

1 A Study of 15-Year Aerosol Optical Thickness and Direct Shortwave Aerosol
2 Radiative Effect Trends Using MODIS, MISR, CALIOP and CERES

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19 **Abstract**

20 By combining Collection 6 Moderate Resolution and Imaging Spectroradiometer (MODIS)
21 and Version 22 Multi-angle Imaging Spectroradiometer (MISR) aerosol products with Cloud and
22 Earth's Radiant Energy System (CERES) flux products, the aerosol optical thickness (AOT, at
23 $0.55\mu\text{m}$) and Short-Wave Aerosol Radiative Effect (SWARE) trends are studied over ocean for
24 the near full Terra (2000-2015) and Aqua (2002-2015) data records. Despite differences in
25 sampling methods, regional SWARE and AOT trends are highly correlated with one another. Over
26 global oceans, weak SWARE (cloud free SW flux) and AOT trends of $0.5 - 0.6 \text{ Wm}^{-2}$ (-0.5 to -0.6
27 Wm^{-2}) and $0.002 \text{ AOT decade}^{-1}$ were found using Terra data. Near zero AOT and SWARE trends
28 are also found for using Aqua data, regardless of Angular Distribution Models (ADMs) used.
29 Regionally, positive SWARE and AOT trends are found over the Bay of Bengal, Arabian Sea,
30 Arabian/Persian Gulf and the Red Sea, while statistically significant negative trends are derived
31 over the Mediterranean Sea and the eastern US coast. In addition, the global mean instantaneous
32 SW aerosol direct forcing efficiencies are found to be $\sim -60 \text{ Wm}^{-2}$ per AOT, with corresponding
33 SWARE values of $\sim -7 \text{ Wm}^{-2}$ from both Aqua and Terra data, and again, regardless of CERES
34 ADMs used. Regionally, SW aerosol direct forcing efficiency values of $\sim -40 \text{ Wm}^{-2}$ per AOT are
35 found over the southwest coast of Africa where smoke aerosol particles dominate in summer.
36 Larger (in magnitude) SW aerosol direct forcing efficiency values of -50 to -80 Wm^{-2} per AOT
37 are found over several other dust and pollutant aerosol dominated regions. Lastly, the AOT and
38 SWARE trends from this study are also inter-compared with aerosol trends (such as active-based)
39 from several previous studies. Findings suggest that a cohesive understanding of the changing
40 aerosol skies can be achieved through the analysis of observations from both passive- and active-
41 based analyses, as well as at both narrow-band and broad-band data sets.

42 **1. Introduction**

43 The significance of aerosol particles on global and regional climate variations has been
44 extensively studied for the past two decades with both observational- and modeling-based
45 approaches (IPCC, 2013). In particular, studies have suggested that the direct shortwave (SW)
46 Aerosol Radiative Effect (SWARE), which refers to the impacts of aerosol particles on Earth's
47 radiation balance through the absorption and scattering of incoming SW solar energy, can be
48 estimated with the combined use of broadband and narrowband observations at the shortwave
49 spectrum (e.g. Zhang et al., 2005a;b; Loeb and Kato, 2002). For example, using one year of
50 collocated Terra Moderate Resolution Imaging Spectroradiometer (MODIS) and Cloud and
51 Earth's Radiant Energy System (CERES) data, Zhang et al., (2005b) derived the spatial
52 distribution of SWARE over global oceans. In that study, the perturbations in Top-of-Atmosphere
53 (TOA) SW energy due to aerosol particles are estimated using Terra CERES observations. The
54 Terra CERES observations have a large footprint of ~20 km at nadir (Wielicki et al., 1996). Thus,
55 collocated finer resolution Terra MODIS observations are used for cloud-clearing and reporting
56 finer scale aerosol optical properties within the CERES field of views (Christopher and Zhang,
57 2002b; Zhang et al., 2005a;b).

58 Terra MODIS, CERES, and Multi-Angle Imaging Spectroradiometer (MISR; Kahn et al.,
59 2010) instruments have been continuously observing Earth's atmosphere for more than 16 years
60 (2000-2016). Similarly, the MODIS and CERES instruments on board the Aqua satellite have
61 also been in operation for 14 years (2002-2016). Taking advantage of these longer term datasets
62 from the Aqua and Terra satellites, several studies have already examined temporal variations in
63 AOT both on regional and global scales (e.g., Zhang and Reid, 2010, Hsu et al., 2012; Li et al.,
64 2014; Alfaro-Contreras, 2016, Toth et al., 2016). For example, using 10 years (2000-2009) of
65 Collection 5 (C5) Terra and Aqua MODIS Dark Target (DT) AOT data, Zhang and Reid, (2010)

66 found a negligible AOT trend over global oceans, but documented three regions with statistically
67 significant increases in aerosol loadings, including the Indian Bay of Bengal, the Arabian Sea, and
68 the eastern coast of China. Several other studies have also investigated AOT trends using ground-
69 based Aerosol Robotic NETwork (AERONET) data (Li et al., 2014), space borne lidar
70 observations (Toth et al., 2016) and other passive-based observations or model simulations
71 (Thomas et al., 2010; Mishchenko et al., 2012; Hsu et al., 2012; Zhao et al., 2013; Chin et al.,
72 2014).

73 Still, to our knowledge, SWARE trends have not been studied with the use of both Terra and
74 Aqua data sets. In addition, the new Collection 6 (C6) MODIS aerosol products have changed the
75 magnitudes of global AOT fields significantly (Levy et al., 2013). Thus, in this study, using C6
76 MODIS and MISR aerosol products, as well as CERES data, we studied AOT and SWARE trends
77 over global oceans with a goal of exploring the following scientific questions:

- 78 1) To what extent have trends changed with the update from MODIS C5 to C6?
- 79 2) What are the regional and global AOT trends over global oceans with the use of near the
80 full Terra/Aqua MODIS and Terra MISR data records?
- 81 3) What are the regional and global trends in MODIS and CERES-based SWARE (Note that
82 although MODIS data are used for cloud clearing, CERES inferred SWAREs are independent of
83 forward calculations of MODIS and MISR)?
- 84 4) What are the instantaneous SW aerosol direct forcing efficiencies and SWARE values on
85 both regional and global scales using near the full Aqua and Terra data records?
- 86 5) Can cohesive conclusions (trend patterns) be achieved among passive-, active-based AOT
87 as well as SWARE trend analyses?

88 This paper is organized as follows. Data used in this study are described in Section 2. In
89 Section 3, differences in AOT trends using C5 and C6 MODIS DT aerosol products are examined
90 for the study period of 2000-2009, and then AOT trends are further derived with the use of near
91 full Terra MODIS and MISR (2000-2015) as well as Aqua MODIS (2002-2015) data records. In
92 Section 4, regional and global SW aerosol direct forcing efficiencies, magnitudes of SWAREs, as
93 well as trends in SWARE are studied using collocated CERES and C6 MODIS DT aerosol
94 products over global oceans. An uncertainty analysis in the derived SWARE trends is also carried
95 in section 4. In Section 5, regional-based AOT and SWARE trends derived from this study are
96 inter-compared with aerosol trend analyses estimated from several other studies that use the
97 CALIOP, MODIS and MISR instruments. Conclusions and discussions are provided in Section
98 6.

99

100 2. Datasets

101 Eight satellite data sets are included in this study (also shown in Table 1). Regional and global
102 over ocean AOTs were extracted from C6 Terra (MOD04_L2, 2000-2015), Aqua (MYD04_L2,
103 2002-2015) MODIS DT level-2 aerosol products (Levy et al., 2013) and Version 22 MISR (2000-
104 2015; Kahn et al., 2010) aerosol products. The Edition 3 Terra and Aqua CERES ERBElite (ES-
105 8; Barkstrom and Wielicki, 1996) and the Edition 3A CERES Single Satellite Footprint (SSF; Loeb
106 and Kato, 2002) Level 2 swath products provide instantaneous broadband SW fluxes. CALIOP
107 Level 2 5-km cloud layer products (Winker et al., 2010) are also used to assist the cirrus cloud-
108 related analysis.

109 *2.1 MODIS DT aerosol products:* The over ocean C6 MODIS DT aerosol products provides
110 spectral AOT at seven wavelengths ranging from visible to Shortwave Infrared at a 10 km nadir

111 spatial resolution, with an increased pixel size of 20x48 km near the edge of the swath (Levy et
112 al., 2013). Only the 550 nm AOT products are used in this study. Compared to the over ocean C5
113 MODIS DT products, aside from changes in upstream products such as L1B reflectance,
114 geolocation, land/sea and cloud mask, one major change included in the over ocean C6 MODIS
115 DT data is the use of non-static near surface wind speeds in the retrieval process (Levy et al.,
116 2013). In this study, only AOT retrievals with a Quality Assurance (QA) flag of marginal
117 confidence or higher are used. The reported uncertainty in AOT data is on the order of $(-0.02 \cdot \text{AOT}$
118 $- 10\%)$, $(+0.04 \cdot \text{AOT} + 10\%)$ (e.g. Levy et al., 2013), although several studies suggest that higher
119 uncertainties could be found for individual retrievals (e.g., Shi et al., 2011).

120 *2.2 MISR aerosol products:* On board the Terra satellite platform, the MISR instrument
121 provides observations at nine different viewing zenith angles ($\text{VZA} = 0$ (nadir), $\pm 26.1^\circ$, $\pm 45.6^\circ$,
122 $\pm 60.0^\circ$, $\pm 70.5^\circ$) at four different spectral bands ranging from 446 to 866 nm, although like MODIS
123 we focus on the green wavelength here (558 nm). Even though MISR has a much narrower swath
124 of ~ 360 km in comparison to MODIS (Diner et al., 2002), the multi-angle observations from MISR
125 enable a more reliable AOT retrieval over bright scenes such as desert regions (Kahn et al., 2010).
126 Thus, unlike the MODIS and CERES-based analyses in this study, which focus on global oceans,
127 trend analyses from MISR include both land and ocean regions, unless otherwise stated.

128 *2.3 CERES SSF products and issues:* The CERES SSF data are constructed through weighted
129 averaging of MODIS aerosol and cloud retrievals within a CERES footprint based on CERES
130 point spread function (PSF, Loeb et al., 2003; Geier et al., 2003). The CERES instrument measures
131 TOA broadband radiance, to convert from radiance to flux, angular distribution models (ADMs)
132 are needed (e.g. Loeb et al., 2003). For the CERES SSF products, CERES ADMs (Loeb et al.,
133 2003) are used to convert CERES radiance to flux. Over cloud free oceans, AOT is accounted for

134 in CERES ADMs through the use of the radiative transfer modeled anisotropic factors, stratified
135 as sea salt AOT values (Loeb et al., 2003), without considering the impacts of absorbing aerosols.
136 The CERES SSF data cannot be directly used in this study, however, simply because it is
137 constructed with the MODIS products in active production at the time of data collection. That is,
138 both Collection 4 (C4; before April 2006) and C5 (after April 2006) MODIS DT aerosol data were
139 used in constructing CERES SSF data ([http://ceres.larc.nasa.gov/products.php?product=SSF-](http://ceres.larc.nasa.gov/products.php?product=SSF-Level2)
140 [Level2](http://ceres.larc.nasa.gov/products.php?product=SSF-Level2)). This creates a problem for using CERES SSF in trend analysis, as changes are expected
141 in both global and regional estimations of AOTs between C4 and C5 MODIS DT aerosol products.
142 In addition, C6 MODIS aerosol data, which are currently available, are not included in the CERES
143 SSF data for the study period. Thus, the CERES SSF data are used in this study by collocating
144 with CERES ES-8 and C6 MODIS DT data, which are explained in detail later.

145 *2.4 CERES ES-8 products:* The CERES ES-8 data are also available for the near full Terra
146 and Aqua data records. The CERES ES-8 data are constructed by using ADMs from the Earth
147 Radiation Budget Experiment (ERBE)-like algorithm (Suttles et al., 1988). No aerosol properties
148 are considered in constructing ERBE ADMs. Thus, CERES ES-8 data are used for evaluating the
149 impact of ADMs on CERES derived SWAREs, and for inter-comparison with CERES SSF-based
150 analyses in this study.

151 *2.5 Collocated CERES SSF, ES-8 and MODIS DT products:* CERES SSF, CERES ES-8 and
152 C6 MODIS DT datasets were collocated in this study using 14 years of Aqua and 16 years of Terra
153 data. This is achieved by collocating CERES SSF and ES-8 data as the first step. Note that CERES
154 SSF data include geolocations at surface yet CERES ES-8 data report geolocations at TOA, thus,
155 the collocation is performed by selecting pairs of pixel-level data points from both products that
156 are in the vicinity of each other (less than 2 degree Latitude/Longitude) and have identical raw

157 observations (CERES upward “TOT filtered radiance” and “SW filtered radiance”). Also, CERES
158 SSF reported “Clear area percent coverage at subpixel resolution” values, which are used to define
159 the clear area percentage (CP) in this study, are applied as the initial cloud screen method. Only
160 collocated CERES SSF / ES-8 pairs that have CP values of 95% or higher are included in further
161 analyses. Note that only CERES pixels that have a MODIS reported cloud fraction of 1% or less
162 are used in the final process. A more relaxed CP threshold of 95% is adopted here, partially for
163 studying the impact of cloud contamination on CERES derived SWAREs as shown in Table 5.

164 As the second step, the collocated CERES SSF and ES-8 data are further collocated with C6
165 MODIS DT data. Only MODIS and CERES data that are from the same satellite platform are used
166 in the collocation. To collocate MODIS and CERES data, surface geolocations
167 (Latitudes/Longitudes) of both datasets are first identified and the two datasets are collocated in
168 space and time based on the PSF of the CERES instrument (Wielicki et al. 1996, Christopher and
169 Zhang, 2002a;b, Zhang and Christopher, 2003). Arithmetic averages are performed for MODIS
170 data points that are within a CERES footprint.

171 CERES data are available from three scan modes: the cross-track, rotating azimuth plane scan,
172 and fixed azimuth plane scan modes. To maintain data consistency, only cross track mode CERES
173 data from Terra and Aqua are used in this study. Also, to further screen potential noisy data, only
174 CERES observations with valid SW flux retrievals (from CERES-ES-8 or CERES SSF) and
175 viewing zenith angle (VZA) as well as solar zenith angle (SZA) less than 60 degrees are considered
176 in this study. Over land observations are further excluded in the study by only using collocated
177 pairs that have CERES ES-8 scene ID of “Clear Ocean”, “Partly Cloudy Over Ocean” and “Mostly
178 Cloudy Over Ocean”. Cloud and aerosol properties within a CERES observation are reported
179 based on the collocated C6 MODIS DT products. The following ancillary data are also recorded

180 for each CERES observation: total number of collocated C6 MODIS DT retrievals, number of
181 valid C6 MODIS DT retrievals (with valid cloud fraction and AOT values), number of valid C6
182 MODIS DT retrievals with QA flags of “marginal”, “good” and “very good”. Lastly, only CERES
183 pixels with CP larger than 99% and a reported MODIS cloud fraction (CF) of less than 1% and are
184 used in this study and the impacts of cloud contamination on the derived SWARE trends are also
185 evaluated later in this paper.

186 *2.6 Collocate CERES ES-8, MODIS DT and CALIOP products:* Using collocated CALIOP
187 and MODIS observations, Several studies have suggested that MODIS AOT retrievals may be
188 contaminated with optically thin cirrus clouds (OTC, e.g. Kaufman et al., 2005, Huang et al., 2011,
189 Feng et al., 2011, Toth et al., 2013). To further study the effects of OTC on the trend analysis, the
190 5 km CALIOP cloud layer product (Winker et al., 2010) is utilized. The CALIOP cloud layer
191 (CAL_LID_L2_05kmCLay) data are spatiotemporally collocated with the already collocated
192 MODIS-CERES data sets on-board the Aqua platform. CALIPSO’s Feature Classification Flag is
193 used to locate residual OTC within CERES observations. It should be noted that CALIOP’s data
194 record spans only about half of our study period (June 2006 – Dec. 2015) and is available only on
195 the Aqua platform, thus it will be used as a secondary analysis presented in Section 4.2. Note the
196 CERES CALIPSO CloudSat MODIS (C3M) products, which are constructed by collocating
197 CERES SSF, CALIPSO, CloudSat and MODIS data (Kato et al., 2011), are also available from
198 2006-2011 (<https://ceres.larc.nasa.gov/products.php?product=CCCM>). However, the C3M data
199 are not available after 2011. Also, to avoid decoupling the impacts of ADMs and cirrus cloud
200 effects, a simple approach, as mentioned in this section is used in this study.

201

202 **3. AOT trends from over ocean DT MODIS data**

203 To initiate this study, we begin with an update to global trend analyses in AOT. Included are
204 two components. First, we evaluate if recent changes in the MODIS aerosol product affect past
205 conclusions on regional aerosol trends over the globe. This is followed by an extension of the trend
206 analysis to the entire 2000-2015 study period (Section 3.2).

207

208 **3.1 Update of AOT trends from Collection 5 to Collection 6**

209 In the Zhang and Reid (2010) paper, 10 years of C5 DT MODIS over ocean data were used in
210 deriving regional and global AOT trends. With the recent release of C6 Aqua and Terra DT
211 MODIS data, including significant updates to calibration and cloud clearing algorithms, it is worth
212 a short reproduction of this work with current products.

213 Similar to Zhang and Reid (2010), Level 2 C6 DT over water Terra MODIS data were binned
214 into $1^\circ \times 1^\circ$ (Latitude/Longitude) monthly averages. “Bad” retrievals, as indicated by the QA flag
215 included in the dataset, are discarded from the analysis, as were MODIS cloud fraction above 80%
216 to minimize the effect of cloud contamination (Zhang and Reid, 2010). Using the monthly gridded
217 over-ocean C6 Terra MODIS DT data from 2000-2009 (excluding August 2000 and June 2001 as
218 these months contained less than 20 days of valid data), regional AOT trends, as well as trend
219 significances (based on WH, as suggested from Zhang and Reid, 2010) were derived and are shown
220 in Figure 1a. Trend significances are computed based on two statistical methods. To be consistent
221 with Zhang and Reid (2010), the Weatherhead method (Weatherhead et al., 1998, hereafter WH),
222 which account for data autocorrelation, is used to calculate trend significances for monthly-based
223 AOT data. For a comparison purpose, the Mann-Kendall method (e.g. Mann, 1945; Kendall, 1975,
224 hereafter MK) is also used. Note that the MK method is also used as it can be applied to estimate

225 trend significances for seasonal-based analysis as discussed in section 4. Both methods are applied
226 in Sections 3 and 4, wherever applicable.

227 To create Figure 1, data are deseasonalized by removing 10-year averages from any given
228 month, for each grid point. Also, AOT trends are derived only for bins which have more than 72
229 months (60%) of valid data records. In Figure 1a, regions with statistically significant trends at a
230 95% confidence interval (from WH), are highlighted with black dots.

231 To inter-compare AOT trend analysis from Zhang and Reid (2010), AOT trends from 10
232 selected regions are computed as shown in Table 2. Also, suggested from Zhang and Reid (2010),
233 the AOT trend from Remote Ocean (RO, 40° S - 0°, 179° W – 140° W) is used as a proxy for
234 unrealized bias in the AOT trend due to issues such as calibration and signal drifts, as this is the
235 region that is least affected by any major aerosol plumes originated from main continents. The
236 ratios and differences in AOT trends for both C6 and C5 Terra MODIS based analysis are also
237 shown in Figs. 1e and 1f, respectively, for the study period of 2000-2009. Only grids with AOT
238 trends above or below ± 0.002 AOT/year are used in this comparison.

239 As suggested from Table 2, both AOT trends and trend significances (based on WH) are similar
240 with the use of C5 and C6 Terra MODIS DT over ocean data for the study period of 2000-2009.
241 This suggests that although documentable changes are made to the C6 MODIS DT over ocean data
242 (Levy et al., 2013), the impact of those changes on global and regional AOT trend analysis is rather
243 marginal. For a comparison purpose, Table 2 also includes trend significances derived using the
244 Mann-Kendall method ($|z|$) for the C6 MODIS DT-based analysis, and consistent results are found
245 from both methods a majority of the time.

246 Lastly, regional and global averages over ocean C5 and C6 Terra MODIS DT AOTs are also
247 shown in Table 2 for the period of 2000-2009. Note that in Zhang and Reid (2010), data-

248 assimilation quality C5 MODIS DT data, which is implemented with extensive QA steps (e.g.
249 Zhang and Reid, 2006; Shi et al., 2011), were used. Here regional and global mean C5 AOTs are
250 derived using similar steps as were used in constructing the C6 AOT data, which are differ from
251 the data-assimilation quality C5 MODIS DT data as used in Zhang and Reid (2010). Still, as
252 suggested from Zhang and Reid (2010), although QA steps could lower the mean global over ocean
253 AOTs from ~0.15 to ~0.11, in part due to the removal of cloud contaminated retrievals, minor
254 impacts on the AOT trend analysis are reported. As suggested from Table 2, a 10% reduction in
255 global mean over ocean AOT is found for the C6 MODIS DT data in comparing with the C5 data,
256 possibly due to a reduction in marine background AOTs (e.g., the Enhanced Southern Ocean
257 Anomaly feature, as shown in Toth et al., 2013, no longer exists in the C6 product).

258

259 **3.2 AOT trends from near full Terra and Aqua data records**

260 Extending the analysis from the previous section, AOT trends are evaluated for the near full
261 available data record (March 2000 – December 2015 for MODIS Terra and MISR, and July 2002
262 – December 2015 for MODIS Aqua) of C6 over ocean MODIS DT and MISR aerosol products.
263 The C6 MODIS DT data are processed and filtered with the same steps as mentioned in section
264 3.1 to construct $1^\circ \times 1^\circ$ (Latitude/Longitude) monthly averages for trend estimates. MISR products
265 are also binned into monthly-averaged $1^\circ \times 1^\circ$ degree bins and filtered according to Zhang et al.,
266 (2017 submitted).

267 Figure 2 depicts the C6 MODIS Terra (Fig. 2a), C6 MODIS Aqua (Fig. 2b) and v22 MISR
268 (Fig. 2c)-based global aerosol distributions (Latitude: -60° to 60°) using monthly gridded AOTs.
269 Only those bins with more than one thousand data counts were considered for this analysis (this is
270 an arbitrary threshold selected for removing some over land water retrievals over scenes such as

271 lakes. It is also partially used for ensuring sufficient data are included in the trend analysis). Figs.
272 2a and 2b show a high level of similarity over most of the globe, which is consistent with what has
273 been reported by Remer et al. (2006) using 3 years of C5 MODIS data . Similar spatial patterns
274 are also found for MODIS- and MISR-based AOT analyses over global oceans (Figure 2c). This
275 is further confirmed from Figures 2d and 2e, which show the ratios and the differences between
276 Terra MODIS and Terra MISR AOTs. Still, the band of high AOT over the southern oceans,
277 which is identified as a potential artifact in both C5 MODIS and MISR aerosol products that may
278 be due to cloud contamination (Toth et al., 2013), is no longer apparent in the C6 MODIS DT
279 aerosol products.

280 Using data shown in Figure 2, the time series of over ocean global mean AOT are also
281 examined and shown in Fig. 3. Figure 3a shows the monthly-averaged C6 MODIS Aqua (red),
282 MODIS Terra (blue) and MISR (green) AOTs over global oceans for the entire time frame of each
283 data set. It should be noted that over land observations from MISR are not included in global
284 averages in order to get a more direct comparison with the over ocean MODIS DT aerosol data
285 sets. Monthly-variations in globally-averaged (simple arithmetic mean) AOTs can be observed,
286 with the solid lines showing the AOT trends for the entire time period for each sensor. Similar to
287 Zhang and Reid (2010), the lowest monthly-averaged MODIS AOTs are found during the
288 Northern-Hemispheric winter months while the highest aerosol loading activity over global oceans
289 seems to occur during the Northern-Hemispheric spring and summer months.

290 Figure 3b shows AOT anomalies after deseasonalizing the monthly data shown in Figure 3a.
291 Terra MODIS and MISR show trends of differing signs; a statistically significant increase/decrease
292 in monthly-mean AOT values of $0.008/-0.005$ AOT decade⁻¹ is found when using Terra
293 MODIS/MISR data for the study period of 2000-2015. In comparison, a statistically insignificant

294 global over ocean AOT trend is found to be $0.0003 \text{ AOT decade}^{-1}$ using Aqua MODIS data for the
295 study period of 2002-2015. A trend difference is clearly seen even if we restrict all datasets to the
296 same study period of 2002-2015, which could be an indication of potential calibration related
297 issues with one or all of the sensors.

298 Zhang and Reid et al., (2010) suggested that since the remote oceans region (defined in Table
299 2) is least affected by major continental originated aerosol plumes, the AOT trend from this region
300 may be used for checking calibration related issues or some other unrealized uncertainties
301 originated from the upstream data used. A caveat here is that we assume that the calibration
302 degradation propagates linearly into AOT. The correction might therefore be an under/over-
303 correction in those higher-AOT areas. Similar to Fig. 3b, Fig. 3c depicts the monthly-averaged
304 deseasonalized AOTs over the remote ocean region where the monthly anomalies and trend lines
305 are visible. Similar to Zhang et al. (2017), an insignificant trend of $0.0003 \text{ AOT decade}^{-1}$ is found
306 for the remote ocean region using Aqua MODIS data, while a statistically significant
307 (Weatherhead method) trend of $0.006/-0.004 \text{ AOT decade}^{-1}$ is found for the same region with the
308 use of deseasonalized Terra MODIS/MISR data. Those differences in AOT trends are not
309 surprising. For example, a recent study suggests a potential cross-talk among Terra MODIS
310 thermal channels, which will affect MODIS cloud detection (Moeller and Frey, 2016) and
311 correspondingly, Terra MODIS AOT trends. Similarly, Limbacher and Kahn, (2016) reported an
312 up to 2% decrease in MISR signals from 2002-2014 that could affect MISR AOT trends. AOT
313 trends estimated from this study are henceforth adjusted based on AOT trends detected from the
314 Remote Ocean region; this is done to reduce potential impacts from upstream data used in the AOT
315 retrievals by assuming that a near zero AOT trend should be observed over the remote ocean region
316 (shown in Table 3).

317 Using monthly gridded data, AOT grid-level trends are also estimated on a global scale, for
318 MODIS Terra- (Fig. 1b), MODIS Aqua- (Fig. 1c) and MISR (Fig. 1d)-based analysis for the entire
319 data record period. Again, the black-dotted areas on the map are for regions with statistically
320 significant trends at a 95% confident interval estimated using the WH method. When comparing
321 with the 10-year analysis as mentioned in Section 3.1 (Fig. 1a), some similarities are clearly
322 visible. For example, increasing AOT trends are observed over the Arabian Sea and Indian Bay
323 of Bengal, while decreasing trends are observed over the Mediterranean Sea and east coast of US
324 from both Figures 1a and 1b. Still for some regions, such as over coastal China, Fig. 1a shows a
325 positive AOT trend, yet near zero AOT trend is found in Figure 1b. A recent study suggests a
326 possible increase in AOT from 2000-2007 over coastal China, followed by a decreasing trend from
327 2008-2015 (Zhang et al., 2017), which can be used to explain the differences as observed in Figure
328 1 over coastal China. Likewise, regional analyses are also conducted as documented by Table 3
329 and Figure 4. In addition to the regions reported by Zhang and Reid (2010), two regions have been
330 added to the study which include Persian Gulf (24° N – 30° N, 50° E – 60° E) and Red Sea (15° N
331 – 30° N, 30° E – 45° E). All regions are outlined by black boxes in Fig. 1.

332 Unlike the insignificant AOT trends on the global scale, both statistically significant positive
333 and negative trends are found for several regions as shown in Figure 4 (as well as Table 3). For
334 example, statistically significant positive AOT trends (where statistically significant trends are
335 denoted by bold font on Table 3) are found from all three datasets (Terra and Aqua MODIS DT
336 and MISR over water aerosol products) over the Bay of Bengal (Fig. 4a), Arabian Sea (Fig. 4b)
337 and Red Sea (Fig. 4d). Note that both the Bay of Bengal and Arabian Sea have been identified in
338 Zhang and Reid (2010) as regions with statistically significant positive trends for the study period
339 of 2000-2009. However, the rates of increase of aerosol loading have plausibly slowed down over

340 the last five years for both regions, indicated by ~20-30% reductions in AOT trends when
341 estimated using the near full Terra data records. Flattening of AOT trends with respect to time can
342 also be observed in Fig. 4 for both regions for 2010-2015. The Red Sea and Persian Gulf are
343 newly introduced for this study but seem to show the highest increase in aerosol loading during
344 the study period (as derived from Terra data). This increase in AOT has been attributed to a number
345 of mechanisms, including a trend in surface wind, precipitation, and soil moisture (Al Senafi and
346 Anis 2015; Klingsmuller et al, 2016), as well as a climatological deepening of the summertime
347 monsoonal low over the Arabian Sea (Solmon et al., 2015). Statistically significant negative trends
348 are found over the Mediterranean Sea (Fig. 4f) and the east coast of N. America (Fig. 4g), again
349 from all three datasets. These findings are also consistent with what has been reported by Toth et
350 al. (2016) with the use of CALIOP data. Also, despite the differences in sampling methods as well
351 as calibration, regional trends from MISR are similar to trends derived using both Aqua and Terra
352 MODIS DT data.

353

354 **4. SWARE Trends**

355 In Section 3, changes in aerosol concentrations over global oceans are studied with respect to
356 AOT trends. The temporal variations in aerosol concentrations could also introduce changes in
357 TOA SW fluxes and thus can be detected using collocated MODIS and CERES (SSF and ES-8)
358 observations. In this section, the SWARE trends derived using MODIS and CERES (SSF and ES-
359 8) data are explored and are inter-compared with AOT trends as mentioned in the previous section.

360

361 **4.1 SWARE trend Analysis using collocated MODIS and CERES data**

362 In several past studies, SWARE values are derived using collocated CERES and MODIS data
363 based on equation 1 (e.g. Loeb and Kato, 2002; Loeb et al., 2003; Zhang et al., 2005b; Christopher
364 and Zhang., 2002a;b):

$$365 \quad SWARE = F_{clear} - F_{aero} \quad (1)$$

366 where F_{clear} represents the TOA SW flux over aerosol and cloud free skies and F_{aero} represents the
367 TOA SW flux over cloud free skies. Taking the derivative of equation 1 with respect to time, we
368 can obtain equation 2:

$$369 \quad \frac{\partial SWARE}{\partial t} = \frac{\partial F_{clear}}{\partial t} - \frac{\partial F_{aero}}{\partial t} \quad (2)$$

370 Here $\partial SWARE/\partial t$ represents the trend in SWARE. $\partial F_{aero}/\partial t$ represents a temporal change in TOA
371 observed SW flux over cloud free skies. $\partial F_{clear}/\partial t$ represents a change in background TOA SW
372 energy over cloud and aerosol free skies. Here F_{clear} is a function of viewing geometry (e.g., solar
373 zenith angle) and near surface wind patterns. By deseasonalizing CERES SW flux data, we can
374 remove the solar zenith angle effect. Also, by using monthly averages of instantaneous retrievals,
375 we assume that there is no viewing zenith or azimuth dependency with respect to time. If we
376 further assume that the changes in near surface wind patterns are negligible for the study period,
377 the $\partial F_{clear}/\partial t$ term can be assumed to be near zero (the impact of near surface wind speed on the
378 SWARE trend is explored in a later section). Thus, we can rewrite equation 2 as:

$$379 \quad \frac{\partial SWARE}{\partial t} = \frac{-\partial F_{aero}}{\partial t} \quad (3)$$

380 As suggested from equation 3, the trends in SWARE can be directly estimated from the temporal
381 variations in SSF/ES-8 TOA SW flux from CERES over cloud free skies (less than 1% cloud
382 fraction and larger than 99% CP). This approach avoids the need for estimating F_{clear} , which cannot
383 be observed and can only be derived through radiative transfer calculations (Christopher, 2011) or
384 extrapolation (e.g., there is always a positive definite AOT).

385 The cloud-free TOA SW fluxes are obtained from CERES (SSF and ES-8) data in this study.
386 This is accomplished by utilizing the collocated MODIS-CERES (SSF and ES-8) data set. As
387 mentioned in Section 2, only those MODIS observations over cloud-free scenes ($CF < 1\%$ and CP
388 $> 99\%$) are used for this analysis as SW flux is sensitive to cloud contamination (Zhang et al.,
389 2005a;b). However, filtering the MODIS data sets with such strict cloud fraction criteria
390 significantly reduces the data volume, which may lead to a sampling bias when working with the
391 MODIS-CERES data set (e.g., Zhang and Reid 2009). Therefore all MODIS-CERES data sets
392 have been averaged into seasons as opposed to monthly averages. In addition, the MODIS-CERES
393 collocated observations are gridded into $2^\circ \times 2^\circ$ (Latitude/Longitude) grids to further alleviate the
394 sampling bias produced by the data reduction in the MODIS-CERES data set.

395 Figure 5 shows the spatial distributions of AOT and cloud-free CERES TOA SW flux over
396 global oceans using collocated MODIS-CERES data (2000-2015 for Terra and 2002-2015 for
397 Aqua). Comparing Figs. 5a and 5b with Figs. 2a and 2b, Terra (5a) and Aqua (5b) AOT plots
398 generated using the collocated MODIS-CERES data are similar to the spatial distributions of AOT
399 generated using the original Terra and Aqua C6 MODIS DT data. Figures 5e and 5f show the
400 gridded cloud-free CERES SSF TOA SW fluxes for Terra and Aqua, respectively. It is interesting
401 to note that the spatial distributions of MODIS AOT and cloud free CERES SSF TOA SW flux
402 (SW_{ssf}), although from two different instruments that measure different physical quantities (narrow
403 band versus broadband energy; dependent versus independent of forward calculations of MODIS),
404 show remarkably similar patterns.

405 Similar to Figs. 5e and 5f, Figs. 5c and 5d show the gridded cloud-free TOA SW fluxes for
406 Terra and Aqua respectively, but with the use of CERES ES-8 SW fluxes. Again, the spatial
407 patterns of cloud-free CERES ES-8 TOA SW flux (SW_{es8}) highly correlate with AOT spatial

408 patterns. Still, an overall difference in CERES SSF and ES-8 TOA SW fluxes is clearly observable
409 (Figs. 5g and 5h) and SW_{ssf} values are generally 8-9 Wm^{-2} higher than SW_{es8} values. Smaller than
410 average differences in cloud free TOA SW fluxes between the two products can be seen over dust
411 aerosol polluted regions such as the northwest coast of Africa, while larger than average
412 differences are found over regions such as east coast of Asia, west coast of South America and
413 Southeast Asia where other type of aerosol particles dominate. For illustrative purposes, data
414 counts for each $2 \times 2^\circ$ (Latitude/Longitude) bin that are used to create Figs. 5a-h are also shown in
415 Figs. 5i and 5j for Aqua and Terra respectively.

416 The relationship between AOT and cloud free TOA SW flux values from Fig. 5 is also
417 evaluated in Figs. 6 and 7 and Table 4. As suggested from Fig. 6a (Aqua) and 6c (Terra), multi-
418 year means of AOTs and SW_{ssf} values share a highly correlated (correlations of 0.72 and 0.73 for
419 Aqua and Terra data, respectively), non-linear relationship. Similar but higher correlations
420 between multi-year mean AOT and SW flux values are also found when using CERES ES-8 data
421 (correlations of 0.83 and 0.87 for Aqua and Terra data, respectively) as shown in Figs. 7a (Aqua)
422 and 7c (Terra).

423 Figure 6b shows the Aqua MODIS AOT and Aqua SW_{ssf} relationship (non-linear) for 5
424 selected regions that have high regional AOT values (e.g., maximum bin averaged AOT > 0.3),
425 including the southwest and northwest coasts of Africa, coastal China, India Bay of Bengal, and
426 Arabian Sea. In particular, a much lower slope of 37.8 Wm^{-2} per AOT is found for the southwest
427 coast of Africa region in-comparing with the other four regions. A similar pattern is observed for
428 using Terra CERES SSF data (slope of 42.5 Wm^{-2} per AOT for the southwest coast of Africa
429 region) as well as for using both Aqua and Terra CERES ES-8 data (slopes of 39.8 and 43.7 Wm^{-2}
430 per AOT for Aqua and Terra respectively, for the southwest coast of Africa region). Note that

431 the slope of AOT and SW flux is a measure of (inversely proportional to) the instantaneous SW
432 aerosol direct forcing efficiency. Smoke aerosol particles dominate high AOTs for the southwest
433 coast of Africa region, while other regions are also influenced by non-smoke aerosols such as dust
434 aerosol particles. Thus Figs. 6 and 7 suggest a lower SW forcing efficiency (in magnitude) for
435 biomass burning aerosols, in part due to a stronger absorption at the visible spectrum (e.g., Remer
436 et al., 2005).

437 We have further explored the topic by estimating SW aerosol forcing efficiencies for the Dec.-
438 May and Jun.-Nov. seasons as shown in Table 4. As indicated in Table 4, SW aerosol direct
439 forcing efficiencies may experience a seasonal dependency such as over the Coastal China region.
440 For example, a CERES SSF-based aerosol SW forcing efficiency value of -88.3 Wm^{-2} per Aqua
441 MODIS AOT is found for the coastal China region for the Dec.-May period. A lower value
442 (CERES SSF-based) of -74.7 Wm^{-2} per Aqua MODIS AOT is found for the Jun.-Nov. season for
443 the same region. Similar conclusions can also be found using Terra data as well as using CERES
444 ES-8 data. The seasonal dependency in SW aerosol forcing efficiency is not surprising for the
445 coastal China region, as dust aerosols are expected for the spring season, while pollutant and smoke
446 aerosols likely dominate for the Jun.-Nov. study period (Zhang et al., 2017). In comparison, less
447 seasonal-based changes are found for the Arabian Sea region, which may be plausibly linked to
448 less significant temporal variation in aerosol speciation over the region. Also indicated in Table
449 4, the derived SWARE has a strong regional-dependency, while the multi-year averaged SWARE
450 is around -6 to -7 Wm^{-2} for the southwest coast of Africa region, over the coastal China region,
451 SWARE values of below -20 Wm^{-2} are found. Note that this conclusion remains unchanged
452 regardless of using Terra or Aqua data, or using CERES ES-8 or SSF ADMs.

453 Over global oceans, the multi-year mean instantaneous SW aerosol direct forcing efficiencies
454 are estimated to be -61 (-58) and -58 (-58) Wm^{-2} per AOT using Aqua and Terra CERES SSF (ES-
455 8) data, respectively. Those numbers are lower than -70 Wm^{-2} per AOT, which is reported from a
456 previous study (Christopher and Jones, 2008). We suspect that the differences in forcing efficiency
457 values may be introduced by different data screening methods as well as a much longer study
458 period. Still, using estimated forcing efficiencies as well as AOTs (Table 4), the global mean (14
459 years of Aqua and 16 years of Terra data) over oceans SWARE values are found to be around -7
460 Wm^{-2} regardless of datasets (Terra or Aqua) and ADMs (SSF or ES-8) used. Note that regional
461 and global mean AOTs as shown in Table 4 are derived using the collocated MODIS and CERES
462 datasets, representing mean AOTs over CERES cloud-free skies. Thus, mean AOTs as reported
463 from Table 4 are different from AOTs as included in Table 2.

464 With the use of seasonally gridded SW flux values, the times series of cloud-free sky CERES
465 SSF and ES-8 TOA SW flux over global oceans are investigated and depicted in Fig. 8a, and the
466 corresponding deseasonalized cloud-free sky flux anomalies are show in Fig. 8b. While Fig. 8a
467 suggests an $\sim 8 \text{ Wm}^{-2}$ difference in mean over ocean cloud-free sky SW flux between CERES SSF
468 and ES-8 products, a small difference in cloud-free sky SW flux trend of 0.2-0.3 $\text{Wm}^{-2} \text{ decade}^{-1}$ is
469 found (Fig. 8b) between the two products for both Terra and Aqua data. For example, negative
470 trends on the order of -0.50 Wm^{-2} and -0.26 Wm^{-2} per decade are found for using Aqua CERES
471 ES-8 and SSF products respectively. Also, although larger cloud-free sky SW flux trends in
472 magnitude are found when using Terra data, the difference between CERES SSF-based and
473 CERES ES-8-based trends is still on the order of 0.2 – 0.3 $\text{Wm}^{-2} \text{ decade}^{-1}$ (Cloud-free sky SW flux
474 trend is -1.50 $\text{Wm}^{-2} \text{ decade}^{-1}$ for Terra CERES ES-8 data and is -1.22 $\text{Wm}^{-2} \text{ decade}^{-1}$ for Terra
475 CERES SSF data). Figures 8a and 8b may imply that different ADMs could significantly impact

476 the derived SW flux values, but their impact on cloud-free sky TOA SW flux trends are rather
477 marginal.

478 Similar to Section 3, we used CERES SW flux trends over the remote ocean region as
479 indicators for potential radiometric calibration related issues. The deseasonalized CERES SSF
480 (ES-8) SW trends over the remote ocean regions (Fig. 8c) seem to suggest plausible artificial trends
481 of -0.25 (-0.50) $\text{Wm}^{-2} \text{decade}^{-1}$ for Aqua and -0.70 (-0.92) $\text{Wm}^{-2} \text{decade}^{-1}$ for Terra, although these
482 trends are also affected by various uncertainties that are further explored in a later section. To
483 examine if we could observe similar issues with the use of full CERES SSF / ES-8 datasets, Fig.
484 9 shows the all sky CERES flux trend for the same study periods as Fig. 8. Decadal changes of
485 SSF (ES-8) all sky flux are less than $0.5(0.7)$ Wm^{-2} and $0.4(0.5)$ Wm^{-2} for Terra and Aqua data,
486 respectively. The Aqua all-sky flux trends are comparable to cloud-free sky trends for both SSF
487 and ES-8 fluxes. However Terra-based all sky trends are much lower in magnitude than the
488 corresponding cloud-free flux, which indicates that cloud-free sky CERES SW energy may be
489 more sensitive to calibration related issues than all sky flux data for Terra-based analysis only.
490 Still, if we account for the changes in SW trends over the remote ocean region, a negligible SW
491 flux (SWARE) trend for Aqua and a negative (positive) SW flux (SWARE) trend of -0.5 Wm^{-2}
492 decade^{-1} (0.5 $\text{Wm}^{-2} \text{decade}^{-1}$) for Terra can be estimated for the global oceans from collocated
493 MODIS-CERES data.

494 Although different cloud-free sky SW flux trends are found while using CERES ES-8 data,
495 after adjusting the detected trends with trends from remote oceans, a zero SW flux (SWARE) trend
496 is found while using collocated Aqua ES-8 SW fluxes from the MODIS-CERES data and a
497 negative (positive) SW flux (SWARE) trend of -0.6 $\text{Wm}^{-2} \text{decade}^{-1}$ (0.6 $\text{Wm}^{-2} \text{decade}^{-1}$) is found
498 using collocated Terra ES-8 SW fluxes from the MODIS-CERES collocated data, both are in

499 good agreement with values estimated using the SSF SW fluxes from the same data. This again
500 may seem to suggest that the impact of ADMs on SWARE trends over global oceans estimated
501 from the collocated MODIS and CERES data are rather marginal.

502 A regional trend analysis for the deseasonalized cloud-free sky SSF and ES-8 SW fluxes is
503 also carried out and presented in Table 3 and Figure 10. A good agreement is shown between
504 regional trends of AOTs (Fig. 4) and cloud-free fluxes (Fig. 10) for a majority of the regions (also
505 shown in Table 3 for a direct comparison). For example, statistically significant positive (based
506 on the Mann-Kendall method) SW flux trends are found over the Arabian Sea, and statistically
507 significant negative trends are found over the Mediterranean Sea and eastern US coast for both
508 Aqua and Terra-based analyses. Also, over the east coast of China, although a near positive trend
509 is found for the study period of 2000-2006 (Terra), the SW flux trend turns negative from 2006-
510 2015 (Figure 11). This is consistent with what has been reported for AOT trends from a recent
511 study (Zhang et al., 2017) as well as in Section 3. Here a piecewise linear fit method from Tomé
512 and Miranda (2004) is applied to detect turning points in trends, similar to what is suggested by
513 Zhang et al. (2017). Also, similar to Zhang et al. (2017), we assume a minimum of 36 months
514 between any two detected turning points. For regions such as the Bay of Bengal, although positive
515 SW flux trends are found, the trends are not statistically significant for one or all datasets.

516 Next, the grid-level AOT and cloud-free flux trends are derived from the collocated
517 MODIS-CERES data sets as shown in Fig. 12. Figures 12a (Terra) and 12b (Aqua) depict the de-
518 biased (applied corrections based on the estimate from the remote ocean region) changes in
519 deseasonalized AOT per year for each $4^\circ \times 4^\circ$ (Latitude/Longitude) grid (averaged from the $2^\circ \times 2^\circ$
520 Latitude/Longitude dataset) over the entire time period (all seasons and years combined). Figures
521 12e and 12f depict the grid level CERES SSF SW flux trends over cloud-free skies similar to Figs.

522 12a and 12b. Similar to the AOT grid level analysis shown in Fig. 1, at least 60 percent of the data
523 record in each grid are required to have valid AOT and SW flux trend values. Comparing between
524 Aqua AOT (Fig. 12b) and CERES SSF cloud free SW (Fig. 12f) trends, similarity can be found.
525 For example, positive trends are found, from both plots, over coastal Indian and the Arabian Sea
526 regions, and negative trends are observable from Europe and the east coast of North America. The
527 similar conclusion can also be reached when using Terra data (Figs. 12a and 12e) as well as when
528 using CERES ES-8 data (Figs. 12c and 12d). Still discrepancies can be found. For example,
529 although the spatial distributions of AOT from both Terra and Aqua show similar patterns,
530 differences between the spatial distributions of Terra and Aqua CERES cloud-free SW fluxes,
531 regardless of ADMs used, are clearly visible. Much larger regions with negative cloud free SW
532 flux trends are found for using Terra data. This may be a result of several possible issues such as
533 SW flux outliers in the CERES data set, quality control applied to the CERES data set, or cloud
534 contamination issues. Thus, this will be examined in the following section.

535

536 **4.2 Uncertainty in Cloud-Free Flux Trend Analysis**

537 In this section, issues that could impact the derived SWARE trends are explored, which
538 include changes in near surface wind patterns, cloud contamination, and uncertainties in the cloud
539 free SW flux trend estimates over the remote ocean region (used as a proxy for radiometric
540 calibration). Note that there are other uncertainty sources that may impact the derived CERES SW
541 flux values, such as uncertainties in converting unfiltered to filtered radiances (Zhang et al.,
542 2005b). However, temporal variations of those uncertainty sources are assumed to be negligible
543 for this study, and thus those terms are not included in the trend uncertainty analysis.

544 *4.2.1 Baseline region (a proxy for radiometric calibration):* As mentioned in Section 4.1, the TOA
545 cloud-free SW flux trend over the Remote Ocean region is used as an indicator for potential
546 calibration related issues. The selection of the Remote Ocean region boundaries is rather arbitrary,
547 and thus the variations in TOA cloud-free CERES SW flux trends over the remote ocean region
548 are investigated by modifying the regional boundaries for four different scenarios as shown in
549 Table 5. Alternate remote ocean regions are chosen by shifting the original boundaries by 10
550 degrees in each direction. The variations in estimated CERES SSF (ES-8) SW flux trends, which
551 correspond to standard deviation values of 0.08 (0.09) and 0.03 (0.08) $\text{Wm}^{-2} \text{decade}^{-1}$ for Terra and
552 Aqua, respectively, provide the first order estimation of the potential variations in the estimated
553 SW trends over the remote oceans.

554 *4.2.2 Cloud fraction:* Similarly, the cloud-free SW flux trends over global oceans are estimated
555 through varying MODIS cloud fractions from 0 to 5% as indicated in Table 5. The standard
556 deviation of the data spread is found to be less than $0.1 \text{ Wm}^{-2} \text{decade}^{-1}$ for both Terra- and Aqua-
557 based CERES SSF and ES-8 SW flux trend analyses, suggesting that cloud contamination has a
558 minor effect on the trend analysis. This conclusion is also confirmed by a sensitivity test by
559 estimating SSF and ES-8 SW flux trends through varying CP values from 95% to 100%.

560 *4.2.3 Thin Cirrus:* Through the use of CALIOP observations, several studies suggest that OTC
561 cloud contamination exists in MODIS detected totally cloud free skies (e.g., Toth et al., 2013).
562 Therefore, the impacts of OTC clouds are evaluated by collocating CALIOP cloud layer data with
563 the already collocated Aqua MODIS and CERES data pairs. All CALIOP observations are
564 spatiotemporally collocated with the current original CERES observation if the temporal
565 difference in the two sensor's scan times is less than or equal to five minutes and if the center of
566 the CALIOP observations lies within 0.3 degrees of the center of the CERES observations. All

567 collocated CERES observations are assigned a cirrus cloud flag depending on whether any of the
568 collocated CALIOP pixels was found to be contaminated by cirrus clouds. The global averaging
569 process is once again performed using the collocated MODIS-CERES-CALIOP observations.
570 CERES observations which are contaminated by cirrus clouds, as identified by CALIOP data, are
571 removed from the averaging process. The resulting global AOT and cloud-free flux trends are
572 presented in Figs. 13a and 13b, respectively for using both CERES ES-8 and SSF SW fluxes. For
573 comparison, the MODIS-CERES trends are also shown (red) over the same time period (summer
574 2006 – fall 2015). Despite differences in globally averaged AOTs, the global TOA SW flux trends
575 derived using the two different data sets are remarkably similar. The standard deviation in global
576 cloud-free CERES SSF flux trend calculations due to OTC is less than $0.1 \text{ Wm}^{-2} \text{ decade}^{-1}$, as shown
577 in Table 5. Thus, OTC clouds may have a minimal impact on the derived cloud-free SW flux
578 trends.

579 *4.2.4 Surface Wind and ADMs:* The uncertainty in cloud-free SW flux trends are also examined as
580 a function of surface wind speeds and ADMs. As mentioned previously, the effect of surface wind
581 speed is included in CERES ADMs (used in the SSF data set). Thus, the SWARE trends derived
582 from the CERES SSF datasets are used to investigate ADMs and surface wind speed related
583 uncertainties in this study. Based on Table 3, the cloud-free sky SW flux trends derived from the
584 CERES SSF SW flux are -0.26 and $-1.22 \text{ Wm}^{-2} \text{ decade}^{-1}$ for using Aqua and Terra datasets
585 respectively, and the numbers are -0.50 and $-1.50 \text{ Wm}^{-2} \text{ decade}^{-1}$ for using CERES ES-8 data.
586 Thus, the cloud-free SW flux trends derived using the CERES ES-8 are on the order of -0.25 Wm^{-2}
587 decade^{-1} (corresponding to standard deviation values of 0.20 and $0.17 \text{ Wm}^{-2} \text{ decade}^{-1}$ for Terra
588 and Aqua, respectively) lower than the same trends derived using CERES SSF data for the same

589 study period. The $\sim 0.25 \text{ Wm}^{-2} \text{ decade}^{-1}$ difference indeed contains combined uncertainties from
590 ADMs as well as the changes in surface wind speeds for both Terra and Aqua datasets.

591 Overall, the largest sources of uncertainty in the SWARE trend estimates are from ADMs
592 / near surface wind speed changes while the impact of cloud contamination is rather minor. If we
593 assume the standard deviation values from Table 5 can be considered as uncertainties, an overall
594 uncertainty in the trend analysis can be estimated based on equation 4 (Penner et al., 1994; Zhang
595 et al., 2005b):

$$596 \quad U_t = e^{[\sum \log U_i^2]^{0.5}} \quad (4)$$

597
598 Where U_t is the overall uncertainty factor and U_i is the uncertainty factor from each item
599 in Table 5. The uncertainty factor is defined as such that if the percentage uncertainty is 8%, then
600 the uncertainty factor is 1.08. As shown in Table 5, estimated from Equation 4, the overall
601 uncertainties for the SWARE trends estimated using CERES SSF data are 0.3 and 0.2 Wm^{-2}
602 decade^{-1} for Terra and Aqua based analyses respectively, shown also in Table 5. Note that similar
603 numbers are also found by repeating the same exercise but using CERES ES-8 data as shown in
604 Table 5.

605

606 **5. Comparison to other aerosol related trend analyses**

607 Both AOT and SWARE trends are estimated in this study. Using CALIOP data from 2006-
608 2014, Toth et al. (2016) studied AOT and aerosol vertical distribution trends over both land and
609 oceans. Alfaro-Contreras et al. (2016) explored temporal variations in above cloud AOT with the
610 combined use of Ozone Monitoring Instrument (OMI) and CALIOP data. Although different
611 spectral widths (narrowband versus broadband), different instruments (passive versus active

612 sensors) and different observing conditions (cloud-free skies versus cloudy skies) are considered
613 in different studies, it is interesting to inter-compare trends derived from those studies, as shown
614 in Table 6. Another reason for selecting these studies is because AOT trends for similar regions
615 are reported.

616 Four studies are listed in Table 6, including passive based AOT analysis (Zhang and Reid 2010;
617 this study); SWARE analysis (this study); CALIOP-based AOT analysis (Toth et al., 2016); and
618 above-cloud AOT analysis (Alfaro-Contreras et al., 2016). It should be noted that only over ocean
619 data are used for the studies utilizing passive-based instruments (Zhang and Reid, 2010; current
620 study). The estimated trends from the active-based studies (Alfaro-Contreras et al., 2016; Toth et
621 al., 2016) included both land and ocean CALIOP data. Also, different data sampling, data
622 screening, and filtering methods are applied for different studies.

623 Table 6 includes estimates for global oceans for 7 selected regions that have reported values
624 from all four studies. It is interesting to note that positive trends in AOT (both from passive and
625 active methods), SWARE, and above cloud AOT are found over the Bay of Bengal and Arabian
626 Sea, although trends from some analyses are insignificant such as from the above cloud AOT
627 analysis (Alfaro-Contreras et al., 2016). Negative trends are found, across all four studies, over
628 the Mediterranean Sea and eastern coast of the US. The cohesive results from studies using
629 different instruments with varying methods, seem to add more fidelity to the trend analysis of this
630 study.

631 Still, over coastal China, while Zhang and Reid (2010) reported a statistically significant
632 positive AOT trend for the study period of 2000-2009, negative AOT trends are found from both
633 this study (2000-2015) and Toth et al., (2016; for 2006-2014). Again, this is because a potential

634 increase in aerosol loading for the early study period (2000-2007) continued with a decreasing
635 trend in aerosol loading after 2008, as suggested by a recent study (Zhang et al., 2017).

636

637 **6. Summary and Conclusions**

638 Using Terra (2000-2015) and Aqua (2002-2015) Collection 6 (C6) Moderate Resolution and
639 Imaging Spectroradiometer (MODIS) Dark Target (DT), Multi-angle Imaging Spectroradiometer
640 (MISR; 2000-2015) and Cloud and Earth's Radiant Energy System (CERES) ES-8/SSF data, both
641 Aerosol Optical Thickness (AOT) and Short-Wave Aerosol Radiative Effect (SWARE) trends are
642 estimated over global oceans. The results of this study are inter-compared with analyses from
643 several other studies that derived AOT trends using different instruments (e.g, active versus
644 passive) over different observing scenes (e.g. cloudy versus cloud free). This study suggests:

- 645 1. Updating the analysis from Zhang and Reid (2010), which examined AOT trend over
646 global oceans using the Collection 5 (C5) Terra MODIS DT aerosol data for 2000-2009,
647 the use of the newly released C6 Terra MODIS DT aerosol products introduces a marginal
648 differences in derived global and regional AOT trends.
- 649 2. Using the near full data record from Terra (2000-2015), Aqua (2002-2015), and MISR
650 (2000-2015), global and regional AOT trends are derived using over ocean C6 MODIS DT
651 and MISR data. A negligible AOT trend ($0.0003 \text{ AOT decade}^{-1}$) is found using Aqua C6
652 MODIS DT data, but a higher AOT trend of $0.008 \text{ AOT decade}^{-1}$ is found using Terra C6
653 MODIS DT data, while a slight negative trend is derived using MISR data (-0.005 AOT
654 decade^{-1}). It is suspected that the difference may be introduced by calibration related issues
655 for one or all sensors, such as the recently reported cross-talk in thermal channels for Terra
656 MODIS (Moeller and Frey, 2016), and a slight decrease in signal sensitivity for Terra

657 MISR (Limbacher and Kahn, 2016). After accounting for potential calibration drifts,
658 negligible AOT trends are found over global oceans using data from all sensors.

659 3. Regionally, statistically significant increases in aerosol loading over time are found over
660 regions such the Indian Bay of Bengal, Arabian Sea, and the Red Sea. Statistically
661 significant negative AOT trends are also found over the eastern US coast and
662 Mediterranean Sea. This is in agreement from all three sensors (MODIS Aqua, MODIS
663 Terra and MISR).

664 4. Using collocated MODIS and CERES data over global oceans, the SW flux (*SWARE*)
665 trends are also estimated for the near-full Terra (2000-2015) and Aqua (2002-2015) data
666 records. After accounting for the potential calibration / angular distribution models
667 (ADMs) / near surface wind related issues, small negative (*positive*) trends of -0.5 to -0.6
668 $\text{Wm}^{-2} \text{decade}^{-1}$ ($0.5 - 0.6 \text{ Wm}^{-2} \text{decade}^{-1}$) are found for Terra based analysis and a near zero
669 trend is found for using Aqua data, and the results are rather consistent regardless of using
670 CERES SSF or ES-8 SW fluxes. Regionally, positive SW flux trends are found over
671 regions such as the Bay of Bengal and Arabian Sea, where statistically significant negative
672 trends are found over the eastern US coast and Mediterranean Sea. The signs of the
673 regional SW flux trends are in good agreement to what has been found for AOT trends.

674 5. Very high correlations are found between MODIS DT AOT and CERES cloud-free SW
675 flux values using $2 \times 2^\circ$ (Latitude/Longitude) gridded multi-year mean Terra (2000-2015)
676 and Aqua (2002-2015) data. The SW aerosol direct forcing efficiency is estimated to be -
677 60 Wm^{-2} per AOT and a *SWARE* value of -7 Wm^{-2} is derived over global oceans. The
678 results are consistent, regardless of using Terra or Aqua data, or using of CERES ES-8 or
679 SSF data. Regionally, over the southwest coast of Africa, where smoke aerosol particles

680 dominate in summer months, a SW aerosol direct forcing efficiency value of $\sim -40 \text{ Wm}^{-2}$
681 per AOT is found, again, regardless of datasets used. SW aerosol direct forcing efficiency
682 values of -50 to -80 Wm^{-2} per AOT are also found for Arabian Sea, northwest coast of
683 Africa, coastal China and Indian Bay of Bengal, where dust and pollutant aerosols
684 dominate. It also worth noting that a non-linear relationship is found between SWARE
685 and AOT.

686 6. Factors that could impact SWARE trend analysis include cloud contamination, calibration
687 drifts, ADMs, ocean wind patterns, and optically thin cirrus (OTC) clouds. The largest
688 sources of uncertainty in the derived SWARE trends are found to be related to
689 ADMs/surface wind speeds, while cloud contamination has a minor impact on the
690 estimated SWARE trends.

691 7. Finally, trend analyses from this study are inter-compared with results from several
692 selected studies (e.g., Zhang and Reid, 2010; Alfaro-Contreras et al, 2016; Toth et al.,
693 2016). Consistency in increasing/decreasing AOT trends is found among the studies, using
694 passive and active based instruments, over cloud free and cloudy skies, as well as using
695 narrowband and broadband observations over regions such as the Bay of Bengal, Arabian
696 Sea, the eastern US coast and Mediterranean Sea. Note that the above mentioned studies
697 are derived with different instruments that have different sampling methods with different
698 uncertainties under different observing conditions. The fact that consistencies are found
699 from those studies, adds fidelity to some of the studies that are difficult to evaluate
700 otherwise.

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859 Table 1. List of datasets used in the study.

Datasets	Study periods	Purposes
C6 Aqua MODIS DT	July 2002- Dec. 2015	AOT trend, cloud fraction
C6 Terra MODIS DT	Mar. 2000 - Dec. 2015	AOT trend, cloud fraction
Terra MISR	Mar. 2000 - Dec. 2015	AOT trend
C6 Aqua CERES-ES-8-SSF	July 2002 - Dec. 2015	Cloud free SW flux trend
C6 Terra CERES-ES-8-SSF	Mar. 2000 - Dec. 2015	Cloud free SW flux trend
CALIOP	June 2006 - Nov. 2015	Thin cirrus cloud mask

860 Table 2. AOT trend analysis for global and selected regions as suggested from Zhang and Reid, 2010. Both trends from Collection 5
861 (C5, Zhang and Reid, 2010) and Collection 6 (C6) over-water Terra MODIS AOT data are shown for the study period of 2000-2009.
862 The trend significances are derived using two different methods ($|\omega/\sigma_\omega|$ and $|z|$ values as estimated from the Weatherhead and Mann-
863 Kendall methods, respectively). The corrected slopes refer to the slopes after accounting for the slope changes over the Remote Ocean
864 region. AOT trend and trend significances for C5 MODIS DT data are obtained from Zhang and Reid, (2010), which are derived
865 using Data-assimilation Quality C5 MODIS DT data. For illustration purposes, C5* and C6 Terra MODIS AOT values, derived using
866 similar methods as mentioned in this study, are also listed. C5* MODIS DT AOTs listed here are not from the Data-assimilation
867 quality products as used in Zhang and Reid (2010).

Region	Latitude	Longitude	Slope AOT / decade Terra		Trend Significance (Terra)			Corrected Slope AOT/decade (Terra)		Mean AOT (Terra)	
			C5	C6	C5 $ \omega/\sigma_\omega $	C6 $ \omega/\sigma_\omega $	C6 $ Z $	C5	C6	C5*	C6
Global			0.010	0.011	3.60	4.85	6.88	0.003	0.005	0.154	0.140
Africa (NW Coast)	8°N - 24°N	60°W - 18°W	-0.006	-0.004	0.61	0.37	0.18	-0.013	-0.010	0.247	0.257
Bay of Bengal	10°N - 25°N	78°E - 103°E	0.076	0.074	5.63	4.79	4.71	0.069	0.068	0.319	0.326
Coastal China	20°N - 40°N	110°E - 125°E	0.069	0.086	4.06	4.69	4.78	0.062	0.080	0.460	0.462
Central America	5°N - 20°N	120°W - 90°W	-0.016	-0.011	1.73	1.12	0.57	-0.023	-0.017	0.151	0.165
Arabian Sea	5°N - 23°N	50°E - 78°E	0.065	0.077	5.40	4.95	4.03	0.058	0.071	0.319	0.329
Mediterranean Sea	30°N - 45°N	0° - 40° E	-0.009	-0.009	0.94	0.96	1.25	-0.016	-0.015	0.200	0.210
Africa (SW. Coast)	23°S - 7°S	20°W - 15°E	0.016	0.018	1.35	1.52	1.46	0.009	0.012	0.179	0.188
N. America (E. Coast)	30°N - 45°N	80°W - 60° W	-0.008	-0.010	1.07	1.50	1.04	-0.015	-0.016	0.157	0.160
Africa (SE. coast)	27° - 15°S	32°E - 45°E	0.017	0.015	2.12	1.93	3.06	0.010	0.009	0.129	0.158
Southeast Asia	15°S - 10°N	80°E - 120°E	0.014	0.016	0.80	0.86	4.89	0.007	0.010	0.176	0.184
Remote Ocean	40°S - 0°	179°W - 140°W	0.007	0.006	N/A	2.32	2.93	0	0	0.100	0.107

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871 Table 3. Multi-year AOT and Cloud-Free Flux trends (2002-2015 for MODIS Aqua; 2000-2015 for MODIS Terra; and 2000-2015 for
872 MISR) for global and selected regions. AOT trends are calculated using monthly-averaged, deseasonalized AOTs derived from the
873 MODIS collection 6 and MISR aerosol products. Cloud-free flux trends are calculated using seasonally-averaged, deseasonalized
874 cloud-free fluxes derived using the collocated MODIS-CERES SSF/ES-8 data set. Various filtering criteria are applied to the data and
875 described in the text. Trends that are statistically significant with a confidence interval of 95% (utilizing the Weatherhead method for
876 monthly-averages, and the Mann-Kendall method for seasonal-averages) are highlighted in bold.

Regional	Lat.	Lon.	AOT decade ⁻¹			Corrected AOT /decade			Cloud-Free Flux wm ⁻² decade ⁻¹		Corrected Cloud- Free Flux wm ⁻² decade ⁻¹	
			MODIS Aqua	MODIS Terra	MISR	MODIS Aqua	MODIS Terra	MISR	Aqua	Terra	Aqua	Terra
									ES-8	ES-8	ES-8	ES-8
									SSF	SSF	SSF	SSF
Global			0.0003	0.008	-0.005	~0	0.002	-0.001	-0.50 -0.26	-1.50 -1.22	0 -0.01	-0.58 -0.52
Africa (NW Coast)	8°N - 24°N	60°W - 18°W	0.002	0.009	-0.008	0.002	0.003	-0.004	0.56 0.71	-1.79 -1.29	1.06 0.96	-0.87 -0.59
Bay of Bengal	10°N – 25°N	78°E –103°E	0.031	0.056	0.018	0.031	0.050	0.022	2.28 1.91	0.79 0.75	2.78 2.16	1.71 1.45
Coastal China	20°N – 40°N	110°E – 125°E	-0.035	0.007	-0.014	-0.035	0.001	-0.01	-0.42 0.04	-2.51 -2.09	0.08 0.29	-1.59 -1.39
Central America	5°N – 20°N	120°W – 90°W	0.007	0.002	-0.011	0.007	-0.004	-0.007	-0.45 -0.11	-1.85 -1.33	0.05 0.14	-0.93 -0.63
Arabian Sea	5°N – 23°N	50°E – 78°E	0.039	0.057	0.033	0.039	0.051	0.037	2.61 2.24	0.90 0.94	3.11 2.49	1.82 1.64
Mediterranean Sea	30°N – 45°N	0° - 40° E	-0.025	-0.014	-0.029	-0.025	-0.020	-0.025	-0.91 -0.72	-2.93 -2.46	-0.41 -0.47	-2.01 -1.76
Africa (SW Coast)	23°S – 7°S	20°W – 15°E	0.016	0.025	0.002	0.016	0.019	0.006	-0.19 0.13	-0.85 -0.57	0.31 0.38	0.07 0.13
East Coast North America	30°N – 45°N	80°W – 60° W	-0.028	-0.016	-0.026	-0.028	-0.022	-0.022	-2.29 -1.65	-3.57 -2.73	-1.79 -1.40	-2.65 -2.03
Africa	27°- 15°S	32°E - 45°E	0.010	0.017	-0.0001	0.010	0.011	0.004	-0.01	-1.46	0.49	-0.54

(SE Coast)										-0.25	-1.27	0	-0.57
S.E. Asia	15°S - 10°N	80°E - 120°E	0.013	0.020	0.004	0.013	0.014	0.008	0.02	0.60	-1.07	0.52	-0.15
Remote Ocean	40°S - 0°	179°W - 140°W	0.0003	0.006	-0.004	0	0	0	-0.50	-0.92	0	0	0
Red Sea	15°N - 30°N	30°E - 45°E	0.081	0.100	0.041	0.081	0.094	0.045	2.52	-0.27	3.02	0.65	2.08
Persian Gulf	24°N - 30°N	50°E - 60°E	0.033	0.081	0.046	0.033	0.075	0.050	1.76	-0.64	2.26	0.28	1.16
									1.16	-0.92	1.41	-0.22	

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878 Table 4. Instantaneous SW aerosol direct forcing efficiencies estimated based on the multi-year means (2000-2015 for Terra and
879 2002-2015 for Aqua) as well as for Dec.-May and Jun.-Nov. seasons using both CERES SSF and ES-8 datasets. Forcing efficiencies
880 are calculated for selected regions that have the maximum 2x2° (Latitude/Longitude) bin-averaged AOT > 0.3, as well as for global
881 oceans. The multi-year mean AOT and SWARE values are estimated using data from all valid bins. Note that values from this table
882 are estimated under CERES cloud free (less 1% cloud fraction, and 99% CP) skies and thus regional and global AOT values may be
883 different from the estimates as shown in Table 2.

	Dec.-May (Wm ² /AOT)		Jun.-Nov. (Wm ² /AOT)		Multi-year Mean (Wm ² /AOT)		Multi-year Mean AOT (0.55 μm)		Multi-year Mean SWARE (Wm ²)	
	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra	Aqua	Terra
	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8	SSF / ES-8
Africa (NW Coast)	-54.1/ -67.0	-52.7/ -63.0	-59.5/ -75.2	-61.1/ -75.9	-54.4 / -65.9	-54.3 / -62.9	0.189 / 0.189	0.204 / 0.204	-10.3 / -12.5	-11.1 / -12.8
Africa (SW Coast)	N/A/ N/A	N/A/ N/A	-40.6/ -44.3	-43.0/ -45.0	-37.8 / -39.8	-42.5 / -43.7	0.160 / 0.160	0.158 / 0.158	-6.0 / -6.4	-6.7 / -6.9
Coastal China	-88.3/ -83.8	-84.0/ -82.4	-74.7/ -74.5	-70.8/ -74.4	-79.0 / -79.5	-74.3 / -79.7	0.293 / 0.293	0.356 / 0.356	-23.2 / -23.3	-26.5 / -28.4
Arabian Sea	-62.0/ -75.3	-66.0/ -75.0	-60.0/ -76.0	-60.6/ -76.5	-61.6 / -76.0	-65.2 / -77.4	0.215 / 0.215	0.238 / 0.238	-13.3 / -16.4	-15.5 / -18.4
Bay of Bengal	-66.4/ -69.3	-52.8/ -63.3	-68.4/ -74.8	-58.4/ -67.8	-74.8 / -80.0	-52.3 / -63.1	0.261 / 0.261	0.295 / 0.295	-19.5 / -20.9	-15.4 / -18.6
Global Oceans	-58.7/ -57.9	-57.3/ -59.4	-56.5/ -59.4	-53.7/ -57.2	-60.9 / -57.7	-57.5 / -58.2	0.116 / 0.116	0.116 / 0.116	-7.1 / -6.7	-6.7 / -6.8

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887 Table. 5 List of uncertainty sources (in $\text{Wm}^{-2} \text{decade}^{-1}$) for the estimated cloud-free SW flux trends.

Region / Sensitivity Test	ES-8/SSF Cloud-free flux trends ($\text{Wm}^{-2} \text{decade}^{-1}$)		Standard Deviation ($\text{Wm}^{-2} \text{decade}^{-1}$)	
	Terra ES-8/SSF	Aqua ES-8/SSF	Terra ES-8/SSF	Aqua ES-8/SSF
Global Oceans / No Data Trim	-1.50/-1.22	-0.50/-0.26	0.09/0.08	0.08/0.03
Remote Ocean / No Data Trim				
R.O. Region Outline				
Lat : 40°S - 0° Lon: 180°W - 140°W	-0.92/-0.70	-0.50/-0.25		
Lat: 40°S - 0° Lon: 170°E - 150°W	-1.00/-0.79	-0.47/-0.23		
Lat: 40°S - 0° Lon: 170°W - 130°W	-0.84/-0.63	-0.43/-0.20		
Lat : 50°S - 10°S Lon: 180°W - 140°W	-0.89/-0.67	-0.43/-0.25		
Lat : 30°S - 10°N Lon: 180°W - 140°W	-1.08/-0.81	-0.62/-0.29		
Global Ocean / Variation of CF %			0.04/0.03	0.03/0.01
0 <0.5 <1 <2 <3 <4 <5	-1.46 -1.49 -1.50 -1.51 -1.54 -1.56 -1.57	-0.46 -0.51 -0.52 -0.54 -0.54 -0.55 -0.55		
	-1.22 -1.24 -1.24 -1.25 -1.27 -1.29 -1.30	-0.24 -0.28 -0.28 -0.28 -0.27 -0.26 -0.26		
Global Ocean / Variation of CP %			0.02/0.08	0.01/0.05
100 >99 >98 >97 >96 >95	-1.44 -1.48 -1.49 -1.49 -1.49 -1.49	-0.54 -0.55 -0.54 -0.53 -0.52 -0.52		
	-1.13 -1.26 -1.30 -1.32 -1.34 -1.35	-0.29 -0.23 -0.20 -0.17 -0.16 -0.15		
Global Ocean / Cirrus Contamination			0.08/0.05	0.08/0.05
MODIS-CERES-CALIOP		-0.59/-0.33		
MODIS-CERES-CALIOP (cirrus filtered)		-0.48/-0.26		
ADMs / Wind Speeds			0.20/0.20	0.17/0.17
Global Full Data Record (ES-8)	-1.50	-0.50		
Global Full Data Record (SSF)	-1.22	-0.26		
Overall Uncertainty			0.3/0.3 Wm ⁻² / decade	0.2/0.2 Wm ⁻² / decade

889 Table 6. Inter-comparison of AOT (AOT decade⁻¹) and SW flux (Wm⁻² decade⁻¹) trends from this study as well as a few previous
 890 studies at both regional and global scales.

	Zhang and Reid (2010) Terra MODIS C5 March 2000- Dec. 2009		This Study Terra MODIS C6 March 2000 – Dec. 2015				Toth et al., 2016 Aqua CALIOP Cloud-Free June 2006 – Dec. 2014	Alfaro-Contreras et al., 2016 Aqua CALIOP Above-Cloud June 2006 – Dec. 2014
Region	ΔAOT /Decade		ΔAOT /Decade		Δ Cloud-Free Flux Wm ⁻² decade ⁻¹ (ES-8/SSF)		ΔAOT /Decade	ΔAOT/Decade
	w/o correction	w/ correction	w/o correction	w/ correction	w/o correction	w/ correction		
Global Ocean	0.010	0.003	0.008	0.002	-1.50/-1.22	-0.58/-0.52	0.002	0.005
Africa (NW Coast)	-0.006	-0.013	0.009	0.003	-1.79/-1.29	-0.87/-0.59	-0.014	0.0007
Bay of Bengal	0.076	0.069	0.056	0.050	0.79/0.75	1.71/1.45	0.016	0.079
Coastal China	0.069	0.062	0.007	0.001	-2.51/-2.09	-1.59/-1.39	-0.017	0.01
Arabian Sea	0.065	0.058	0.057	0.051	0.90/0.94	1.82/1.64	0.027	0.055
Med. Sea	-0.009	-0.016	-0.014	-0.020	-2.93/-2.46	-2.01/-1.76	-0.006	-0.010
Africa (SW Coast)	0.016	0.007	0.025	0.019	-0.85/-0.57	0.07/0.13	0.009	0.007
N. America (East Coast)	-0.008	-0.015	-0.016	-0.022	-3.57/-2.73	-2.65/-2.03	-0.013	-0.02
Remote Ocean	0.007	0	0.006	0	-0.92/-0.70	0/0		0.005

891 **Figure Captions**

892 **Figure 1.** Spatial distribution of trends for (a) over ocean Terra MODIS DT AOT for 2000-2009,
893 (b) over ocean Terra MODIS DT AOT for 2000-2015, (c) over ocean Aqua MODIS DT AOT for
894 2002-2015 and (d) over land and ocean Terra MISR AOT for 2000-2015 for every $1^\circ \times 1^\circ$ bin. (e)
895 Ratios of MODIS C6 to C5 AOT trends for the study period of 2000-2009, and (f) Differences in
896 MODIS C6 to C5 AOT trends for the study period of 2000-2009. Regions with statistically
897 significant trends at a confidence interval of 95% are highlighted with black dots. Figs. 1e and 1f
898 are constructed with the use of grids with AOT trends above or below ± 0.0002 AOT/year.

899
900 **Figure 2.** (a) The global distribution of daytime AOTs constructed using sixteen years (2000-
901 2015) of monthly-averaged over ocean C6 Terra MODIS AOTs at a $1^\circ \times 1^\circ$ resolution. Only those
902 bins with more than one thousand data counts were considered for this analysis. (b) Similar to Fig.
903 2a, but using over ocean C6 Aqua MODIS AOTs for the study period of 2002-2015. (c) Similar to
904 Fig. 2a, but using both over ocean and over land Terra MISR AOT data for the study period of
905 2000-2015. (d) Differences in gridded AOTs between Terra MODIS and Terra MISR. (e) The
906 ratios of gridded AOTs between Terra MODIS and Terra MISR.

907
908 **Figure 3.** (a) Monthly-averaged global AOTs derived using operational MODIS C6 aerosol
909 products for Aqua (red), Terra (blue) and MISR (green). Straight lines are the linear fits for the
910 monthly data. (b) Similar to Fig. 3a, but for the deseasonalized, monthly-averaged AOTs. (c)
911 Similar to Fig. 3b, but for the Remote Ocean region as described in Table 3.

912

913 **Figure 4.** The deseasonalized, monthly and regionally averaged AOTs for eight selected regions
914 utilizing operational MODIS C6 and MISR aerosol products. Straight lines are linear fits to the
915 monthly data.

916

917 **Figure 5.** (a) The spatial distribution of seasonally-averaged AOTs using Terra MODIS DT AOT
918 data from the collocated Terra MODIS-CERES dataset for the study period of 2000-2015, at a
919 spatial resolution of $2 \times 2^\circ$ (Latitude/Longitude). (b) Similar to Figure 5a but using the collocated
920 Aqua MODIS-CERES dataset for the study period of 2002-2015. (c) Seasonally averaged CERES
921 ES-8 cloud-free SW flux constructed using the collocated Terra MODIS-CERES dataset for the
922 study period of 2000-2015. (d) Similar to Figure 5c, but using the collocated Aqua MODIS-
923 CERES dataset for the study period of 2002-2015. (e-f) Similar to Figs 5c and 5d but for the
924 seasonally-averaged CERES SSF cloud-free SW fluxes. (g) Difference between cloud-free SW
925 flux from Figure 5e and 5c. (h) Similar to Fig. 5g, but for Aqua. (i) Collocated Terra MODIS-
926 CERES data counts for every $2^\circ \times 2^\circ$ (Latitude/Longitude) bin. (j) Similar to Fig. 5i, but for Aqua.
927

928 **Figure 6.** (a) Scatter plot of Aqua MODIS AOT versus CERES SSF SW flux (at a $2 \times 2^\circ$
929 resolution) using data as shown in Fig. 5. Color lines are for selected regions and the black thick
930 line is for global oceans. (b) Similar to Fig. 6a, but for 5 selected regions that have a maximum
931 AOT > 0.3 as indicated from Fig. 5. (c) Similar to Fig. 6a, but for Terra. (d) Similar to Fig. 6b,
932 but for Terra.

933

934 **Figure 7.** Similar to Fig. 6, but for using collocated MODIS and CERES ES-8 cloud-free SW flux
935 data.

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937

938 **Figure 8.** (a) Time series of seasonally-averaged, deseasonalized cloud-free SW fluxes over
939 global oceans utilizing the collocated MODIS-CERES (SSF/ES-8) datasets for Terra (green) and
940 Aqua (red). (b) Similar to Fig. 8a but using data from the Remote Ocean region. The ES-8 SW
941 fluxes are depicted by solid lines where SSF SW fluxes are depicted by dashed lines.

942

943 **Figure 9.** Time series of all-sky SW flux over the entire globe (land and ocean). The trends are
944 calculated from monthly-globally averaged all-sky SW fluxes derived from the CERES SSF / ES-
945 8 data. SW fluxes from all scenes including cloudy, clean, land and ocean are taken into account
946 when calculating the monthly averages, which are gridded into a similar resolution as the
947 collocated MODIS-CERES dataset ($2 \times 2^\circ$).

948

949 **Figure 10.** The temporal variations of deseasonalized, seasonally- and regionally- averaged
950 CERES SSF / ES-8 cloud-free fluxes (seasonal anomaly) for 8 selected regions, constructed using
951 the collocated Aqua and Terra MODIS-CERES datasets. The blue lines represent the Terra-based
952 analysis while the red lines represent the Aqua-based analysis and the solid lines represent the ES-
953 8 SW fluxes where the SSF SW fluxes are depicted by dashed lines.

954

955 **Figure 11.** The de-seasonalized, seasonally averaged cloud-free fluxes over the Coastal China
956 region derived utilizing the collocated MODIS-CERES (SSF / ES-8) datasets. Straight lines show
957 piecewise linear fits for the study periods of 2000-2015 (Terra only).

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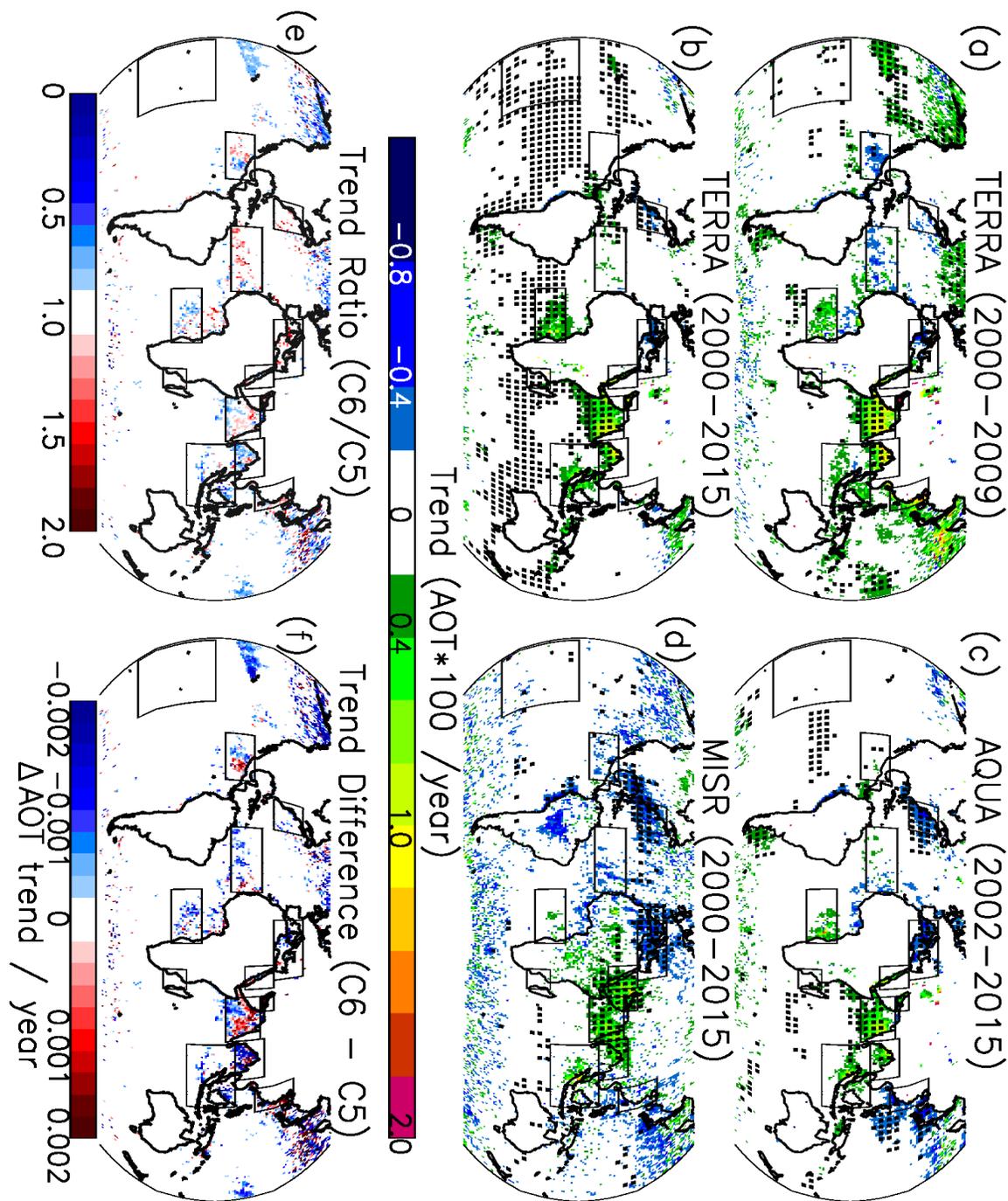
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960 14 year Aqua (2002-2015) for every $4 \times 4^\circ$ (Latitude/Longitude) bin derived from the collocated
961 MODIS-CERES dataset. AOT trends are constructed using seasonally-averaged AOTs. (c) Spatial
962 distribution of cloud-free-sky CERES ES-8 SW flux trends estimated using the collocated Terra
963 MODIS-CERES data for the study period of 2000-2015. (d) Similar to Figure 12c, but using the
964 collocated Aqua MODIS-CERES (ES-8) dataset for the study period of 2002-2015. (e-f) Similar
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966 AOT/clear-sky SW flux trends at the 95 % confidence interval are shown in black dots.

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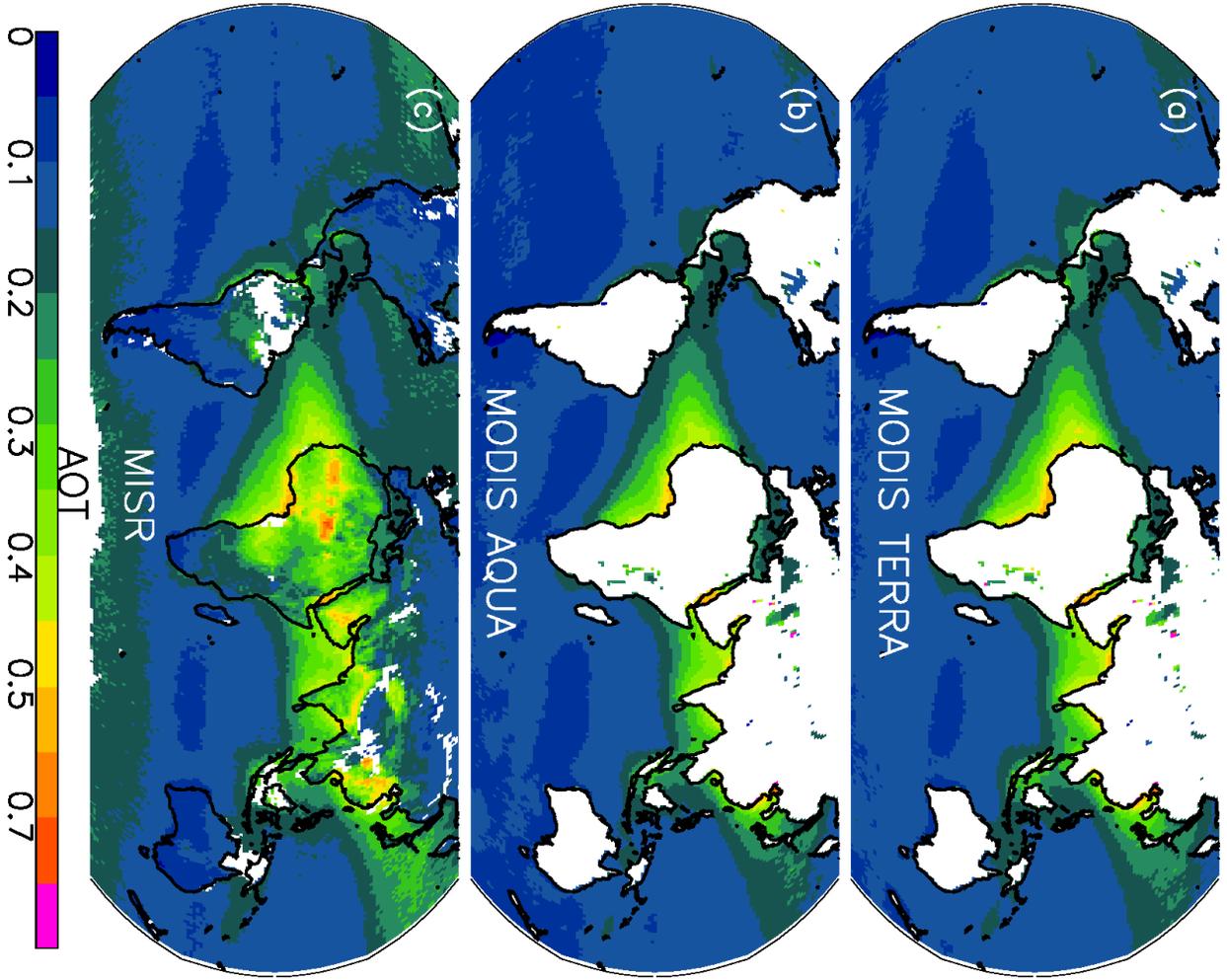
968 **Figure 13.** Global AOT trends derived from the (red) MODIS-CERES dataset, (green) MODIS-
969 CERES-CALIOP dataset and (blue) MODIS-CERES-CALIOP dataset after filtering for cirrus
970 clouds. Both CERES SSF and ES-8 data are included. Time series have been derived utilizing
971 seasonal AOT averages. CALIOP is used to locate and remove CERES observations contaminated
972 with cirrus clouds. (b) Depicts the same thing as Fig. 13a, except for the cloud-free flux. This
973 analysis is carried out for the Aqua-based study only.

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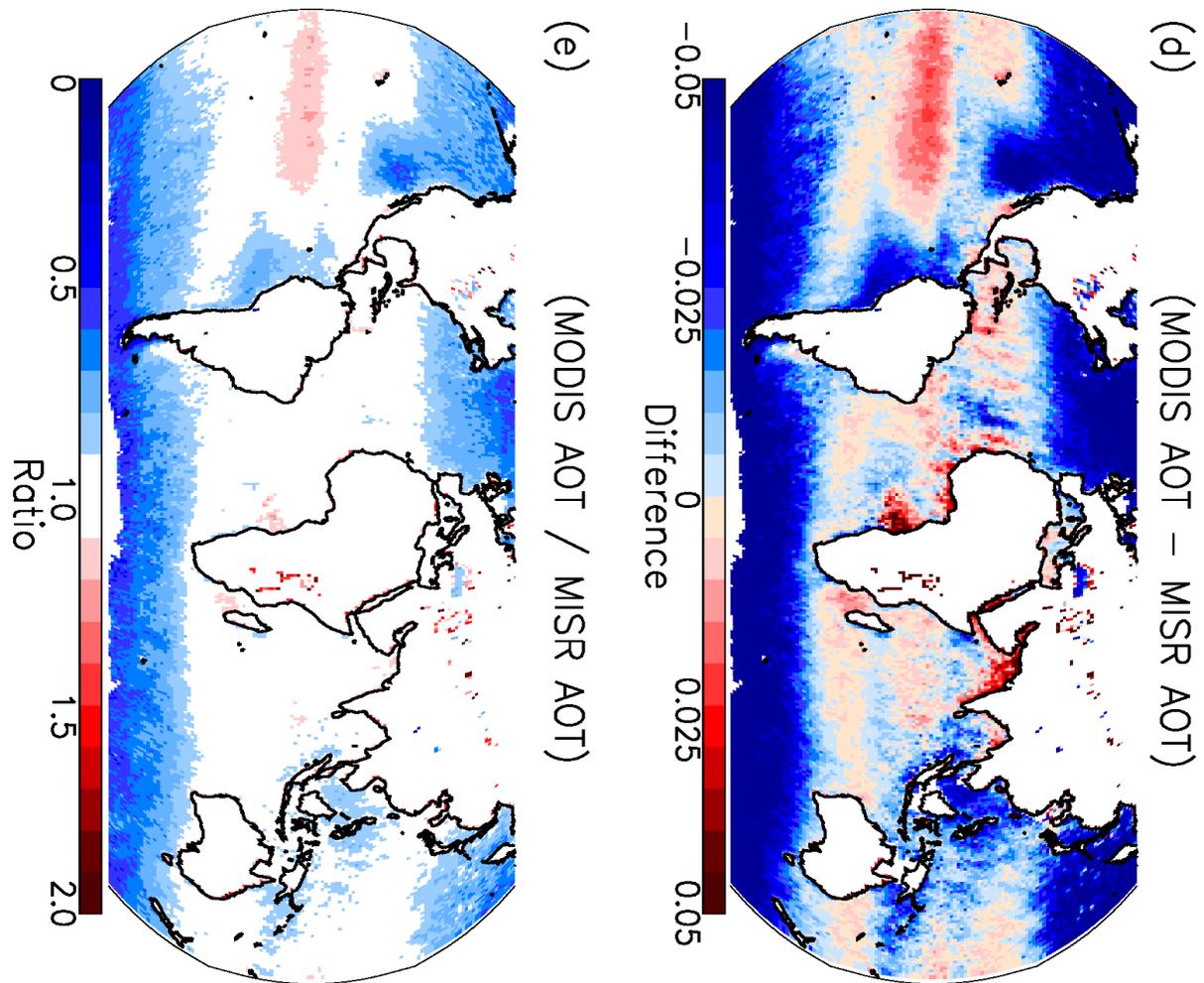
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 983 are constructed with the use of grids with AOT trends above or below ± 0.0002 AOT/year.

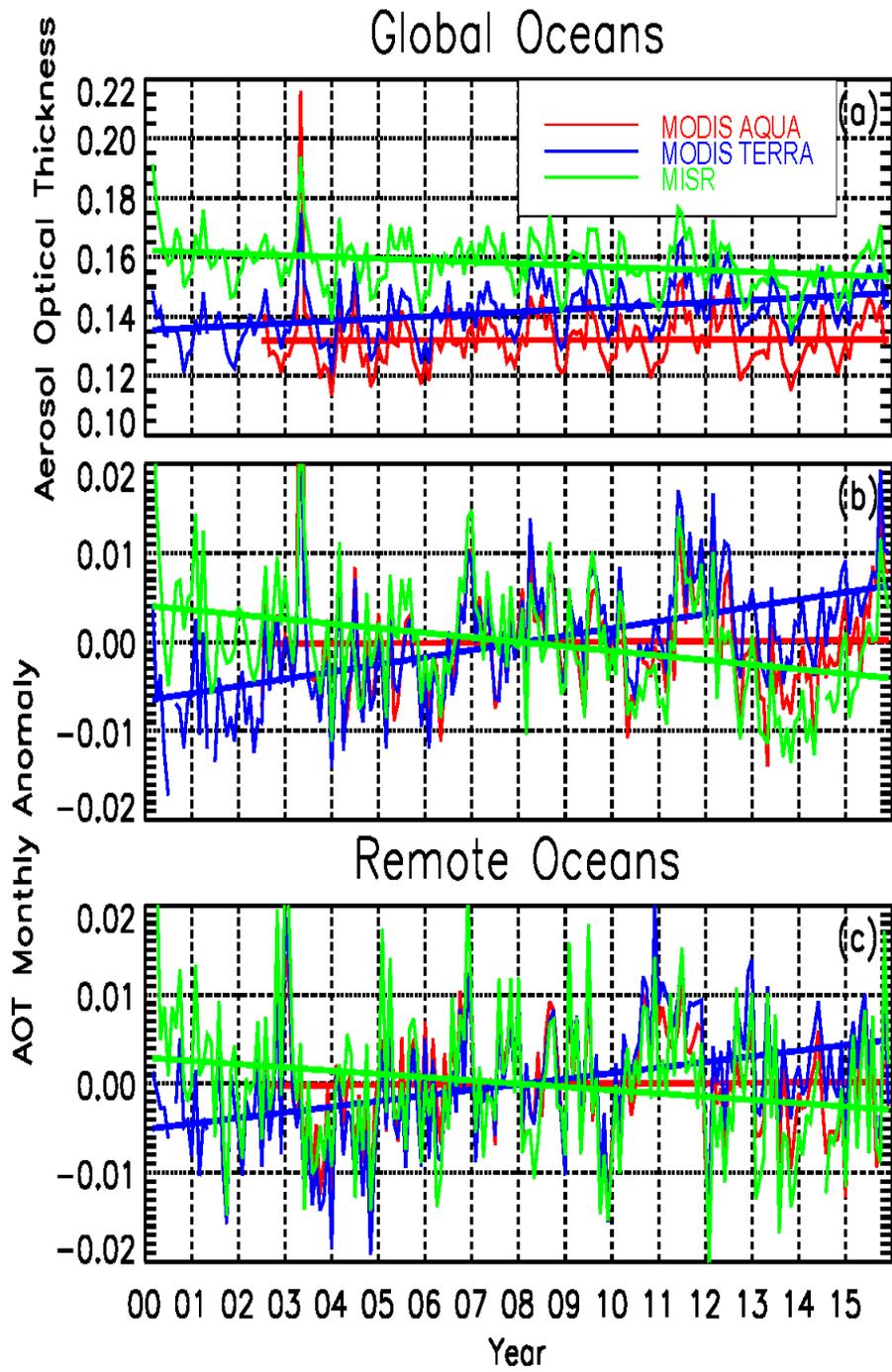


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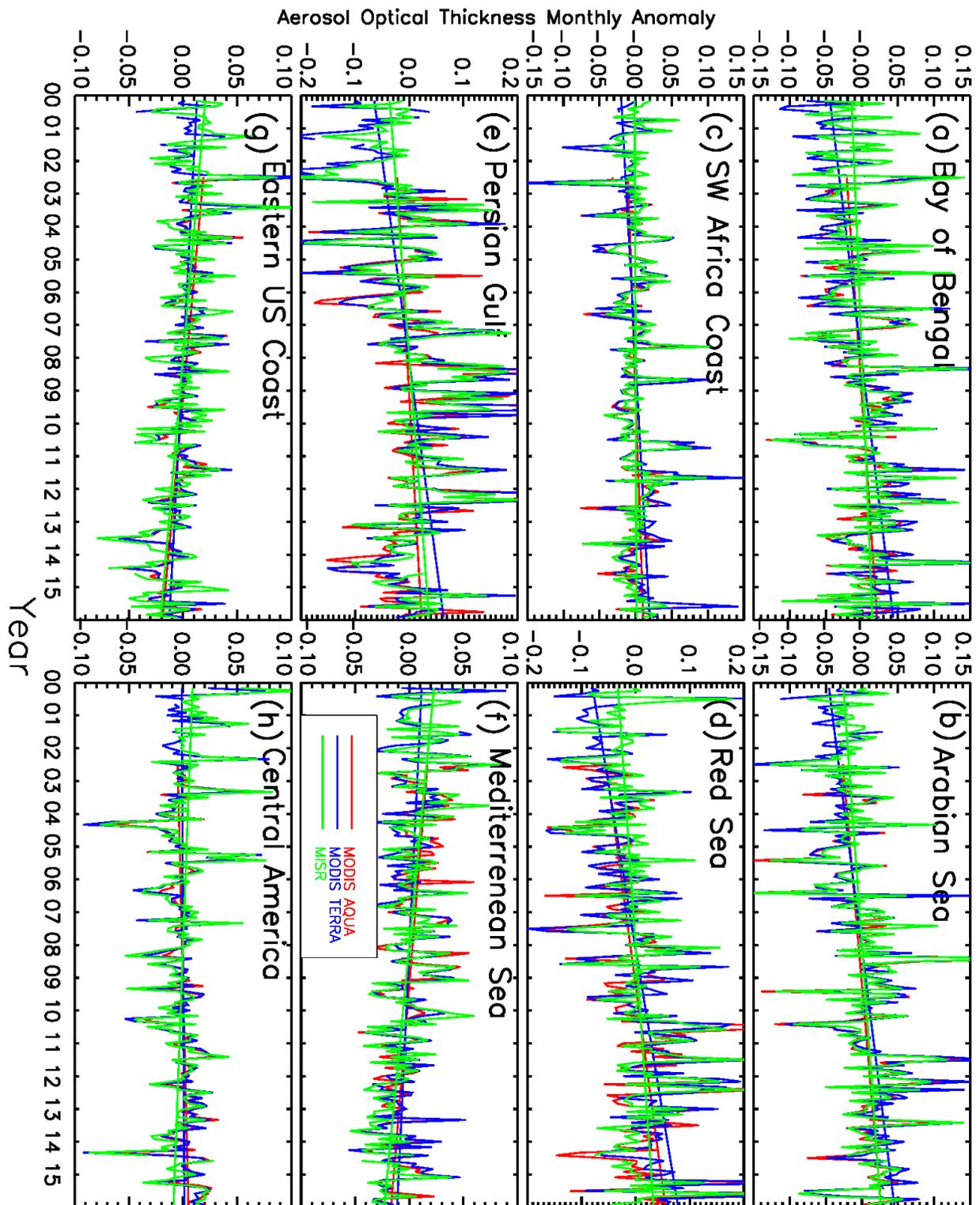


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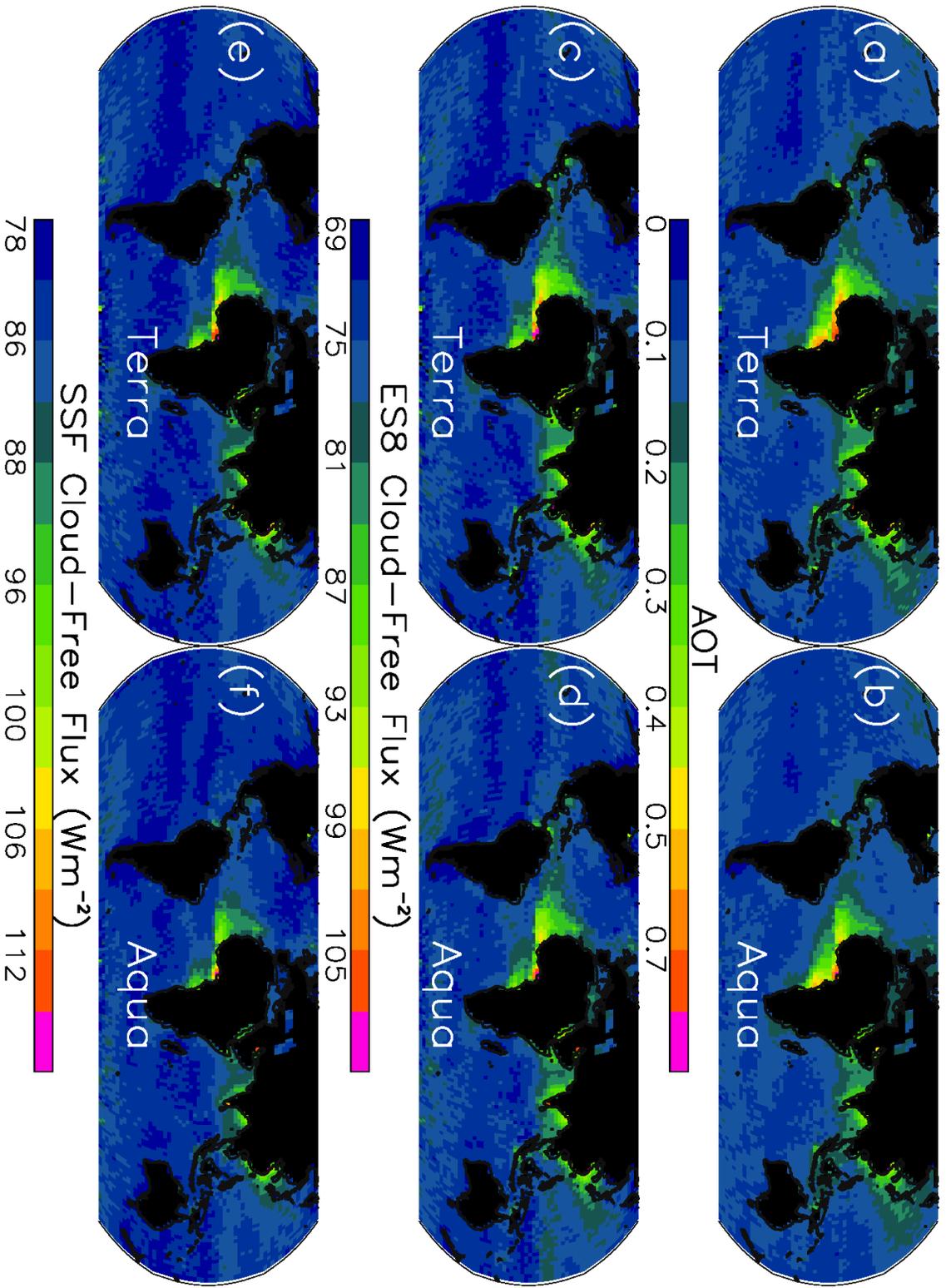
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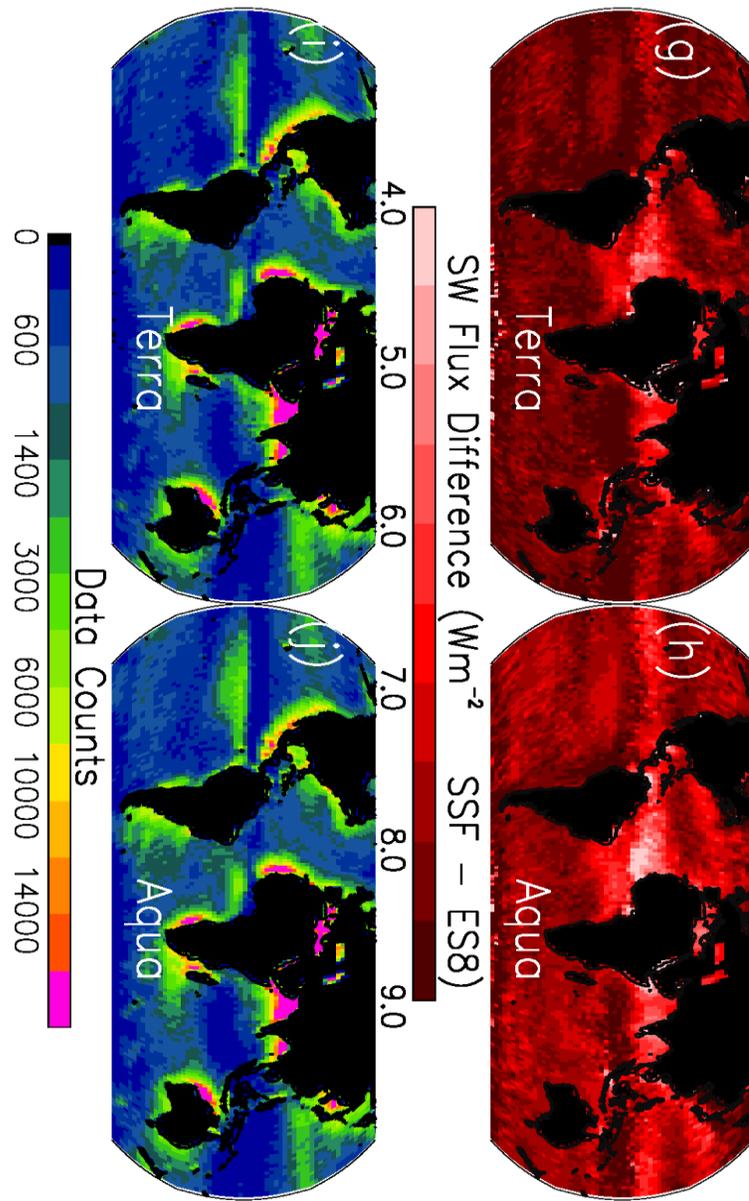
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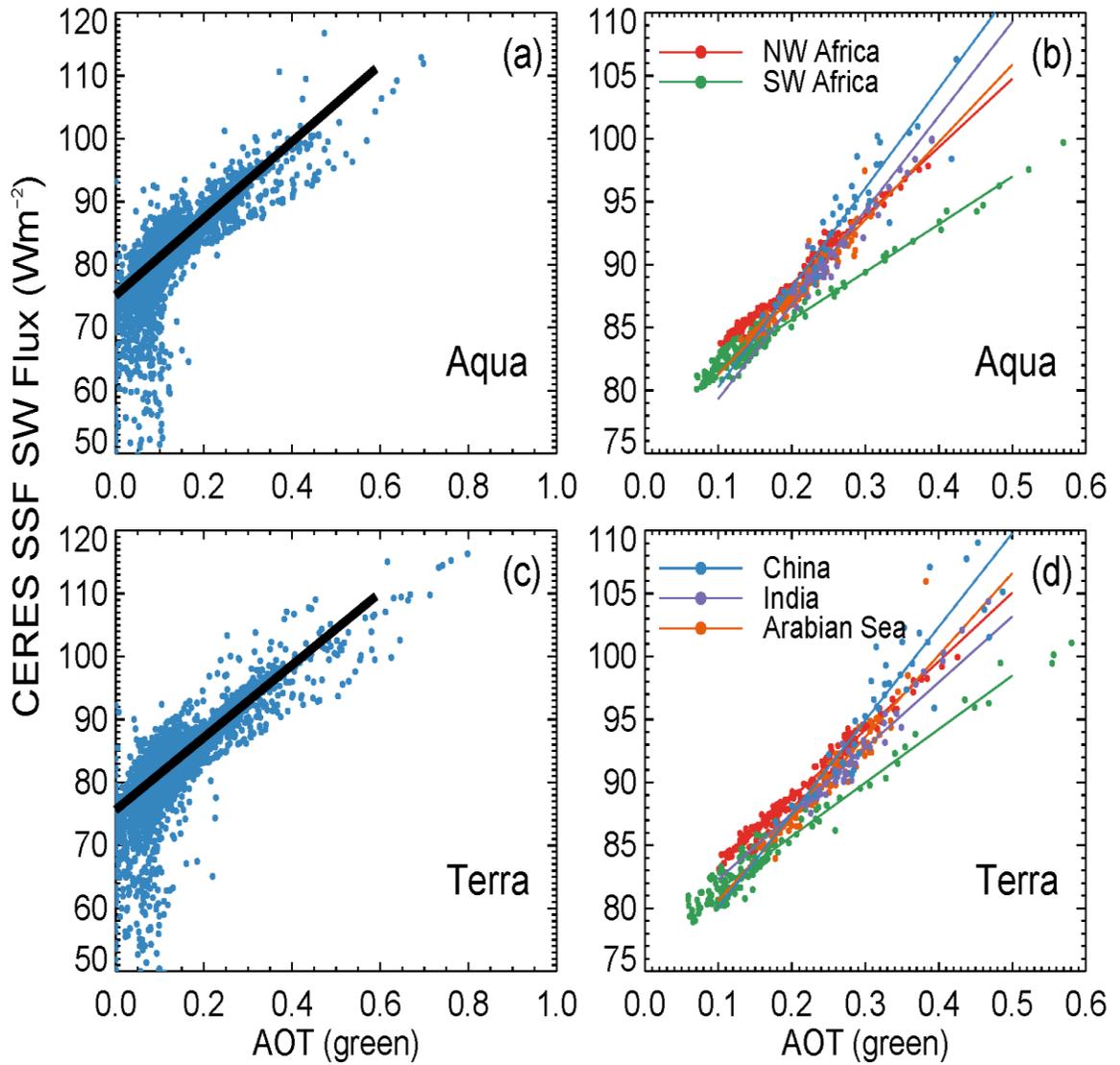
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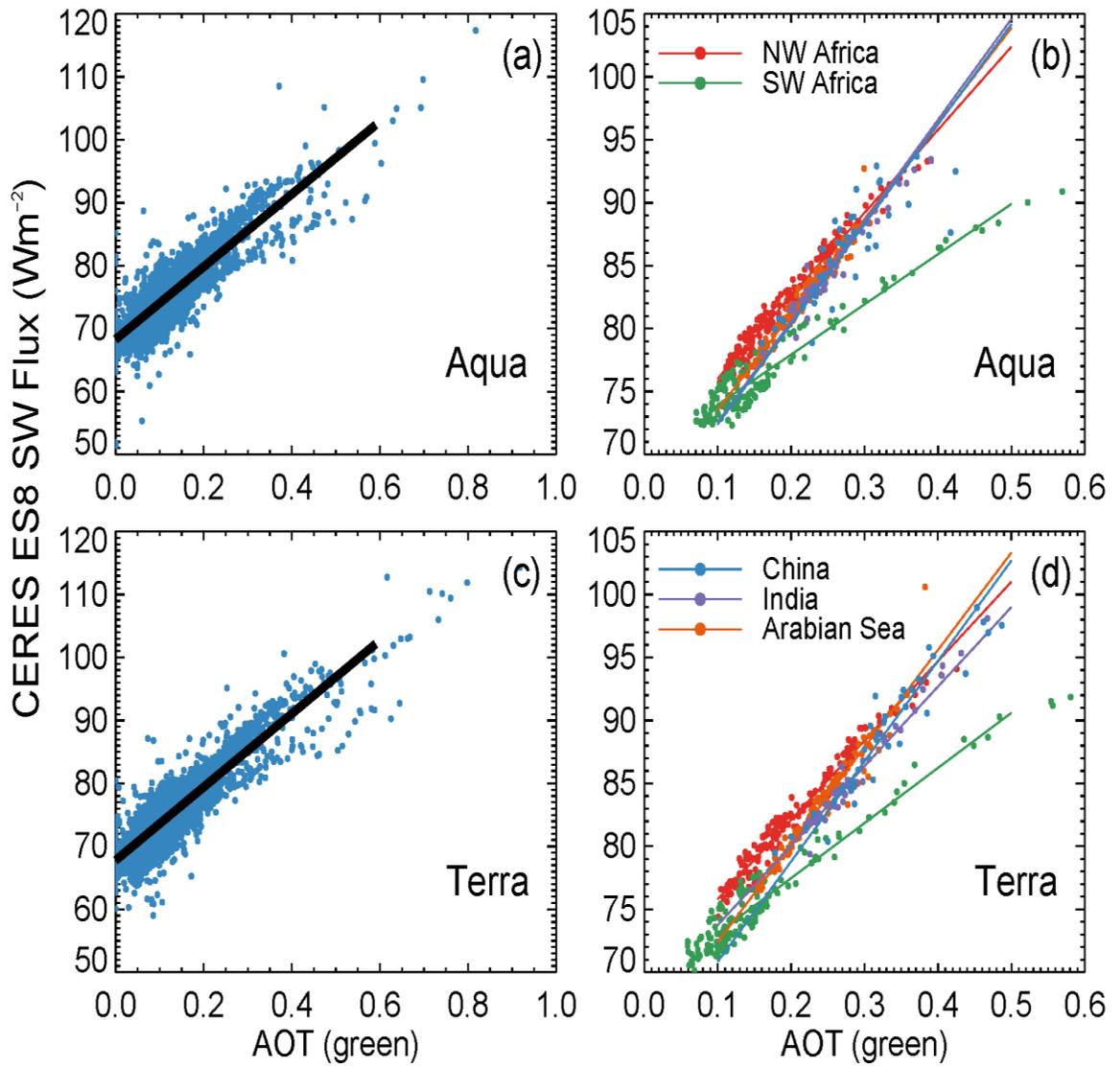
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 1011 Aqua MODIS-CERES dataset for the study period of 2002-2015. (c) Seasonally averaged CERES
 1012 ES-8 cloud-free SW flux constructed using the collocated Terra MODIS-CERES dataset for the
 1013 study period of 2000-2015. (d) Similar to Figure 5c, but using the collocated Aqua MODIS-
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 1015 seasonally-averaged CERES SSF cloud-free SW fluxes. (g) Difference between cloud-free SW
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1018
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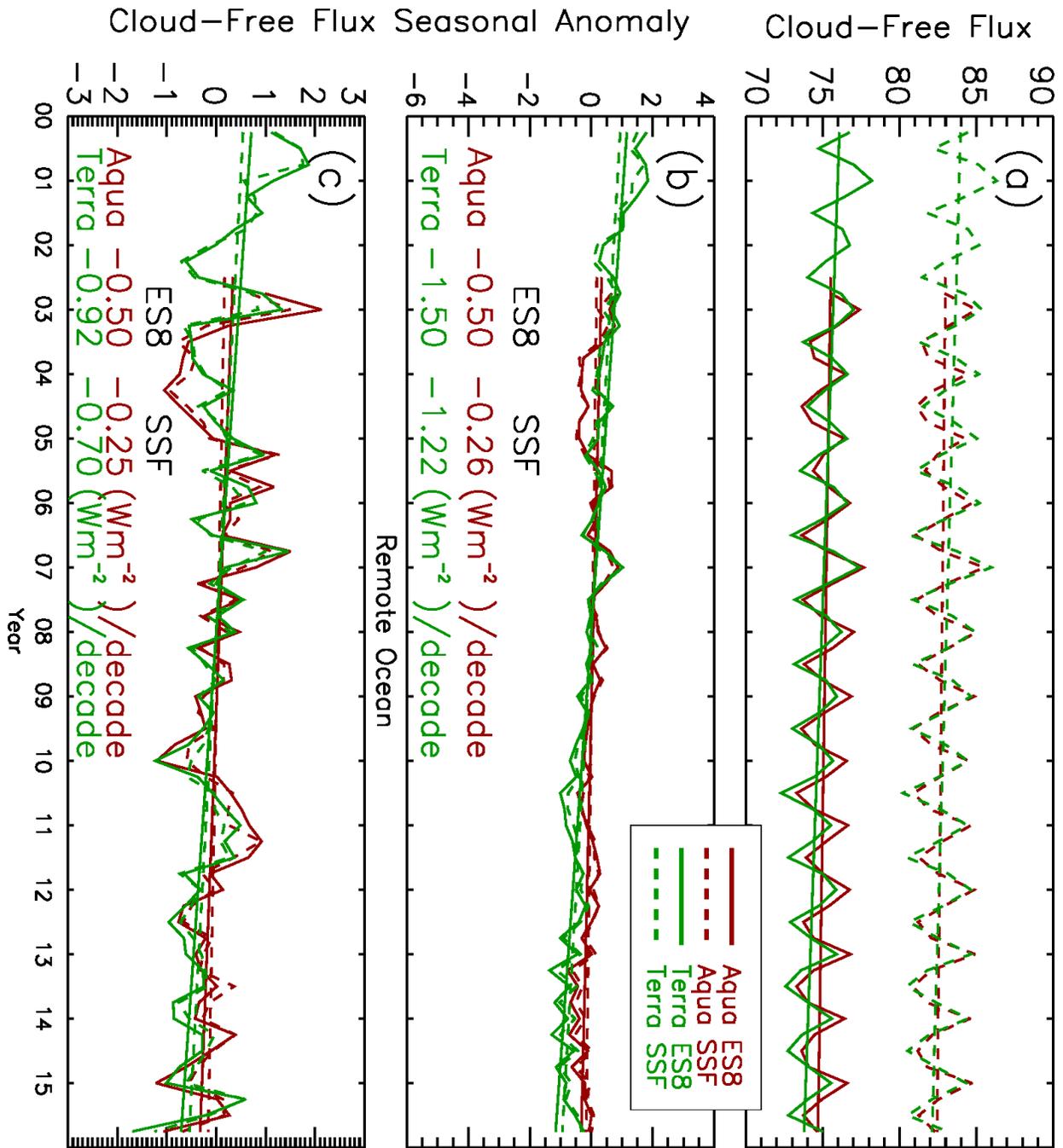
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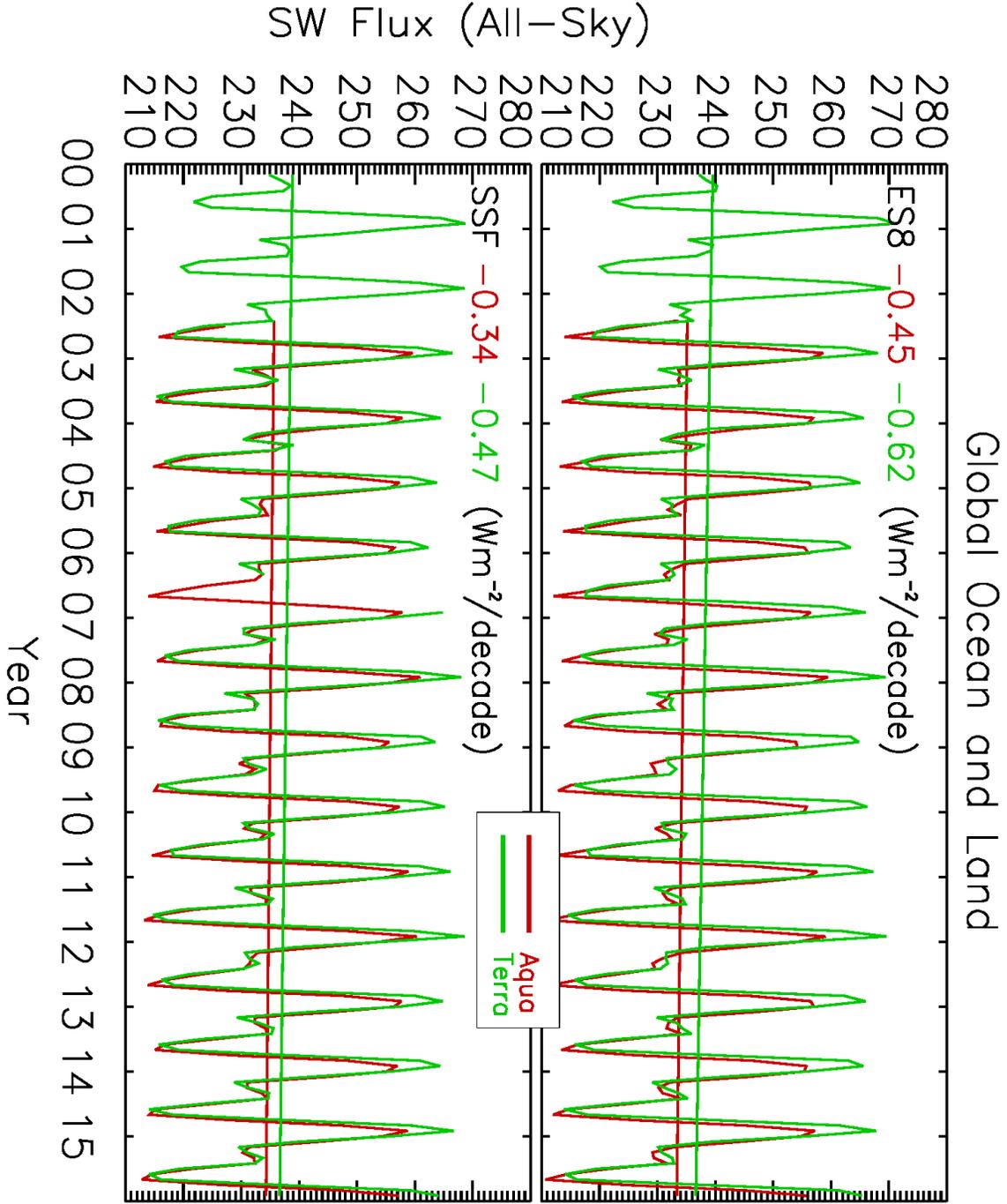
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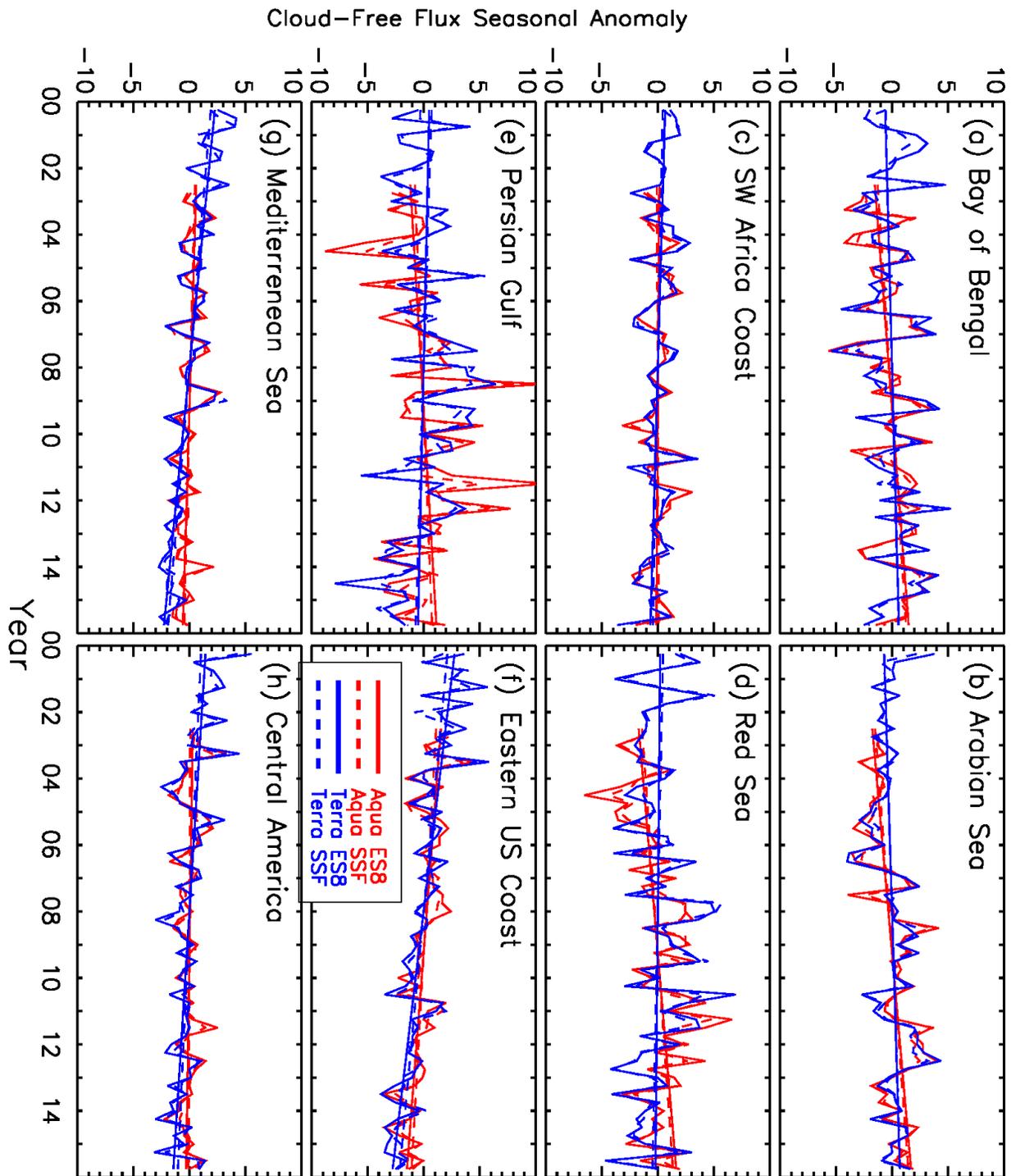
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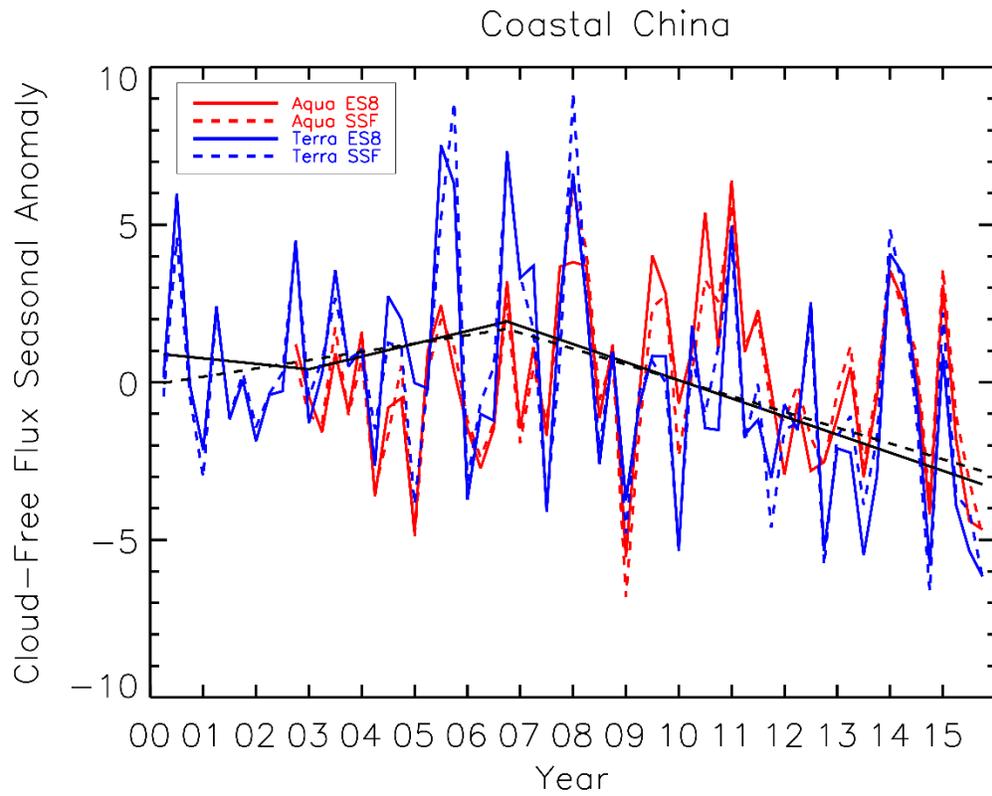
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 1048 collocated MODIS-CERES dataset (2 x 2°).
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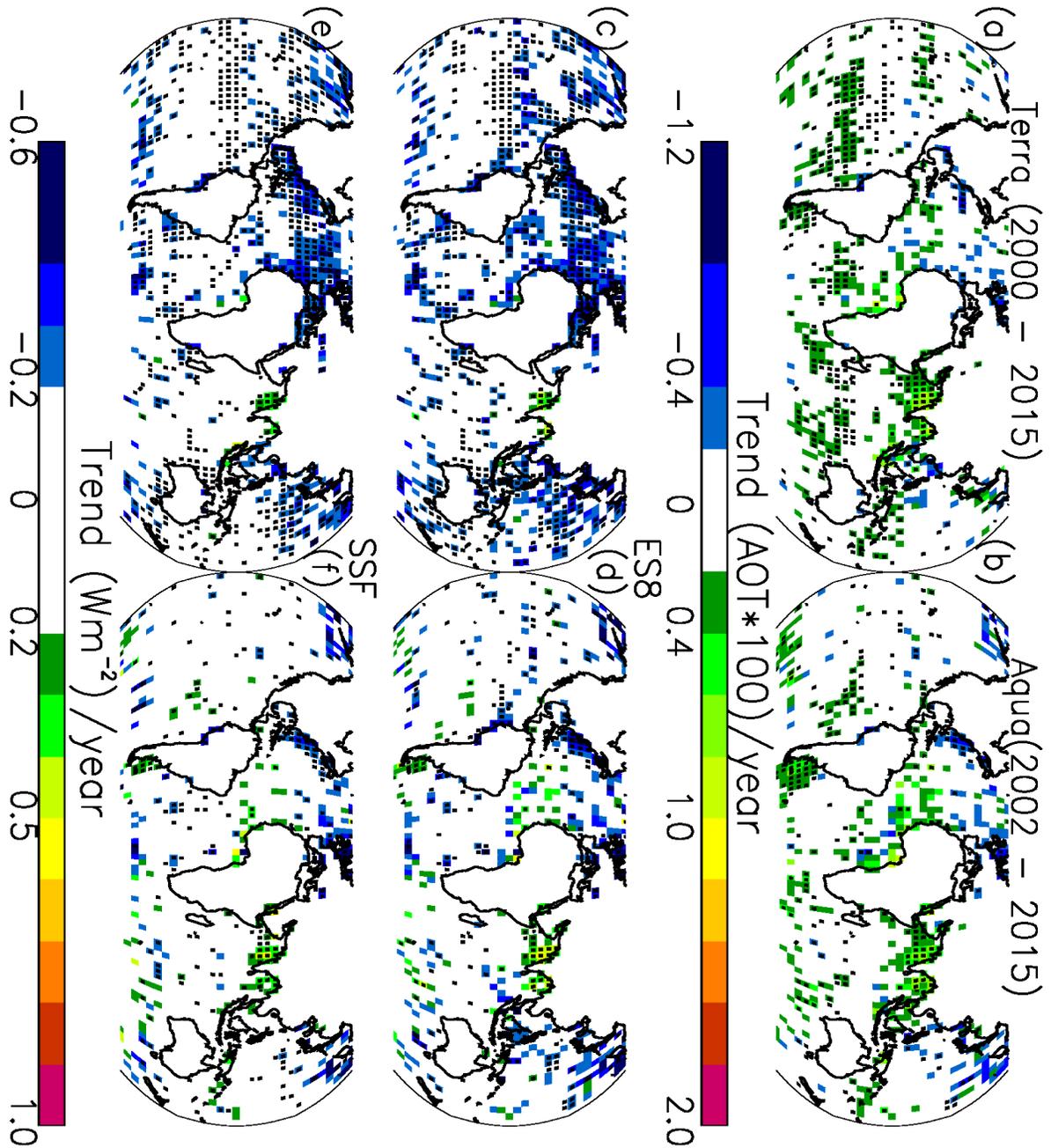
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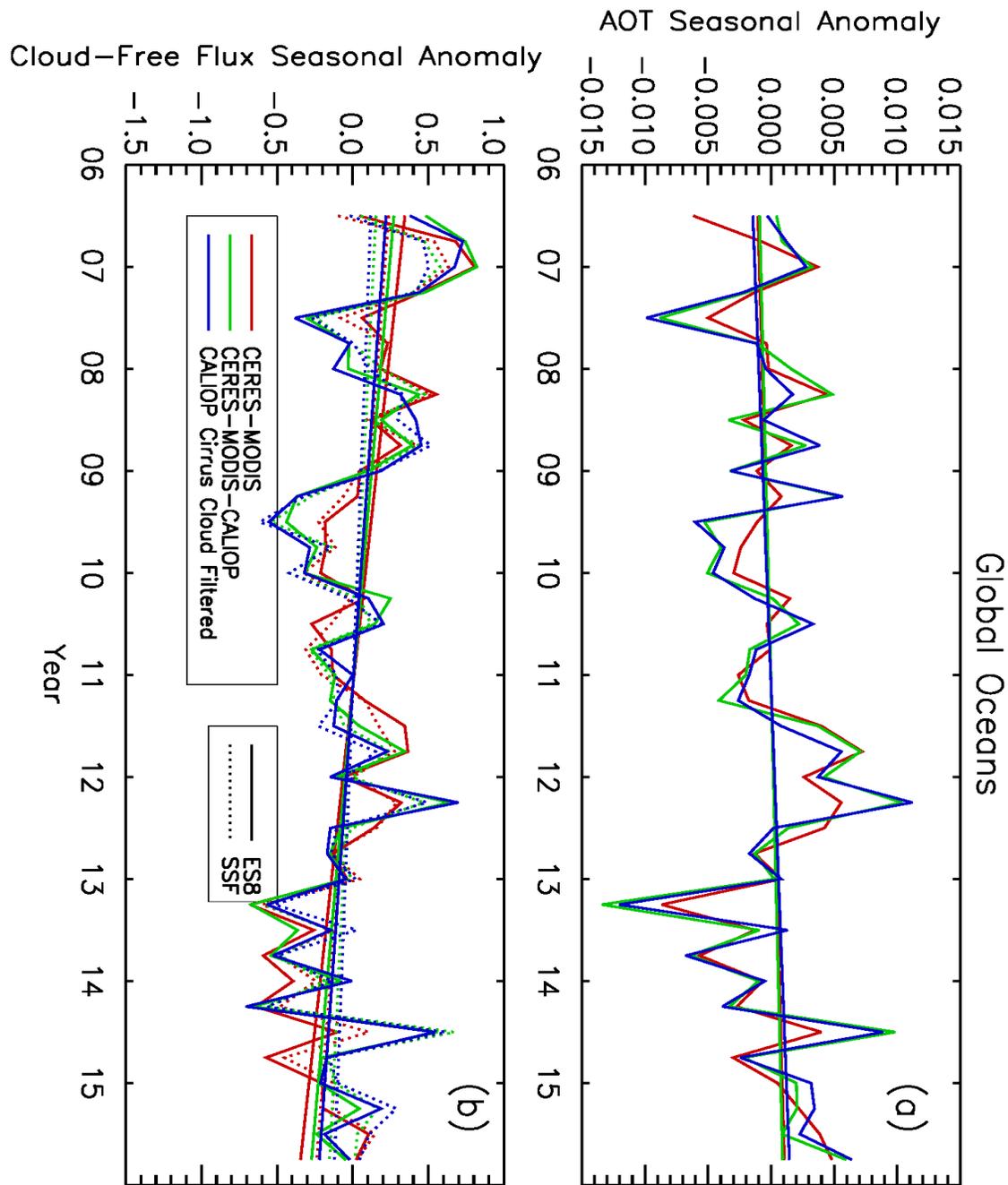
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1069

1070 **Figure 13.** Global AOT trends derived from the (red) MODIS-CERES dataset, (green) MODIS-
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