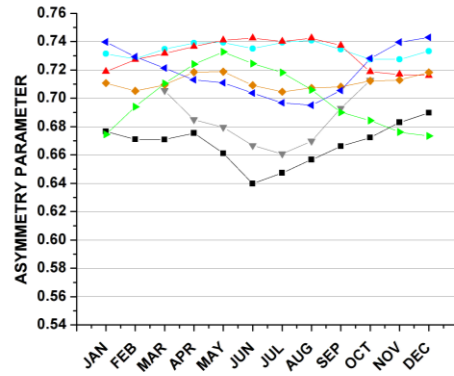
819 over the period 2002-2010.

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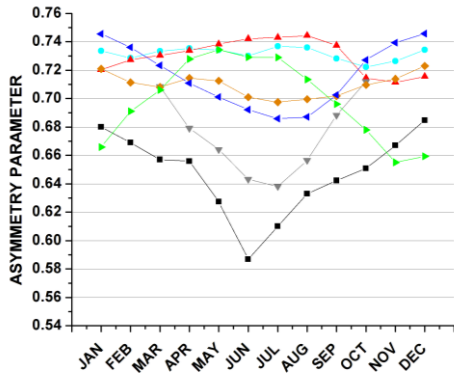


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(i-a)

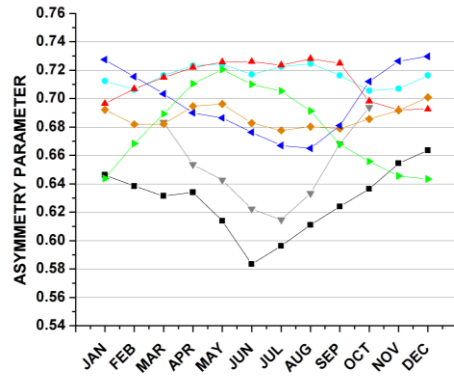


(i-b)

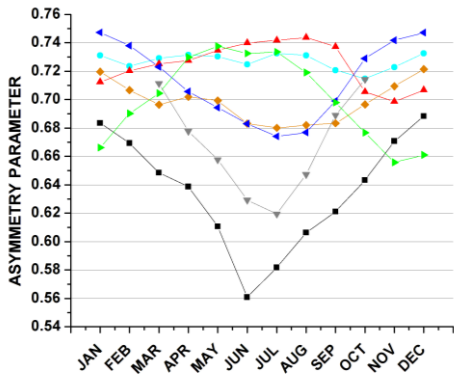


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(ii-a)

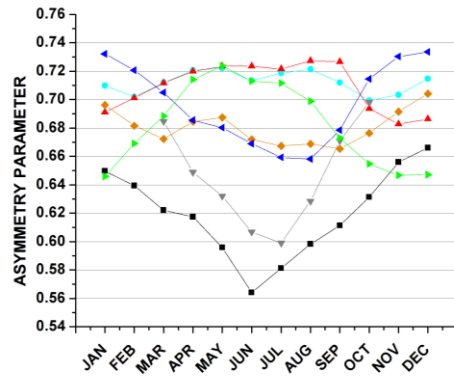


(ii-b)



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(iii-a)



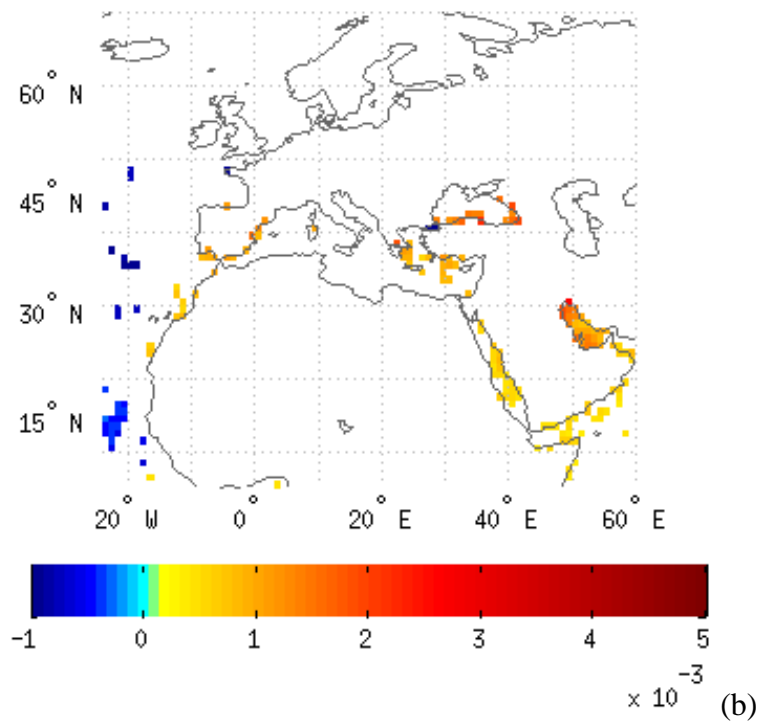
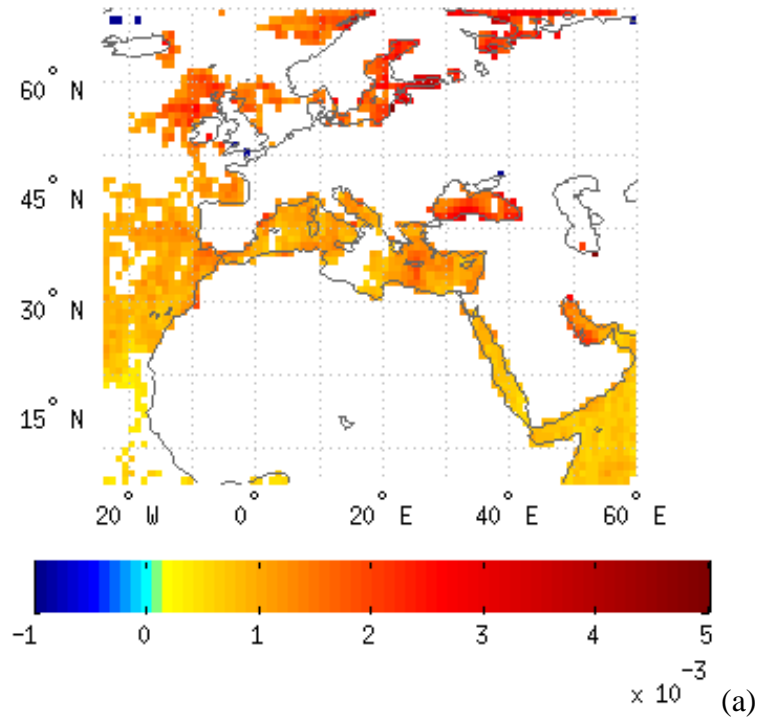
(iii-b)



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825 **Figure 4.** Intra-annual variation of MODIS Terra (-a, left column) and Aqua (-b, right
 826 column) g_{aer} values averaged over seven selected sub-regions (Fig. 1). Results are
 827 given for g_{aer} values at: 470 nm (i-, top row), 660 nm (ii-, middle row) and 870 nm
 828 (iii-, bottom row), averaged over the period 2002-2010, respectively.

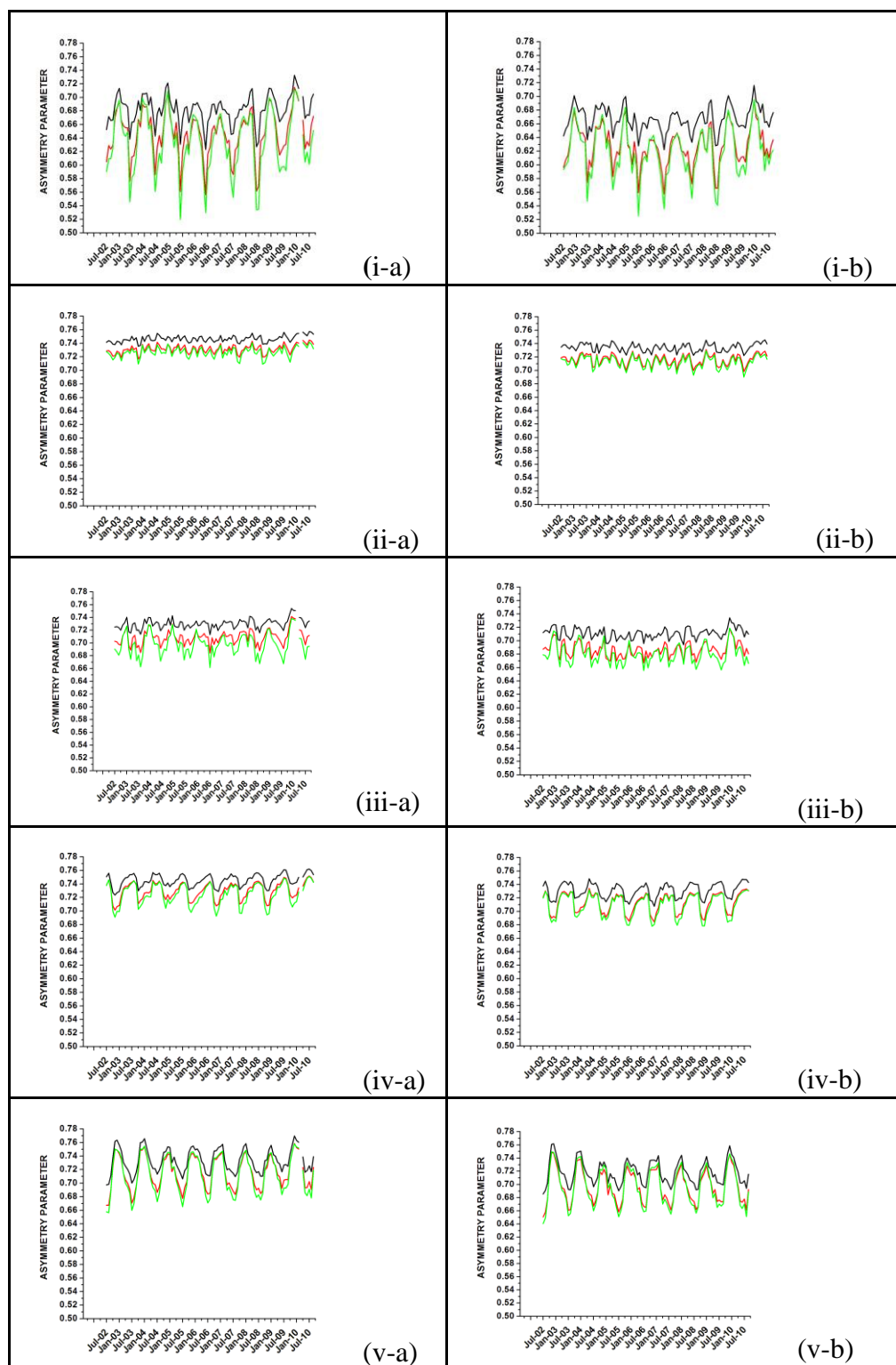
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833 **Figure 5.** Slope (in units decade⁻¹) of MODIS g_{aer} deseasonalized anomalies over the
834 period 2002-2010 from MODIS-Terra (-a, top) and MODIS-Aqua (-b, bottom), for
835 the wavelengths of 470 nm.

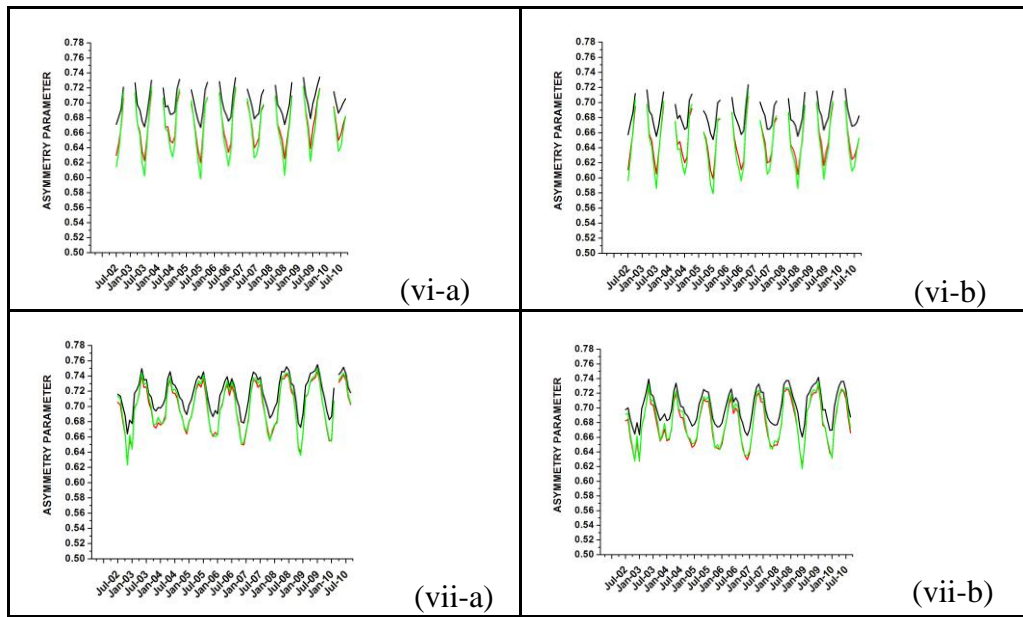
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840 **Figure 6.** Inter-annual (2002-2010) variation of monthly mean g_{aer} values at 470 nm
 841 averaged over the sub-regions of: (i) Black Sea, (ii) Eastern Atlantic Ocean, (iii)
 842 Mediterranean Sea, (iv) Middle East, (v) North-eastern Atlantic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

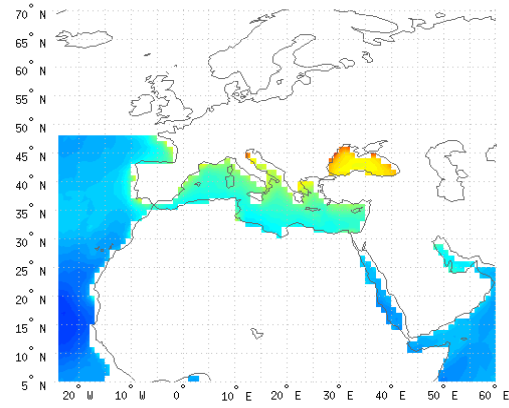
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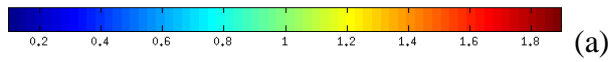
847

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

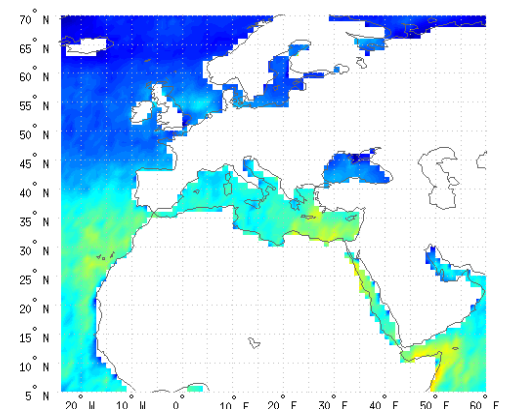
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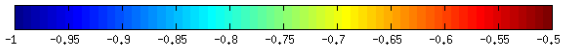
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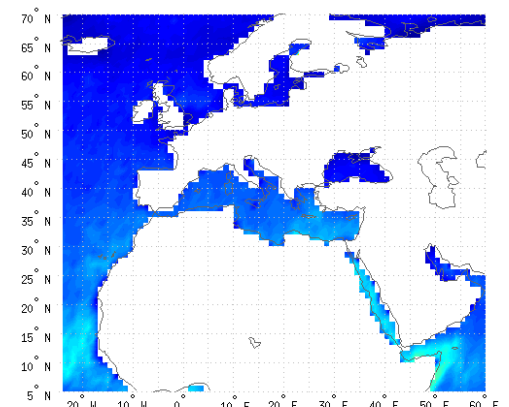
(a)



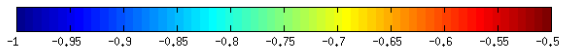
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(b)



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(c)

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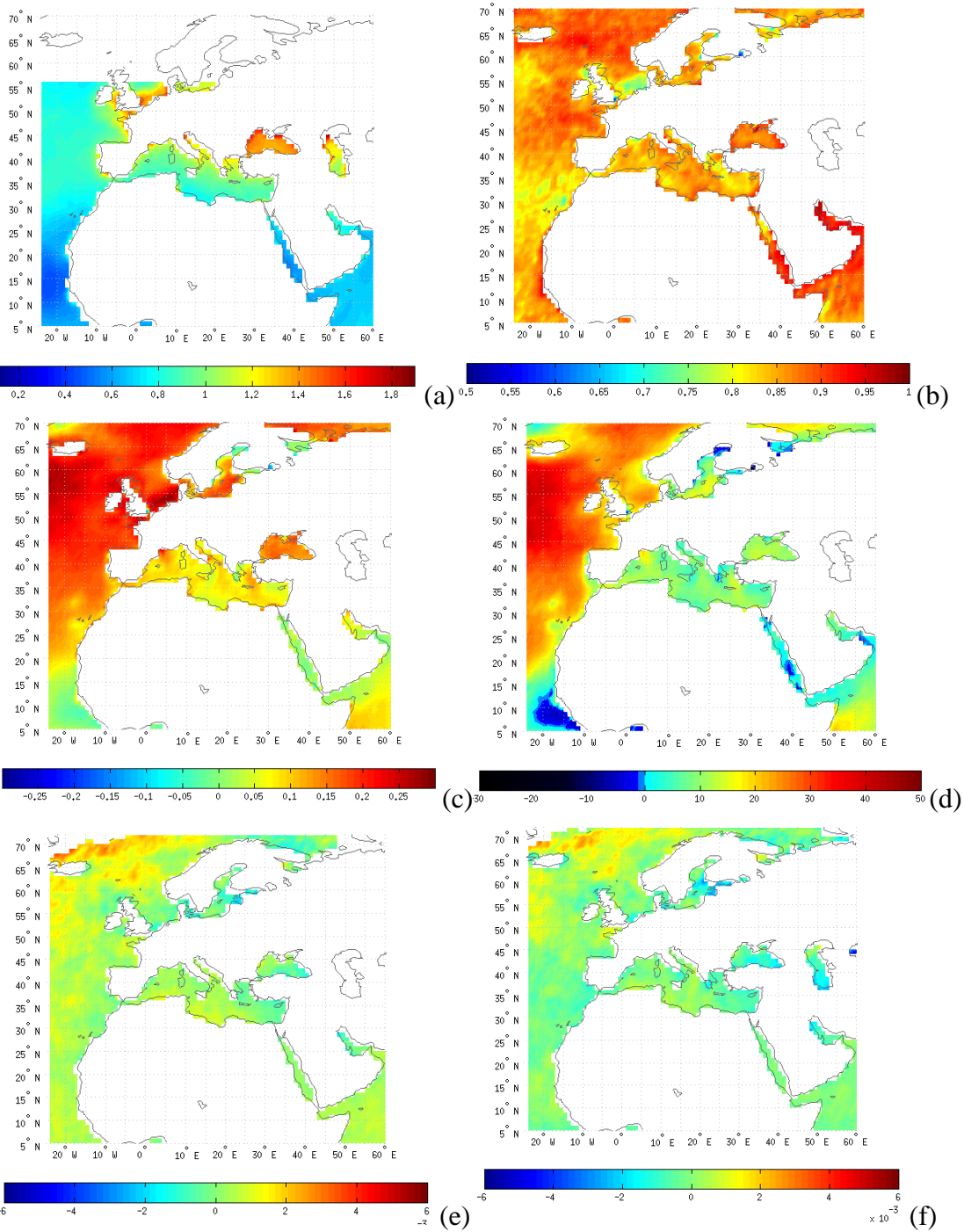
858 **Figure 7.** Geographical distribution of MODIS-Aqua C005 Angström exponent

859 ($AE_{550-865}$) values averaged over 2002-2010, at the wavelength pair of 550-865 nm.

860 The correlation coefficients between $AE_{550-865}$ and g_{aer} data at 660 and 870 nm are

861 given in (b) and (c), respectively.

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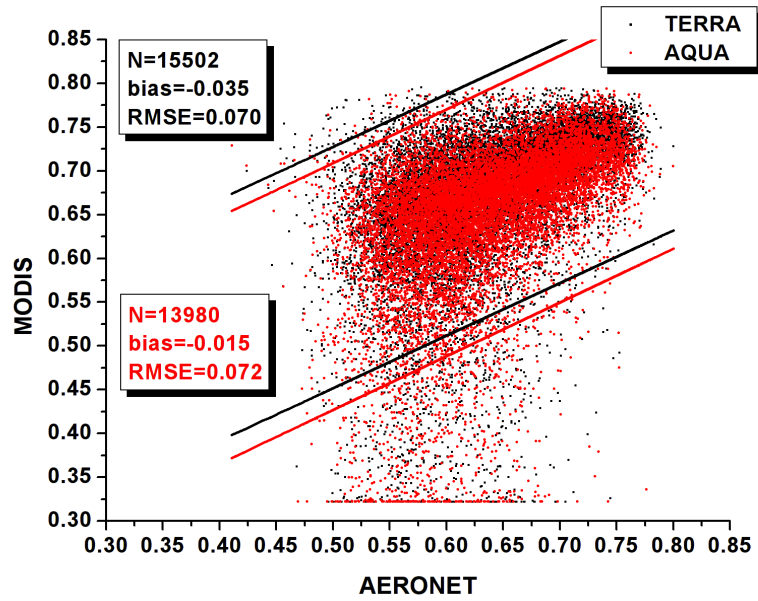
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867 **Figure 8.** Geographical distribution of MODIS-Aqua C006 Angström exponent
 868 ($AE_{550-865}$) values averaged over 2002-2010, at the wavelength pair of 550-865 nm. In
 869 (b), (c) and (d) are given the correlation coefficients, the absolute biases and the
 870 relative percent biases, respectively, between the C006 and corresponding C005
 871 $AE_{550-865}$ data. In (e) and (f) are given the computed deseasonalized trends of
 872 MODIS Aqua C005 and C006 $AE_{550-865}$ slope values for years 2002-2010,
 873 respectively.

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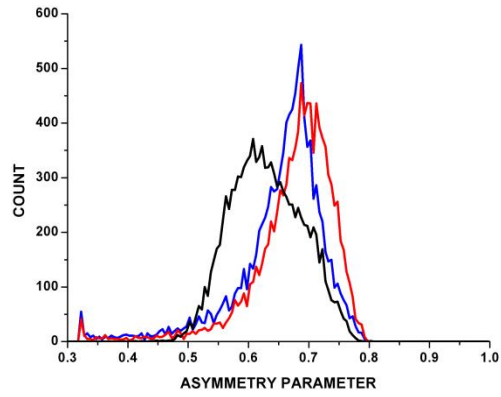
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877 **Figure 9.** Scatterplot comparison between g_{aer} values at 870 nm from MODIS Terra

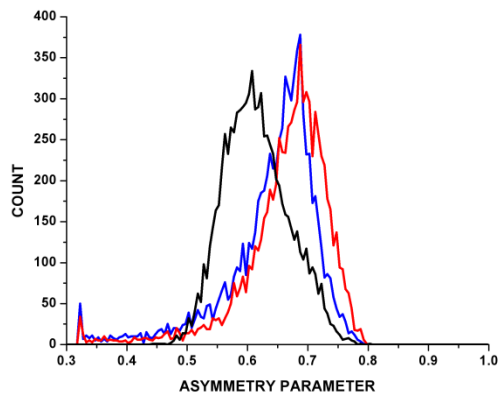
878 (black color) and Aqua (red color) and corresponding values from AERONET stations

879 (blue squares, Fig. 1). The 95% prediction bands as well as the mean bias and root

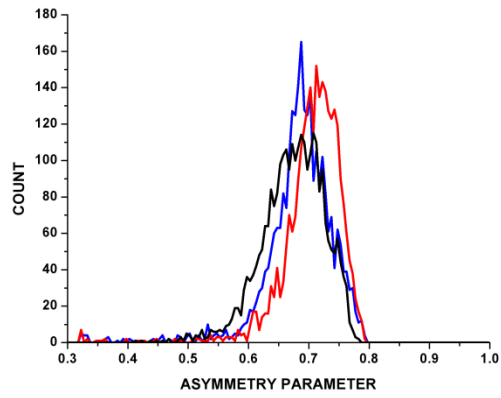
880 mean squared error are given.



(a)

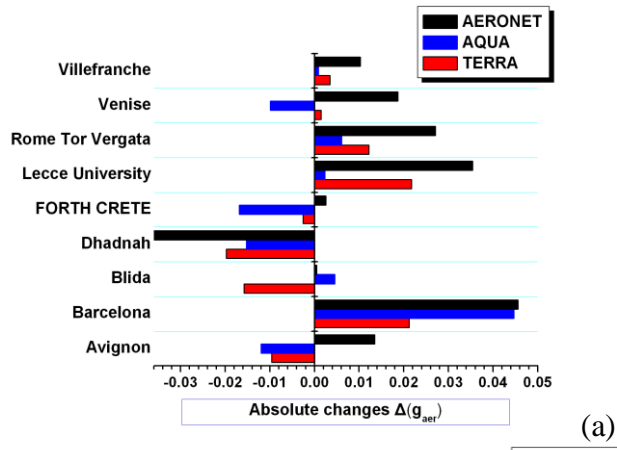


(b)

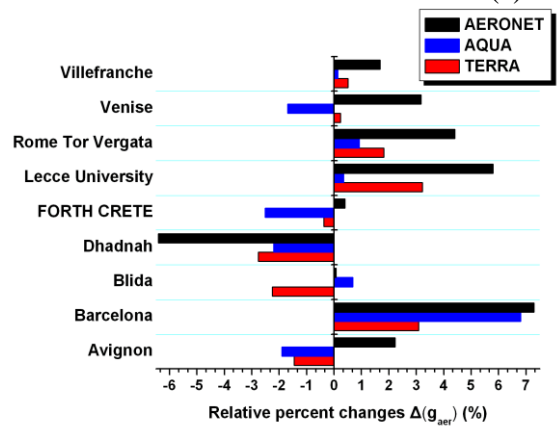


(c)

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.



(a)



(b)

Figure 11. 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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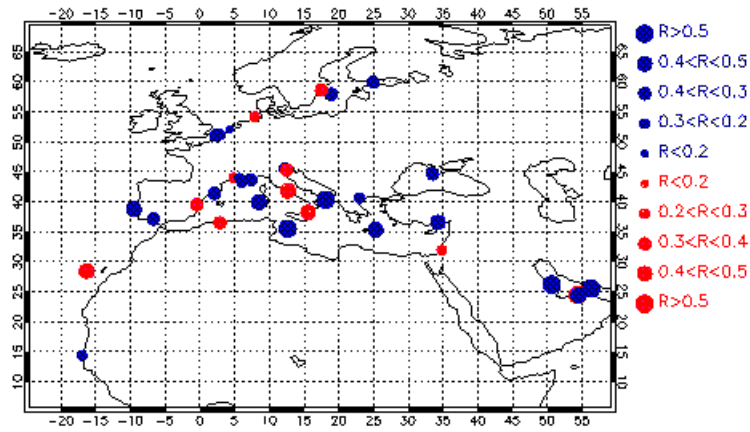
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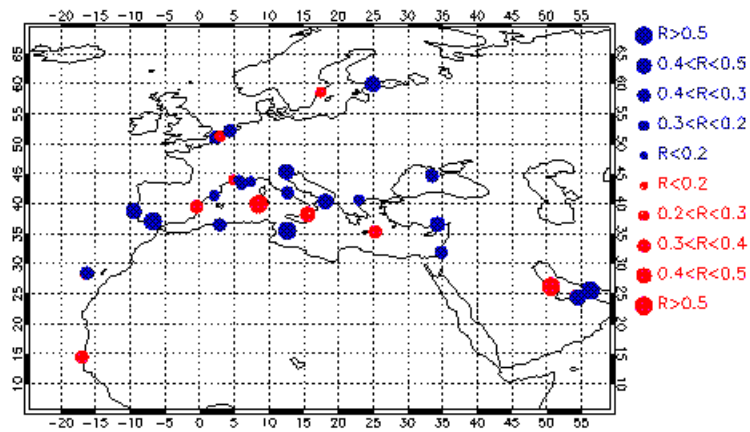
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(a)



(b)

Figure 12. 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.