

1 **Changes in TOA SW Fluxes over Marine Clouds When Estimated via**
2 **Semi-Physical Angular Distribution Models**

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ABSTRACT

11 Top-of-atmosphere (TOA) shortwave (SW) angular distribution models (ADMs) approximate –
12 per angular direction of an imagined upward hemisphere – the intensity of sunlight scattered back
13 from a specific Earth-atmosphere scene. ADMs are, thus, critical when converting satellite-borne
14 broadband radiometry into estimated radiative fluxes. This paper applies a set of newly developed
15 ADMs with a more refined scene definition and demonstrates tenable changes in estimated fluxes
16 compared to currently operational ADMs. Newly developed ADMs use a semi-physical framework
17 to consider cloud-top effective radius, \bar{R}_e , and above-cloud water vapor, $ACWV$, in addition to
18 accounting for surface wind speed and clouds' phase, fraction, and optical depth. In effect,
19 instantaneous TOA SW fluxes for marine liquid-phase clouds had the largest flux differences (of up
20 to 25 W m^{-2}) for lower solar zenith angles and cloud optical depth greater than 10 due to extremes
21 in \bar{R}_e or $ACWV$. In regions where clouds had persistently extreme levels of \bar{R}_e (here mostly for
22 $\bar{R}_e < 7 \mu\text{m}$ and $\bar{R}_e > 15 \mu\text{m}$) or $ACWV$, instantaneous fluxes estimated from Aqua, Terra, and Meteosat
23 8 and 9 satellites using the two ADMs differed systematically, resulting in significant deviations in
24 daily mean fluxes (up to $\pm 10 \text{ W m}^{-2}$) and monthly mean fluxes (up to $\pm 5 \text{ W m}^{-2}$). Flux estimates
25 using newly developed, semi-physical ADMs may contribute to a better understanding of solar
26 fluxes over low-level clouds. It remains to be seen whether aerosol indirect effects are impacted by
27 these updates.

28 **1. Introduction**

29 Clouds reflect and absorb sunlight and, thus, crucially modify how much light reaches the
30 surface, which atmospheric vertical levels experience heating from light absorption, and how
31 much light eventually leaves the Earth-atmosphere-system through the top-of-atmosphere (TOA).
32 Representing cloud reflection and absorption in climate models is challenging as both processes
33 are function of the amount and phase of cloud condensate, its micro-physical character (Twomey
34 1977), and its inhomogeneous three-dimensional structure (Cahalan et al. 1994; Barker et al. 1996;
35 Hogan et al. 2019). This complexity, combined with the need for computationally efficient radiative
36 transfer calculations require climate models to make simplifying assumptions (e.g. Fu and Liou
37 1992; Clough et al. 2005; Bender et al. 2006; Pincus et al. 2003). The benchmark to assess the
38 realism of a climate models' radiative response is TOA radiative fluxes (Ramanathan 1987; Bony
39 et al. 1992; Li et al. 2013) as they can be estimated from many satellites that have been sampling
40 vast regions frequently for more than 50 years (Dewitte and Clerbaux 2017).

41 Marine boundary-layer clouds are particularly relevant as they – and their relatively high albedo
42 – replace an otherwise dark (i.e. low albedo) ocean surface. These clouds are potentially exposed
43 to natural and anthropogenic sources of aerosol such as biomass burning events closer to continents
44 (Painemal et al. 2014) or ship exhaust along merchant routes (e.g. Toll et al. 2017). Acting as cloud
45 condensation nuclei, aerosol can redistribute cloud condensate locally towards more numerous,
46 smaller droplets that reflect sunlight more efficiently (Twomey 1977). Ensuring that we understand
47 the cloud radiative effect of boundary-layer clouds is important, as their dynamics and therefore
48 their temporal evolution is in large part determined by cloud-top radiative cooling (Lilly 1968;
49 Wood 2012).

50 To estimate TOA solar fluxes from satellite observations, three components are required: 1)
51 knowledge of the underlying scene properties of surface and atmospheric constituents which are
52 usually retrieved from an on-board multi-spectral imager or collocated from auxiliary data sets
53 if irretrievable; 2) broadband radiance that is either inferred from narrow multi-spectral channels
54 across the solar spectrum or preferably measured via an on-board broadband radiometer; and 3) a
55 carefully designed model with assumptions about how solar radiation will be scattered into different
56 angular directions of an imagined upward hemisphere so that the broadband radiance, measured
57 at one angular direction can be transformed into a broadband flux (Suttles et al. 1988; Smith et al.
58 1986; Loeb et al. 2003, 2005; Su et al. 2015). The angular variation of reflected solar radiation
59 is referred to as anisotropy and can vary considerably across scenes and sun-observer geometries.
60 For clouds over ocean (the central topic of this study) any scene with a cloud fraction smaller than
61 100% may encompass specular reflection from the water surface that changes the intensity and
62 angular distribution as higher surface wind speeds roughen the surface and create tilted reflecting
63 facets (Cox and Munk 1954). Overcast cloudy scenes distribute sunlight differently with cloud
64 optical depth as increased multi-scattering leads to a more Lambertian-like reflection (e.g. Gao et al.
65 2013). Tornow et al. (2020) developed a method to incorporate cloud micro-physical characteristics
66 (represented through cloud-top effective radius \bar{R}_e) and amount of absorbing above-cloud water
67 vapor (*ACWV*) into anisotropy-predicting angular distribution models (ADMs).

68 Any ADM development that further refines the definition of Earth-atmosphere scenes, and
69 therefore enables consideration of additional effects impacting anisotropy, warrants a look at how
70 resulting flux estimates compare against the current standard (demonstrated briefly for past ADM
71 developments in e.g. Su et al. 2015). And as newly estimated fluxes are potentially more accurate
72 and may help the community improve their understanding of cloud-aerosol-radiation interaction,
73 newly developed ADMs have not only been applied to upcoming satellite missions but also to the

74 existing wealth of past and current satellite observations. For example, such development-driven
75 reprocessing led to several versions of the widely used CERES SSF (Cloud and Earth’s Radiant
76 Energy System Single Scanner Footprints) data product (e.g. Smith et al. 2011, or as documented
77 in https://ceres.larc.nasa.gov/data/documentation/#historical-editions_).

78 This study makes a case for newly developed, semi-physical ADMs to be considered as the next-
79 generation solution and to refine flux estimates in past, current, and future missions. We investigated
80 whether TOA shortwave (SW) fluxes are significantly different when using currently operational
81 ADMs, employed in most-recent CERES SSF Edition 4A (Loeb et al. 2005; Su et al. 2015),
82 versus newly developed, semi-physical ADMs (Tornow et al. 2020). Analysis of instantaneous flux
83 estimates found differences of up to 25 W m^{-2} in cases of extremes in $\overline{R_e}$ and $ACWV$. To investigate
84 impacts across larger regions and longer time scales, we processed two months of polar-orbiting
85 and geostationary satellite observations over the tropical and southern Atlantic where there are
86 often low-level clouds (e.g. Cesana et al. 2019) of varying microphysical properties (e.g. Bennartz
87 and Rausch 2017) as well as above-cloud water vapor. Daily and monthly mean fluxes indicate
88 that systematic instantaneous differences between the ADMs can propagate into time means with
89 significant differences of up to 10 W m^{-2} for daily means and up to 5 W m^{-2} for monthly means.

90 This paper is organized as follows. Section 2 introduces ADMs, their estimated uncertainty, and
91 the satellite data used as input for the ADMs. Section 3 presents our main results and Section 4
92 discusses them.

93 **2. Materials and Methods**

94 The following subsections explain the fundamentals of currently operational as well as newly
95 developed, semi-physical ADMs and how we obtained anisotropy from them (Sec. 2a), the way

96 we approximate uncertainty of flux estimates (Sec. 2b), and the satellite observations we used as
97 input for both ADMs (Sec. 2c).

98 *a. Empirical Angular Distribution Models for Marine Clouds*

99 Empirical ADMs describe the expected angular distribution of sunlight reflected back towards
100 TOA based on CERES-MODIS observations (Wielicki et al. 1996) collected over a 5-year period
101 (2000-2005) by taking measurements using a rotational azimuth plane scan mode. An imagined
102 upward hemisphere was discretized into 2° angular elements, forming an angular bin per joint
103 combination of solar zenith angle θ_0 , viewing zenith angle θ_v , and relative azimuth angle φ interval
104 (their intervals denoted by superscript Δ generally or by letters when pointing at specific intervals).
105 For each angular bin, available CERES SSF (described in more detail in Section 2c) samples
106 were used to train the currently operational, sigmoidal approach (Loeb et al. 2005; Su et al. 2015)
107 described in Sec. 2a1, and newly developed, semi-physical approach (Tornow et al. 2020) discussed
108 in Sec. 2a2 – each striving to minimize residuals between radiance-predicting models and CERES-
109 MODIS observations. The key difference between the two approaches is scene definition and
110 parameters used to define a scene that may result in different TOA SW anisotropies for the same set
111 of measurements. We reproduced the currently operational CERES approach to ensure consistent
112 fusion with simulated radiances (Sec. 2a3) and coherent anisotropy estimation (Sec. 2a4).

113 1) CURRENTLY OPERATIONAL ADMs USING SIGMOIDAL FITS

114 Footprint level data from the CERES SSF were first classified into groups of MODIS-retrieved
115 cloud phases: ice phase, liquid phase, and mixed phase. Note that CERES SSF footprints can
116 contain a layer of ice and a layer of liquid condensate with some horizontal displacement so both
117 layers are visible from space. Per cloud phase, scenes were defined in a continuous manner through

118 the product of layer cloud optical depth $\tilde{\tau}$ (the exponential of the average over logarithmic τ values
 119 over a cloud layer retrieved by MODIS) and cloud fraction f . A sigmoid curve was used to fit
 120 CERES-observed radiances I and the parameter $x = \log \tilde{\tau} f$ which is based on MODIS retrievals

$$I(\theta_0^\Delta, \theta_v^\Delta, \varphi^\Delta) = I_0 + \frac{a}{\left[1 + e^{-\frac{(x-x_0)}{b}}\right]^c} \quad (1)$$

121 where I_0 , a , b , c , and x_0 were free parameters.

122 In case of low cloud fraction or low optical thickness ($x < 6$) and viewing geometries potentially
 123 affected by sun-glint (i.e. sun-glint angles $< 20^\circ$), a look-up-table for that angular bin stored the
 124 average of observed TOA SW radiances per interval of x (intervals: <3.5 , $3.5-4.5$, $4.5-5.5$, $5.5-6$)
 125 and 10 m wind speed (intervals: $0-2$, $2-4$, $4-6$, $6-8$, $8-10$, and > 10 m s $^{-1}$). This approach is
 126 currently used in CERES SSF Edition 4A to estimate fluxes (Su et al. 2015).

127 2) NEWLY DEVELOPED, SEMI-PHYSICAL APPROACH

128 Like the currently operational CERES approach (Sec. 2a1), cloudy footprints were separated
 129 by their phase and scene definition was a continuous function of the following parameters. Per
 130 cloud phase, a semi-physical approach served to define scenes via a footprint albedo α and via
 131 an absorption through above-cloud water vapor $ACWV$. The footprint albedo incorporated each
 132 footprint's clear portion (that involved a clear-sky albedo α^{ocean} as well as potential sun-glint
 133 contribution r^{SunGlint} predicted from a Cox-Munk model using 10 m wind speed) as well as up to
 134 two cloudy portions (each with a statistically predicted two-stream cloud albedo $\alpha_{1,2}^{\text{TwoStream}}$ using
 135 MODIS-retrieved $\tilde{\tau}_{1,2}$, $\overline{R}_{e,1,2}$) together weighted by their respective cloud fraction. This approach
 136 was used in its log-linear form for ordinary least square fitting to CERES-MODIS observations to
 137 find optimal, free parameters A , B , and C :

$$\log I(\theta_0^\Delta, \theta_v^\Delta, \varphi^\Delta) \approx A + B \cdot \log \alpha + C \cdot ACWV \quad (2)$$

138 With footprint albedo α

$$\alpha = f_0 \cdot (\alpha^{\text{ocean}} + r^{\text{SunGlint}}) + f_1 \cdot \alpha_1^{\text{TwoStream}} + f_2 \cdot \alpha_2^{\text{TwoStream}} \quad (3)$$

139 and using the functional form of the two-stream cloud albedo $\alpha^{\text{TwoStream}}$ to ingest MODIS-inferred
140 $\tilde{\tau}$ and \bar{R}_e :

$$\alpha^{\text{TwoStream}} = \frac{\alpha^{\text{ocean}} + (1 - \alpha^{\text{ocean}})(1 - g)\tilde{\tau}/2}{1 + (1 - \alpha^{\text{ocean}})(1 - g)\tilde{\tau}/2} \quad (4)$$

141 To capture observations best, two additional optimization steps were performed: 1) instead of a
142 fixed asymmetry parameter $g(\bar{R}_e)$ in the two-stream albedo (Eq. 4) we allowed variation with sun-
143 observer-geometry and found optimal $g(\bar{R}_e)$ through an approximation of $g(\bar{R}_e) = a + b \cdot \bar{R}_e + c \cdot \bar{R}_e^2$
144 and an exhaustive search of the a - b - c parameter space; 2) for least-square fitting (Eq. 2) and
145 $g(\bar{R}_e)$ optimization we only used horizontally homogeneous clouds (i.e. a $\nu = \left(\frac{\tilde{\tau}}{\sigma(\tilde{\tau})}\right)^2 > 10$) of
146 high MODIS retrieval quality (>80% of pixel within the cloud layer reported as well-retrieved) to
147 avoid sampling biases and maximize consistency of predicted radiances across angular bins. We
148 performed step 1 first for footprints of pure liquid and ice clouds so that we could use optimized
149 $g(\bar{R}_e)_{\text{liq}}$ and $g(\bar{R}_e)_{\text{ice}}$ for footprints of mixed phase. Tornow et al. (2020) provide more details on
150 this methodology and also explain that alternative approaches, such as separate sigmoidal fitting
151 over subsets of CERES-MODIS data defined by \bar{R}_e and $ACWV$ intervals, may be infeasible due to
152 partially sparse sampling of \bar{R}_e and $ACWV$.

153 3) USE OF RADIATIVE TRANSFER SIMULATIONS

154 Observations covered between 71-98% of sun-observer-geometries (for $\theta_0 > 20^\circ$, Table 3 in
155 Tornow et al. 2020). To produce complete hemispheric fields of upwelling TOA SW radiances
156 we used the plane-parallel radiative transfer code MOMO (Matrix Operator Model, Hollstein and
157 Fischer 2012) to simulate solar radiances. MOMO has foremost been applied to narrow-band

158 retrievals (e.g. Lindstrot et al. 2009; Diedrich et al. 2013) and was adapted to simulate the solar
 159 spectrum through 53 separate spectral intervals covering 0.25 – 4.0 μm , each using non-correlated
 160 k-binning (Bennartz and Fischer 2000) based on HITRAN-2008 (Rothman et al. 2009). Liquid-
 161 phase clouds were represented through sub-adiabatic prototypes (e.g. Brenguier et al. 2000) over
 162 up to 18 vertical layers and covered cloud optical depths between 0.3 and 27.1, cloud-top effective
 163 radii between 4.7 and 30.1 μm and above-cloud water vapor between 3.3 and 39.1 kg m^{-2} . Phase
 164 functions of liquid cloud droplets were calculated via Mie theory. For ice clouds, we used a single
 165 layer at higher altitude and produced optical depth ranges between 1 and 50, ice crystal effective
 166 radii between 20 and 50 μm , and above-cloud water vapor between 0.32 and 0.33 kg m^{-2} . Phase
 167 functions of ice crystals with a General Habit Mixture and severely roughened were extracted from
 168 the data base of Baum et al. (2011, 2014). To cover ranges in above-cloud water vapor, we used four
 169 different radiosonde vertical profile that represented moisture conditions over a 12 month-period
 170 well (see Fig. 2 in Tornow et al. 2018). In order to produce radiances for mixed phase clouds, we
 171 linearly combined liquid and ice cloud radiances.

172 The resulting radiances were interpolated onto a 2° angular grid. To produce consistent fields
 173 when joined with observation-based radiances \bar{I} (predicted from either model), simulated radiances
 174 I^{sim} were empirically adjusted. Adjustment factors were determined from hemispheric portions
 175 captured by observations (denoted as k and l for viewing zenith and relative azimuth angles,
 176 respectively) and, then, applied to all other angular portions (denoted as p and q , respectively).
 177 The empirical adjustments stem from Loeb et al. (2003) and was applied separately for currently
 178 operational and for newly developed, semi-physical ADMs.

$$\hat{I}(\theta_0^i, \theta_v^p, \varphi^q) = \frac{1}{mn} \sum_{k=1}^m \sum_{l=1}^n \bar{I}(\theta_0^i, \theta_v^k, \varphi^l) \frac{I^{\text{sim}}(\theta_0^i, \theta_v^p, \varphi^q)}{I^{\text{sim}}(\theta_0^i, \theta_v^k, \varphi^l)} \quad (5)$$

179 Lastly, adjusted simulated radiances \hat{I} served to as input to fit currently operational and newly
 180 developed, semi-physical models for angular bins without observations.

181 4) EXTRACTING ANISOTROPY

182 For each set of currently operational and newly developed, semi-physicals ADMs, we joined
 183 radiance-predicting models from observations and simulations. To ensure that hemispheric radi-
 184 ance fields were consistent and to avoid outliers from poor statistical or semi-physical observation-
 185 based fits, we screened for fits that had residuals with absolute biases lower than $1.5 \text{ W m}^{-2} \text{ sr}^{-1}$
 186 as well as standard deviations smaller than $10 \text{ W m}^{-2} \text{ sr}^{-1}$ (or $50 \text{ W m}^{-2} \text{ sr}^{-1}$ for sun-glint angles
 187 smaller 20°) and additionally set a minimum number of CERES-MODIS samples per angular bin
 188 (1000 for liquid and mixed-phase, 100 for ice phase). Bins with fewer observations or larger
 189 residuals relied entirely on simulated radiances.

190 Merged models produced radiances over half-hemispheres that were integrated to fluxes using
 191 an analytic solution of the integral discretized from $\int_0^{2\pi} \int_0^{\pi/2} I \cos \theta_v \sin \theta_v d\theta_v d\varphi$:

$$\bar{F}(\theta_0^i) = \sum_{k=1}^m \sum_{l=1}^n \bar{I}(\theta_0^i, \theta_v^k, \varphi^l) [\sin^2(\theta_v^k + 1) - \sin^2(\theta_v^k - 1)] \Delta\varphi \quad (6)$$

192 Finally, we extracted anisotropy \bar{R} as follows:

$$\bar{R}(\theta_0^i, \theta_v^k, \varphi^l) = \frac{\bar{I}(\theta_0^i, \theta_v^k, \varphi^l) \pi}{\bar{F}(\theta_0^i)} \quad (7)$$

193 In line with the current CERES methodology (pers. comm. Wenying Su) we performed a
 194 reference level correction (Loeb et al. 2002) that accounts for limb brightening as the intercept
 195 point is raised from the surface level to some altitude h (here $h = 20\text{km}$) and adjusted the anisotropy
 196 as follows (Loeb et al. 2003), where $r_e = 6371\text{km}$ is Earth's radius:

$$R(\theta_0^i, \theta_v^k, \varphi^l) = \bar{R}(\theta_0^i, \theta_v^k, \varphi^l) \left(\frac{r_e}{r_e + h} \right) \quad (8)$$

197 This step amplified all fluxes by 0.3% and did not impact the comparison of estimated fluxes from
 198 both ADMs.

199 Finally, this anisotropy was used to convert the instantaneously-observed radiance I into a flux
 200 \hat{F} :

$$\hat{F}(\theta_0) = \frac{I(\theta_0, \theta_v, \varphi) \cdot \pi}{R(\theta_0^i, \theta_v^k, \varphi^l)} \quad (9)$$

201 *b. Estimating Uncertainty*

202 To approximate the uncertainty behind each instantaneous flux estimate, we propagated the
 203 standard deviation of model fits $\Delta I = \sigma(dI)$, where dI are residuals from fitting of observations
 204 or simulations via currently operational or newly developed, semi-physical approach, into flux
 205 integrals ΔF and anisotropy estimates ΔR :

$$\Delta F(\theta_0^i) = \sqrt{\sum_{k=1}^m \sum_{l=1}^n \Delta I^2(\theta_0^i, \theta_v^k, \varphi^l) [\sin^2(\theta_v^k + 1) - \sin^2(\theta_v^k - 1)] \Delta \varphi} \quad (10)$$

206 as well as

$$\Delta R(\theta_0^i, \theta_v^k, \varphi^l) = R(\theta_0^i, \theta_v^k, \varphi^l) \sqrt{\left(\frac{\Delta I}{I}\right)^2 + \left(\frac{\Delta F}{\bar{F}}\right)^2} \quad (11)$$

207 The relative anisotropy uncertainty was then applied to instantaneous flux estimates:

$$\Delta \hat{F}(\theta_0) = \hat{F}(\theta_0) \frac{\Delta R(\theta_0^i, \theta_v^k, \varphi^l)}{R(\theta_0^i, \theta_v^k, \varphi^l)} \quad (12)$$

208 Uncertainties of daily and monthly flux averages $\Delta \bar{\bar{F}}$ were calculated as follows, where s is the
 209 number of samples:

$$\Delta \bar{\bar{F}} = \sqrt{\frac{1}{s} \sum_{d=1}^s \Delta \hat{F}_d^2} \quad (13)$$

210 Uncertainties from co-registration with auxiliary data, radiance unfiltering, or calibration were
 211 not incorporated. To determine whether fluxes from both ADMs and collected over diurnal and
 212 monthly time scales differed significantly we also used Student's t-test. To assess their mean
 213 difference we also calculated standard errors.

214 *c. Satellite Observations and collocated Parameters*

215 The current operational and newly developed, semi-physical ADMs were applied to inputs for the
216 month of January and July 2007 from CERES SSF Edition4A (Su et al. 2015) of the polar-orbiting
217 Aqua and Terra satellites (FM3 and FM1 instrument, respectively) and GERB Edition 1 (Dewitte
218 et al. 2008) from the geostationary Meteosat-8 (January) and Meteosat-9 (July) satellites. We
219 filtered footprints for water surfaces only in the region bound by 60°S - 30°N and 60°W - 25°E and
220 for solar geometries of $\theta_0 < 82^{\circ}$. As briefly introduced in Sec. 1, in this region there are often
221 low-level clouds of a wide range of cloud microphysical properties that arise from varying distances
222 to continents acting as a source of aerosol and a large span in above-cloud water vapor facilitated
223 by latitudinal slopes in precipitable water.

224 CERES SSF footprints cover approximately 20 km at nadir and solar channels measure the solar
225 radiance over the spectral range of 0.3-5.0 μm . During the time period the CERES instruments
226 operated in cross-track mode sampling Earth homogeneously (Barkstrom 1990). As part of the
227 SSF data set, statistics of MODIS-retrieved cloud properties (~ 1 km resolution) were produced
228 for up to two cloud layer per CERES footprint (given their cloud-top pressure differed by at least
229 50hPa). Statistics include cloud layer averages of effective radius \bar{R}_e , cloud-top pressure \bar{p}^{top} , and
230 visible cloud optical depth $\bar{\tau}$ as well as additional statistics on cloud optical depth including layer
231 standard deviation of optical depths $\sigma(\tau)$ and the exponential of the average over logarithmic values
232 of cloud optical depth $\bar{\tau}$, and clear and cloudy fractions, f_0 and $f_{1,2}$, respectively. Wind speed at 10
233 m provided with the SSF dataset were extracted from GEOS (Goddard Earth Observing System)
234 version 5.4.1 (Rienecker et al. 2008). In order to apply the novel ADMs, we extracted above-cloud
235 water vapor $ACWV$ for each CERES FOV by collocating vertical profiles of temperature $T(p)$
236 and relative humidity $rh(p)$ from ERA-20C (Poli et al. 2016) via nearest neighbor interpolation

237 accounting for MODIS-detected cloud-top height (of the layer with larger fraction):

$$ACWV = \frac{1}{g} \int_{p^{ctop}}^0 mr(p, T, rh) dp = \frac{1}{g} \int_{p^{ctop}}^0 \frac{e_s(T)}{p} \frac{mol_{h_2o}}{mol_{air}} rh(p) dp \quad (14)$$

238 with mixing ratio $mr(p, T, rh)$, saturation vapor pressure $e_s = 6.112e^{\frac{17.67T}{T+243.5}}$ (using T in degree
 239 Celsius, Bolton 1980), gravitational acceleration g , and molecular weights of water and dry air
 240 mol_{h_2o} and mol_{air} respectively. Using radiosonde data from St. Helena from 4 month in 2003, we
 241 determined that ERA20C-based ACWV values agreed to within $-1.06 \pm 4.35 \text{ kg m}^{-2}$. Currently
 242 operational CERES TOA SW fluxes were estimated using the sigmoidal approach (Sec. 2a1), as
 243 described in Su et al. (2015).

244 Data from GERB had a footprint size of approximately 10 km at nadir and the instrument's
 245 solar channel covered 0.32-4.0 μm . Although data was available every 15 minutes, we decided to
 246 process it hourly, with some timeslots missing at the time of acquisition (downloaded Feb. 2019
 247 from `ftp://oma.gerb.be`). Retrieved cloud properties from the multi-spectral imager SEVIRI
 248 (circa 3 km pixel size at subsatellite point) include cloud fraction f and visible cloud optical
 249 thickness $\tilde{\tau}$, as described above for CERES SSF. Cloud properties from SEVIRI were adjusted
 250 to match MODIS-retrieved properties to ensure coherent scene definition (Ipe et al. 2004). To
 251 apply newly developed, semi-physical ADMs, we extracted additional SEVIRI parameters from
 252 the Cloud Physical Property (CPP) data set version CLAAS 2.0 (Finkensieper et al. 2016; Benas
 253 et al. 2017): R_e , cloud phase, p^{ctop} and f at pixel-level. This allowed production of phase-specific
 254 layer average optical thicknesses and cloud fractions that we adjusted so that their sum would
 255 correspond to GERB values which were paired with layer-averaged effective radii and cloud-top
 256 pressure. As above, we used ERA-20C vertical profiles and SEVIRI cloud-top pressure to derive
 257 each FOV's ACWV and additionally extracted 10 m wind speed from ERA-20C. Applying the same
 258 ADMs to radiometers with slightly shifted spectral coverage inherently assumes proper radiance

259 unfiltering as well as negligible change in actual anisotropic behavior. Note that original GERB
260 processing used older ADMs (Loeb et al. 2003), and application of currently operational or newly
261 developed, semi-physical ADMs may cause deviations from official flux estimates that were not
262 investigated here.

263 To obtain complete diurnal cycles of TOA SW fluxes (as captured by e.g. Rutan et al.
264 2014; Gristey et al. 2018) and fill temporal and spatial gaps (due to $\theta_0 > 82^\circ$, missing
265 time slots, and faulty scan lines), we used the CERES SYN1deg-1hour data set (DOI:
266 10.5067/TERRA+AQUA/CERES/SYN1DEG-1HOUR_L3.004A) from January and July 2007 as a
267 complement. As described in Doelling et al. (2013, 2018), flux estimates in SYN1deg were
268 a composite based on measurements from multi-spectral imagers onboard geostationary satel-
269 lites and broadband radiometers from polar-orbiting Aqua and Terra satellites, and GERB
270 was used for validation in that study. Daily and monthly mean cloud properties were taken
271 from the CERES-SYN1deg-day (DOI: 10.5067/Terra+Aqua/CERES/SYN1degDAY_L3.004A)
272 and CERES-SYN1deg-month (DOI: 10.5067/Terra+Aqua/CERES/SYN1degMonth_L3.004A)
273 products.

274 3. Results

275 Angular distribution models provide crucial knowledge to transform a satellite-perceived TOA
276 SW radiance paired with underlying scene properties into an outgoing TOA SW flux. A newly
277 developed ADM based on semi-physical models (Sec. 2a2) expanded the list of properties defining
278 cloudy scenes over ocean and may, thus, produce significantly different flux estimates compared to
279 the currently operational methodology, introduced as sigmoidal model (Sec. 2a1). In order to verify
280 significance and understand differences, we started by analyzing the impact onto instantaneous flux
281 estimates (Sec. 3a) and, then, examined spatial and temporal flux averages (Sec. 3b).

282 *a. Impact on Instantaneous Flux Estimates*

283 To investigate whether instantaneous flux estimates significantly change when switching from
284 currently operational to newly developed, semi-physical models, we predicted radiance fields from
285 each model and derived anisotropies for three cases of liquid-phase clouds over ocean (Fig. 1). Each
286 case (separated by panel) was further stratified by varying a newly incorporated property which
287 the currently operational approach was insensitive to: cloud-top effective radius \bar{R}_e (Fig. 1, panels
288 I and II) and above-cloud water vapor $ACWV$ (Fig. 1, panel III). Values were chosen to roughly
289 represent the outer margins (red and blue) and most frequent values (green) of observed spectra
290 (exemplary histograms shown in lower panels of Fig. 2). Anisotropy fields were characterized by
291 cloud scattering features, such as cloud bow and cloud glory, as well as sun-glint (for a broken
292 cloud field in Fig. 1, panel II). Anisotropy deviations between ADMs were generally within the
293 uncertainty range, except for angular portions in the backscattering direction (Fig. 1, panel I).

294 [Figure 1]

295 For varying \bar{R}_e (Fig. 1, panels I and II), anisotropies exhibit systematic differences in the
296 backscattering region, such that larger droplet sizes caused reduced anisotropies (θ_v between -60°
297 and 0°), a sharper response around the cloud glory (θ_v between -31° and -35°), and a cloud bow
298 shifted further away from the cloud glory (θ_v between -63° and $+7^\circ$). These systematic deviations
299 in the backscattering region were consistent with Mie-calculated single-scattering intensities (cf.
300 Fig. 1 in Tornow et al. 2018). The anisotropy response to $ACWV$ affected predominantly the
301 forward-scattering direction and will be discussed in the Sec. 4.

302 [Figure 2]

303 To understand for which scene properties both ADMs agree, we selected three viewing geometries
304 for solar illumination $\theta_0 \in [32, 34^\circ]$ and two cloud optical thicknesses and varied \bar{R}_e and $ACWV$.

305 Fig. 2 illustrates where currently operational ADMs (shown as circle) that were insensitive to these
 306 variations intersected newly developed, semi-physical ADMs (shown as triangles). For \bar{R}_e (left
 307 panel of Fig. 2), the intercepts were found mostly between 10 and 15 μm and correspond to most
 308 prevalent values of CERES-MODIS data (shown in histograms in the bottom left panel) that were
 309 used to train both sets of ADMs. As shown across three $\tilde{\tau}$ groups, most frequent \bar{R}_e can marginally
 310 shift. And as analyzed during ADM development, \bar{R}_e median values across angular bins of the
 311 same upward hemisphere can vary substantially (between 7 and 17 μm for selected θ_0 , shown in
 312 Table 3 of Tornow et al. 2020). For ranging $ACWV$ (right panel of Fig. 2) the anisotropy response
 313 was lower and we found fewer matches between ADMs. Missing matches for ranging $ACWV$
 314 could be caused by picking a single \bar{R}_e in this analysis that – as we listed above – could deviate
 315 sufficiently from median \bar{R}_e in individual angular bins to which currently operational ADMs may
 316 have been biased. For selected scenes and viewing geometries, the shading highlights 5 W m^{-2}
 317 flux deviation that could be facilitated by deviations of $\pm 5 \mu\text{m}$ or $\pm 15 \text{ kg m}^{-2}$ from \bar{R}_e and $ACWV$
 318 intercepts, respectively. Note that – because anisotropy integrates by definition to 1 over the upward
 319 hemisphere – negative anisotropy differences in one viewing direction must be balanced by positive
 320 differences in another direction, and vice versa. Consequently, for the same scene but different
 321 viewing geometries, two ADMs can produce flux differences of opposite sign across geometries.
 322 This can be seen from different slopes in forward and backscattering direction, for example.

323 [Figure 3]

324 To more accurately quantify the impact on resulting flux estimates, we predicted radiance fields
 325 \bar{I} using newly developed, semi-physical models (sensitive to \bar{R}_e and $ACWV$) applied anisotropies
 326 from both semi-physical and currently operational models, and measured their difference:

$$\delta \hat{F} = \hat{F}_{\text{loglinear}} - \hat{F}_{\text{sigmoidal}} = \frac{\bar{I}\pi}{R_{\text{loglinear}}} - \frac{\bar{I}\pi}{R_{\text{sigmoidal}}} \quad (15)$$

327 Fig. 3 presents the expected flux difference $\delta\hat{F}$ for two settings of solar geometry and cloud
 328 optical depth (by row) as well as cloud micro-physical setup (by column). Given the observer's
 329 perspective (defined by θ_v and φ), magnitude and sign of $\delta\hat{F}$ vary considerably. For either small
 330 or large droplets sizes (left or right panels of Fig. 3), cloud bow features stand sharply out from
 331 cloud glory and were of opposite sign but comparable magnitude. Nadir and forward scattering
 332 regions were equally intense and of alternating sign but showed more gradual changes in $\delta\hat{F}$ across
 333 viewing geometries. An effective radius of 10 μm had much reduced $\delta\hat{F}$ – possibly the result
 334 of median conditions being reflected in currently operational models, as demonstrated above –
 335 although some angular portions showed persistent differences (e.g. nadir directions of $\tau = 5.5$
 336 and $\theta_0 = [40^\circ, 42^\circ]$ had positive differences regardless of \bar{R}_e). Flux differences for various ACWV
 337 scenarios (not shown) marked a pronounced response in the forward scattering direction (with
 338 positive flux differences for high ACWV values) and a less intense response of opposite sign in the
 339 backscattering portion.

340 [Figure 4]

341 To quantify an expected upper bound of instantaneous flux changes due to a switch in ADMs and
 342 to exclude systematic differences, we took hemispherically resolved flux differences of extreme
 343 scenarios (e.g. the left and right column of Fig. 3 for \bar{R}_e), measured the span in $\delta\hat{F}$ per angular
 344 portion across extremes, and from absolute values divided by 2 (assuming that median conditions
 345 of zero difference were roughly centered between extremes) we extracted the 95th percentile:

$$\delta\hat{F}_{\text{max}} = 95^{\text{th}} \text{percentile of } \frac{|\delta\hat{F}_{20\mu\text{m}} - \delta\hat{F}_{5\mu\text{m}}|}{2} \quad (16)$$

346 We repeated quantification for a range of cloud optical depths and solar geometries. Fig. 4
 347 shows how the upper bound was generally within 25 W m^{-2} . For extremes in \bar{R}_e , expected flux

348 differences increased with τ up to 10-20 then decreased for $\tau > 20$. This was likely a result of both
349 an increasing cloud albedo and the diminished fraction of single-scattering versus multi-scattering
350 events (thus reducing the dominance of cloud bow and glory features) that are both associated with
351 larger τ . Maxima (at τ of 10-20) grew larger in $\delta\hat{F}_{\max}$ for lower θ_0 , as more downwelling radiation
352 reached the cloud for reflection. Accounting for increased solar influx for lower solar zenith angles,
353 resulting peak albedo changes were roughly comparable and amounted to 0.015-0.020. Maxima
354 shifted towards larger τ for lower θ_0 ; likely the result of more single-scattering features (i.e. cloud
355 bow and cloud glory) positioned closer to the nadir and rather influencing the intensity fields for
356 higher τ values. For extremes in *ACWV*, we found a rising impact with larger τ and lower θ_0 . We
357 conclude that increased water vapor absorption effects, associated with *ACWV*, occurred for more
358 available solar radiation (lower θ_0) and higher albedos (larger τ).

359 [Figure 5]

360 *b. Impact on Daily and Monthly Averages*

361 Figure 3 indicated that regions of persistently low or high \bar{R}_e could repeatedly experience flux
362 differences of similar magnitude and sign when observed under similar viewing geometry over
363 the course of a day (i.e. varying θ_0 and possibly varying τ , but comparable φ and θ_v , e.g.
364 by geostationary platforms) or over the course of a month (e.g. by polar-orbiting satellites).
365 This suggests that temporal and spatial averages of such regions could result in systematic flux
366 differences. To investigate whether flux estimates from newly developed, semi-physical ADMs and
367 their difference to currently operational estimates affect daily and monthly averages, we processed
368 two months (January and July 2007) of CERES-MODIS and GERB-SEVIRI (Sec. 2c) data over
369 the tropical and southern Atlantic (all water surfaces between 60°W and 25°E as well as 60°S and
370 30°N). Processing involved application of currently operational ADMs (thus reproducing values

371 in CERES-MODIS and upgrading GERB-SEVIRI data, see Sec. 2c) and of newly developed,
372 semi-physical ADMs. Following Doelling et al. (2013), we first averaged fields onto an equal-area
373 grid. An example is shown in Fig. 5 for orbits of Aqua and Terra as well as a time slot of MSG.
374 Flux estimates across missions resulted in roughly similar flux difference pattern in magnitude
375 and sign and could be associated with \bar{R}_e (e.g. the around 12°W and 25°S). Each pixel was then
376 adjusted so GERB fluxes matched with coincident CERES-based estimates. This step assumed an
377 absolute calibration differences between both instruments (Doelling et al. 2013). Figure 6 shows
378 examples of pixels before the GERB-to-CERES adjustment. To fill the diurnal cycle beyond a
379 solar zenith angle (SZA) of 82° and to fill missing time slots or faulty scan lines, we used CERES-
380 SYN1deg-1hour data (Sec. 2c). Figure 6 shows the overall agreement between CERES-MODIS,
381 GERB-SEVIRI, and CERES-SYN1deg-1hour data sets.

382 [Figure 6]

383 Resulting daily averages (Fig. 7) show significant flux differences between the equator and 40°S
384 with a magnitude of 10 W m⁻². Regions of extreme differences line up with extremes in \bar{R}_e . For
385 other days, differences were less pronounced or canceled out (not shown). As seen in Figure 6, daily
386 mean differences can be traced back to GERB-based flux differences on the hourly time scale (e.g.
387 for "Tropical Atlantic" between 1200-1400 UTC, or "Offshore" during most time slots). These
388 were then amplified by CERES-based flux differences (e.g. for "Offshore" and "Tropical Atlantic")
389 through GERB-to-CERES adjustment (see above). The region around 40°W, 20°N presents an
390 example of similar cloud properties and yet flux differences of opposite sign that resulted from
391 slightly different viewing geometries.

392 [Figure 7]

393 Finally, we computed monthly mean flux differences. Fewer areas show significant differences
394 (Figs. 8 and 9). However, there are regions with significant difference in fluxes, up to 5 W m^{-2} .
395 These regions are less coherent with extremes in \bar{R}_e and $ACWV$. We presume that ranging sun-
396 observer-geometries over the course of a months paired with ranging levels of \bar{R}_e and $ACWV$ led
397 to non-significant or vanishing flux differences.

398 [Figure 8 & 9]

399 For two selected pixels (marked in Fig. 7, 8, and 9) that showed significant differences in July, we
400 plotted daily average scene properties and resulting flux differences (Fig. 10). “Coastal” (shown
401 in red) was characterized by persistently low \bar{R}_e and ranging $ACWV$, producing varying flux
402 differences that were on average negative. “Offshore” (shown in blue), on the other hand, showed
403 persistently low $ACWV$ and varying \bar{R}_e that mostly resulted in positive differences and, thus,
404 had a positive average difference. Monthly averages of strongly varying \bar{R}_e and $ACWV$ (shown
405 as triangles) resulted in non-extreme levels, while flux differences failed to cancel out towards
406 vanishing monthly means.

407 [Figure 10]

408 In summary, we demonstrated that the choice of ADM matters for flux estimation in particular
409 when encountering extremes in \bar{R}_e and $ACWV$ and that can lead to instantaneous flux differences of
410 up to 25 W m^{-2} . Instantaneous fluxes differed significantly when captured from the backscattering
411 direction. Flux differences originating at the instantaneous level can lead to systematic differences
412 in daily and monthly means on the order of 10 W m^{-2} and 5 W m^{-2} , respectively. Regions
413 associated with significant differences were mainly closer to the continent or further offshore,
414 where effective radii showed particularly small ($\bar{R}_e < 7\mu\text{m}$) or large ($\bar{R}_e > 15\mu\text{m}$) values, respectively.

4. Discussion

Angular distribution models allow to convert radiance observations from satellite platforms such as Aqua, Terra, or Meteosat 8 and 9 into outgoing radiative fluxes at TOA. A newly developed set of ADMs using semi-physical models (Tornow et al. 2020) offers a novel way to estimate solar fluxes over marine clouds. For liquid-phase clouds, we show here that instantaneous flux estimates from newly developed, semi-physical models may deviate up to 25 W m^{-2} from currently operational models employed in CERES SSF Edition 4A; given particularly small or large effective radii or loads of absorbing water vapor above clouds. We further demonstrate that these flux differences may occur systematically and affect daily and monthly means causing significant differences of the order of 10 W m^{-2} and 5 W m^{-2} , respectively.

Updated solar fluxes in regions of low-level clouds may improve our understanding of aerosol indirect effects and resulting radiative properties. Recent studies using instantaneous CERES SSF Edition 4 satellite observations to quantify the change in cloud radiative effects due to cloud-aerosol interaction (e.g. Painemal 2018; Gryspeerdt et al. 2019) may present less intense slopes using updated values of this study that indicated higher TOA SW fluxes for cloud of larger \overline{R}_e and vice versa. The upcoming satellite mission EarthCARE (Illingworth et al. 2015) may also benefit from improved flux estimation. The mission is dedicated to assessing our current understanding of cloud-aerosol-radiation interaction by comparing observation-based TOA fluxes with radiative transfer calculations acting on cloud and aerosol retrievals from active-passive instruments. Scenes exhibiting solar or terrestrial flux differences beyond 10 W m^{-2} will be considered poorly understood. Mitigating hypothetical differences coming from the observation-based end (e.g. due to micro-physical extremes as seen in this study) will help narrowing the focus on scenes that deserve attention. Using ADMs that fail to produce adequate anisotropy responses to

438 \overline{R}_e or $ACWV$, on the other hand, could cause $> 10 \text{ W m}^{-2}$ deviations over relatively straightforward
439 Earth-atmosphere scenes and may divert valuable resources by searching for possible insufficiencies
440 in other parts of the closure assessment. Furthermore, data sets like CERES EBAF (Loeb et al.
441 2018) that rely on monthly means, as produced here, are crucial to evaluate the performance of
442 climate models (e.g. Li et al. 2013; Ahlgrimm et al. 2018).

443 The physically plausible response of TOA SW anisotropy to \overline{R}_e is evidence in favor for newly
444 estimated fluxes being realistic. However, few instantaneous estimates could be identified as
445 significantly different from currently operational estimates as error bars overlapped. The basis
446 for uncertainty estimates were radiance residuals after fitting currently operational and newly
447 developed, semi-physical models trying to capture all CERES-MODIS observations over a 5-year
448 period. A case study using homogeneous samples of specific optical depth from this period dealt
449 with much reduced radiance fluctuations and exhibited significant anisotropy differences (Tornow
450 et al. 2018). We believe that the errors we estimate in this study (Sec. 2b) may have been too
451 conservative and produced too large uncertainties for clouds of lower optical depth.

452 Daily mean differences between ADMs exceed other sources of uncertainty (Loeb et al. 2009),
453 such as instrument calibration (2 W m^{-2}) or imperfect knowledge of solar irradiance (1 W m^{-2}).
454 Additional uncertainty may originate from inaccurate retrievals of $ACWV$ (caused by cloud-top
455 height retrieval or water vapor profiles) as well as \overline{R}_e . Systematic retrieval errors could be harmless
456 if samples resemble the "training data" (here 5 years of CERES SSF observations) and if all
457 viewing geometries for a selected illumination geometry are affected likewise. On the other hand,
458 systematic errors could pose problems when predicting fluxes for other platforms (such as MSG
459 satellites in this study). Future efforts need to ensure that SEVIRI retrieval of \overline{R}_e and cloud-
460 top height are consistent with CERES SSF data. Similarly, when switching to other sources for

461 coincident profiles of atmospheric water vapor, cross-checks should be performed between the new
462 source and the source used for training.

463 The response of TOA SW anisotropy to *ACWV* was different than expected. Instead of a θ_v -
464 dependency (expecting absorption to increase as the atmospheric mass grows with θ_v), we found
465 dominant anisotropy sensitivity in the forward scattering direction (Fig. 1C), an angular portion
466 associated with multi-scattering. We suspect that the observed response may have captured in-cloud
467 absorption of multi-scattered sunlight and that could have served as proxy of in-cloud water vapor.
468 These effects in the forward scattering direction may have clouded a θ_v -dependency that we found
469 in simulated radiances (not shown). In this study, we found significant flux differences in regions
470 that had low *ACWV* values and little variation, and, therefore, can assume that we avoided potential
471 anisotropy artifact. Future efforts should try to separate in-cloud from above-cloud absorption by
472 e.g. introducing a single scattering albedo into the two-stream cloud albedo of newly developed,
473 semi-physical models. This single scattering albedo ω could be a function of both \bar{R}_e and in-cloud
474 water vapor *ICWV*, $\omega = \omega(\bar{R}_e, ICWV)$.

475 With the advent of operational products reporting cloud-topped aerosol optical thickness (e.g.
476 Peers et al. 2019; Sayer et al. 2016), the present semi-physical model could be readily expanded to
477 incorporate absorption effects due to aerosol. Regions such as the southeast Atlantic experiencing
478 seasonal plumes of biomass burning aerosol could be particularly benefiting from further refined
479 models.

480 Finally, this study focused on liquid-phase clouds. We expect less flux differences for ice clouds
481 as their *ACWV* levels are much reduced and scattering effects on ice crystals (and their rich natural
482 variety in shape treated as bulk) shows less marked changes with \bar{R}_e (cf. Tornow et al. 2020).
483 We do, however, expect differences for mixed-phase footprints (i.e. footprints which usually
484 contain an ice cloud next to a liquid cloud) as their representation through newly developed, semi-

485 physical models is more refined. Future work will carefully assess flux differences to the currently
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498 *Data availability statement.* Data analyzed in this study were a re-analysis of existing data, which
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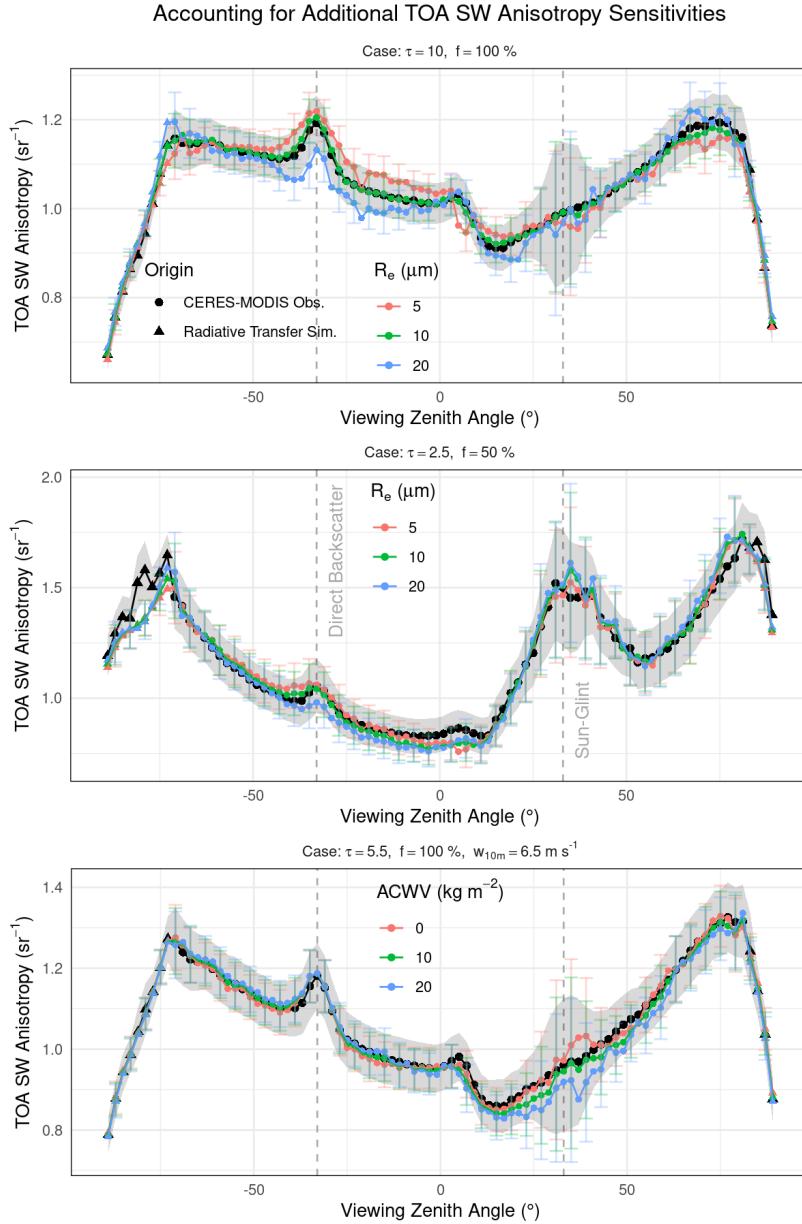
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698 **LIST OF FIGURES**

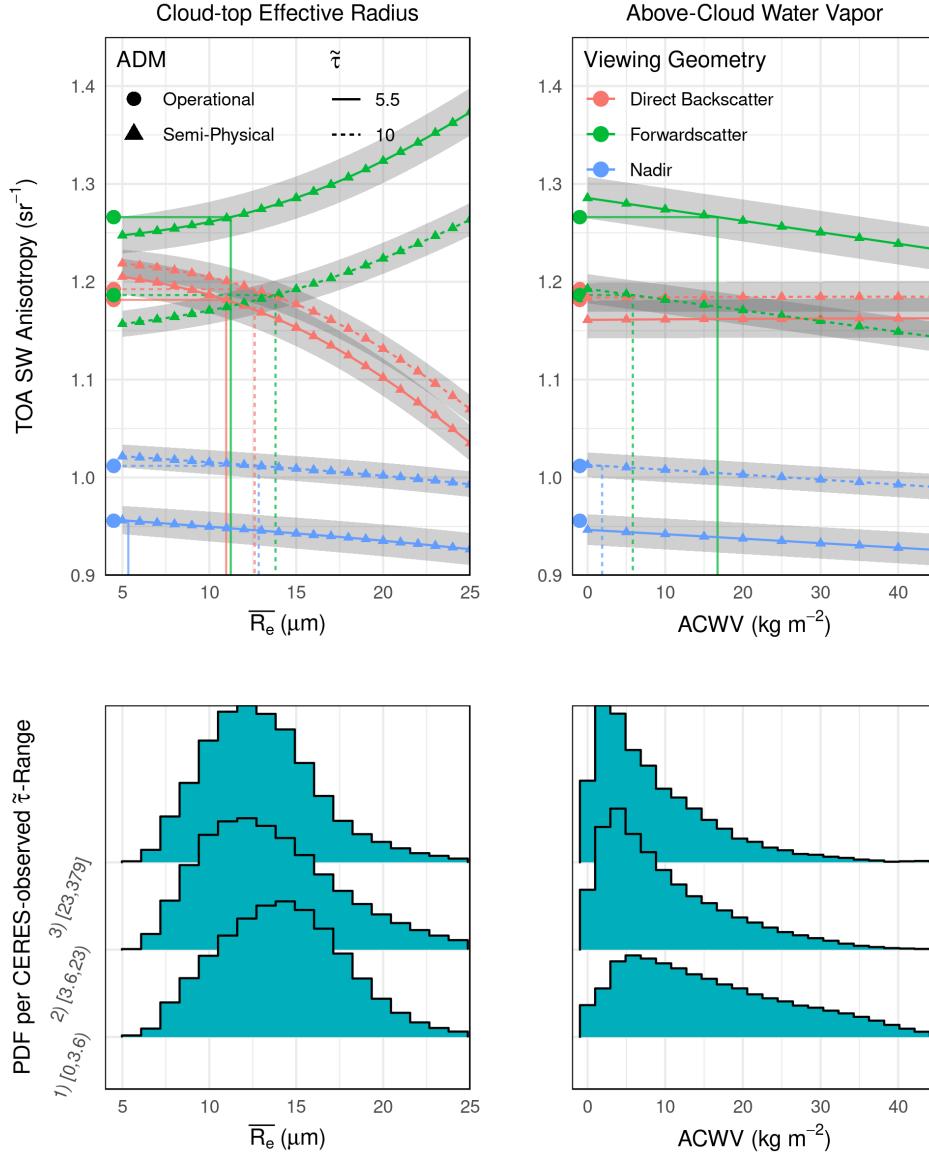
- 699 **Fig. 1.** Along the principal plane of solar geometry $\theta_0 \in [32, 34^\circ]$, we present the response of TOA
700 SW anisotropy to newly introduced parameters of semi-physical ADMs (shown in colors) and
701 comparison to currently operational ADMs (shown in black) that were insensitive to these
702 parameters. Negative x-values correspond to the backscattering direction ($\varphi \in [178, 180^\circ]$),
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751	$\bar{\tau}$, \bar{R}_e , and $ACWV$ (from top to bottom). Triangles on the right mark monthly mean and	
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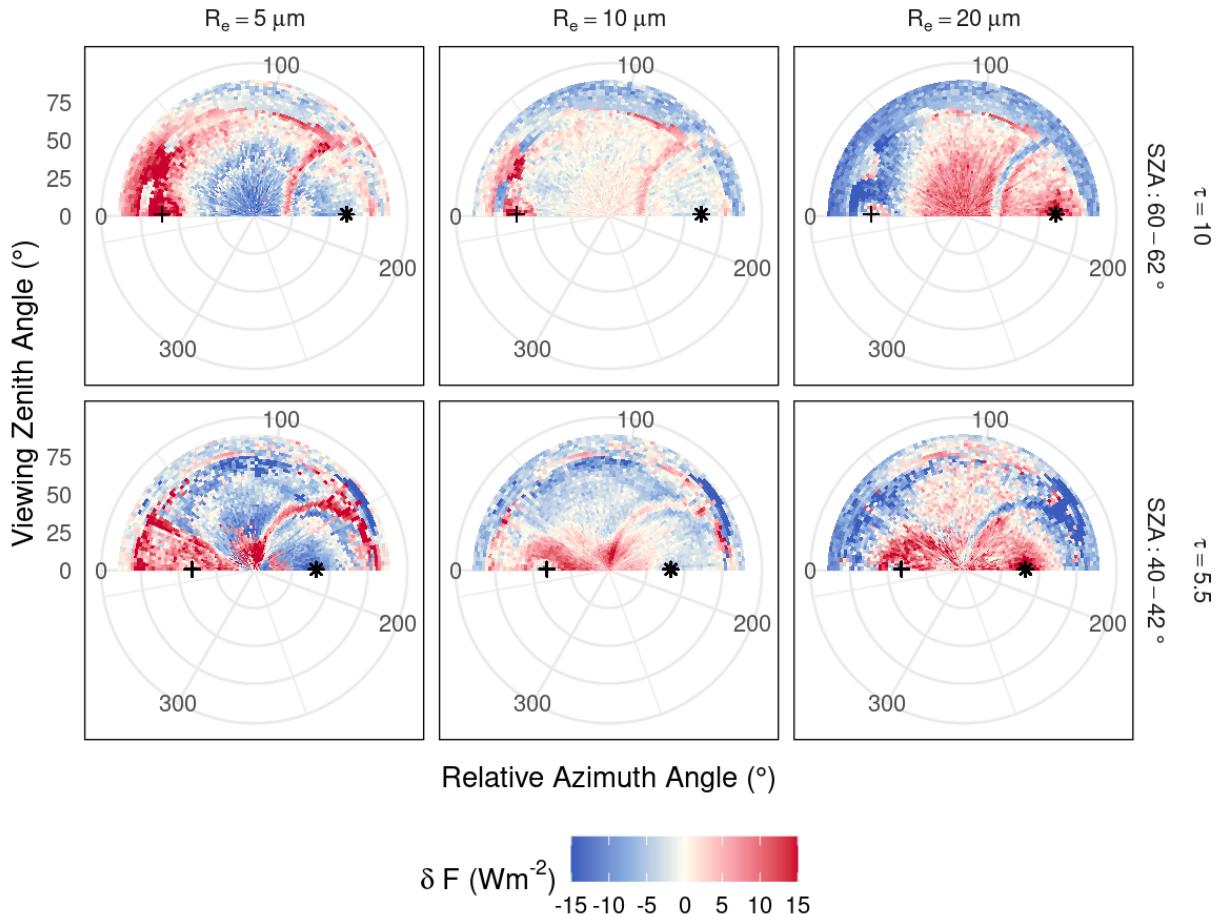
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Correspondence between Currently Operational and Semi-Physical ADMs



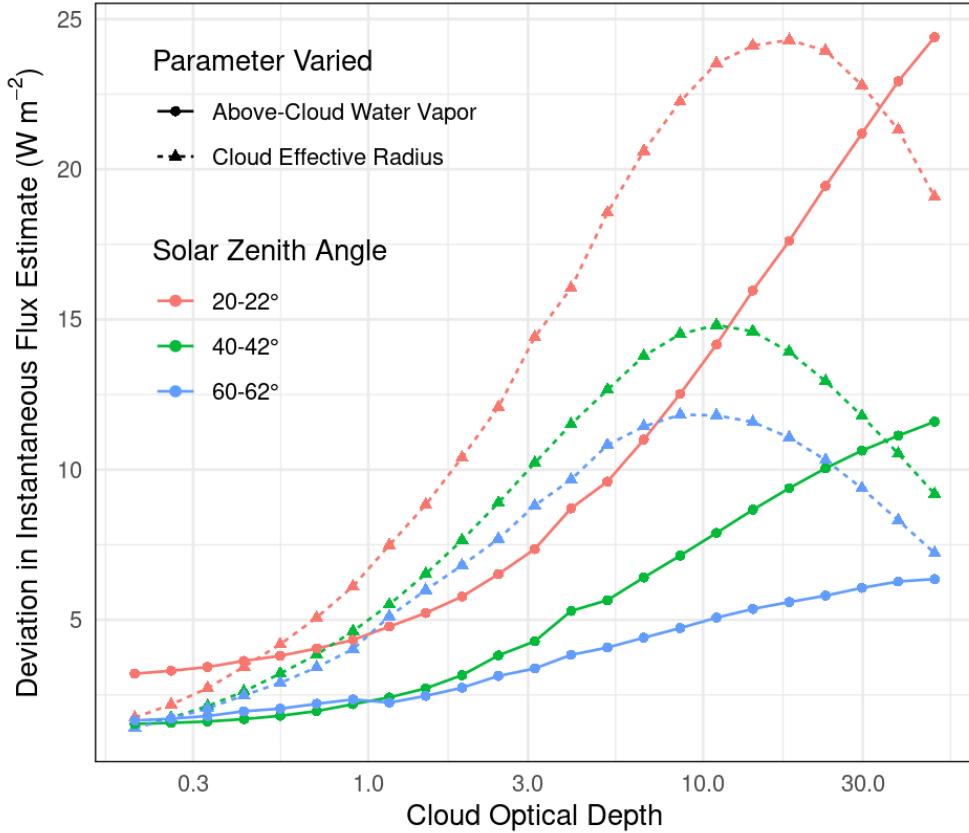
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 765 and a constant solar geometry of $\theta_0 \in [32, 34^\circ]$, we highlight for which \bar{R}_e and ACWV both ADMs agree. Bottom
 766 plots show histograms of respective variable over three τ -ranges and help understanding potential sampling biases
 767 in currently operational ADMs.

Expected Deviation in Flux Estimation when using Semi-Physical instead of Currently Operational ADMs

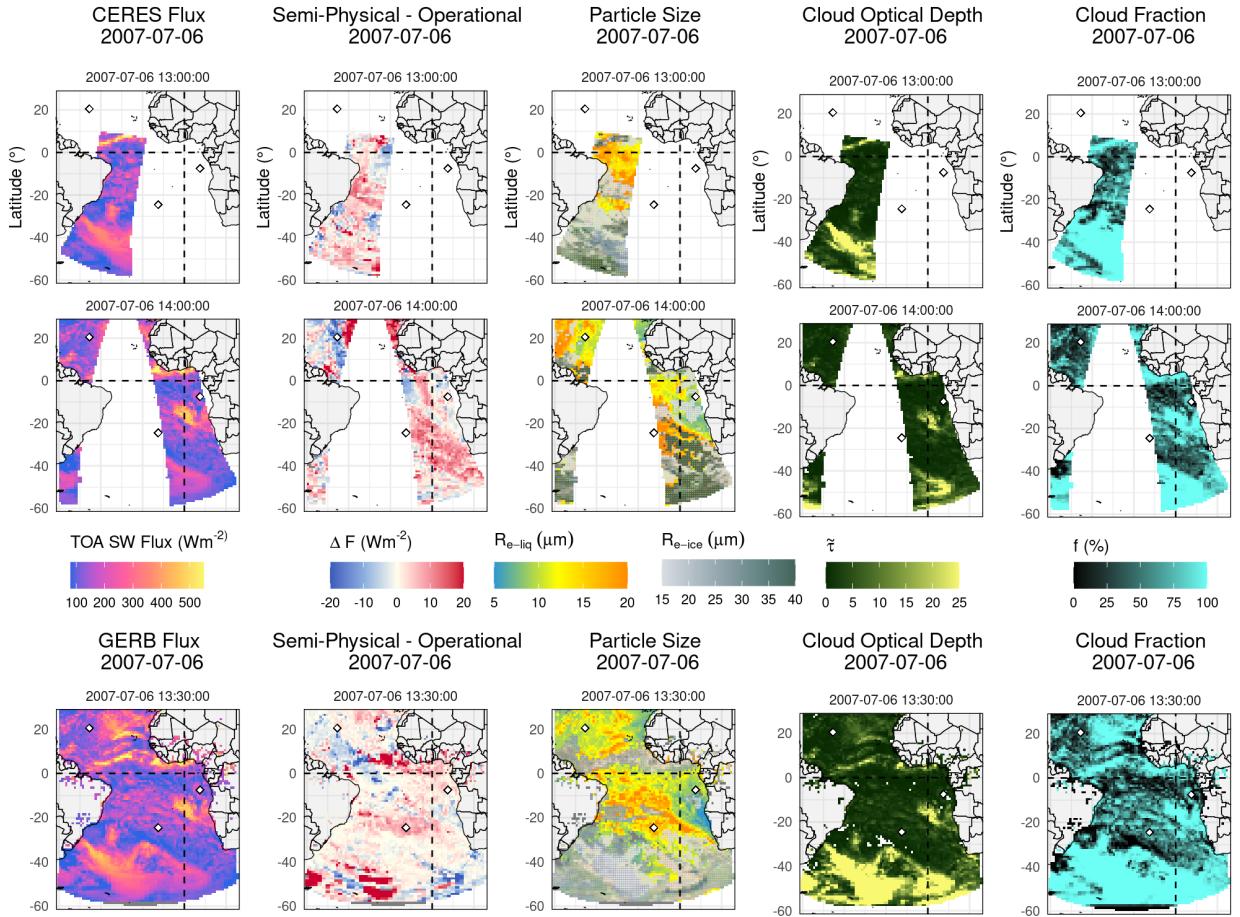


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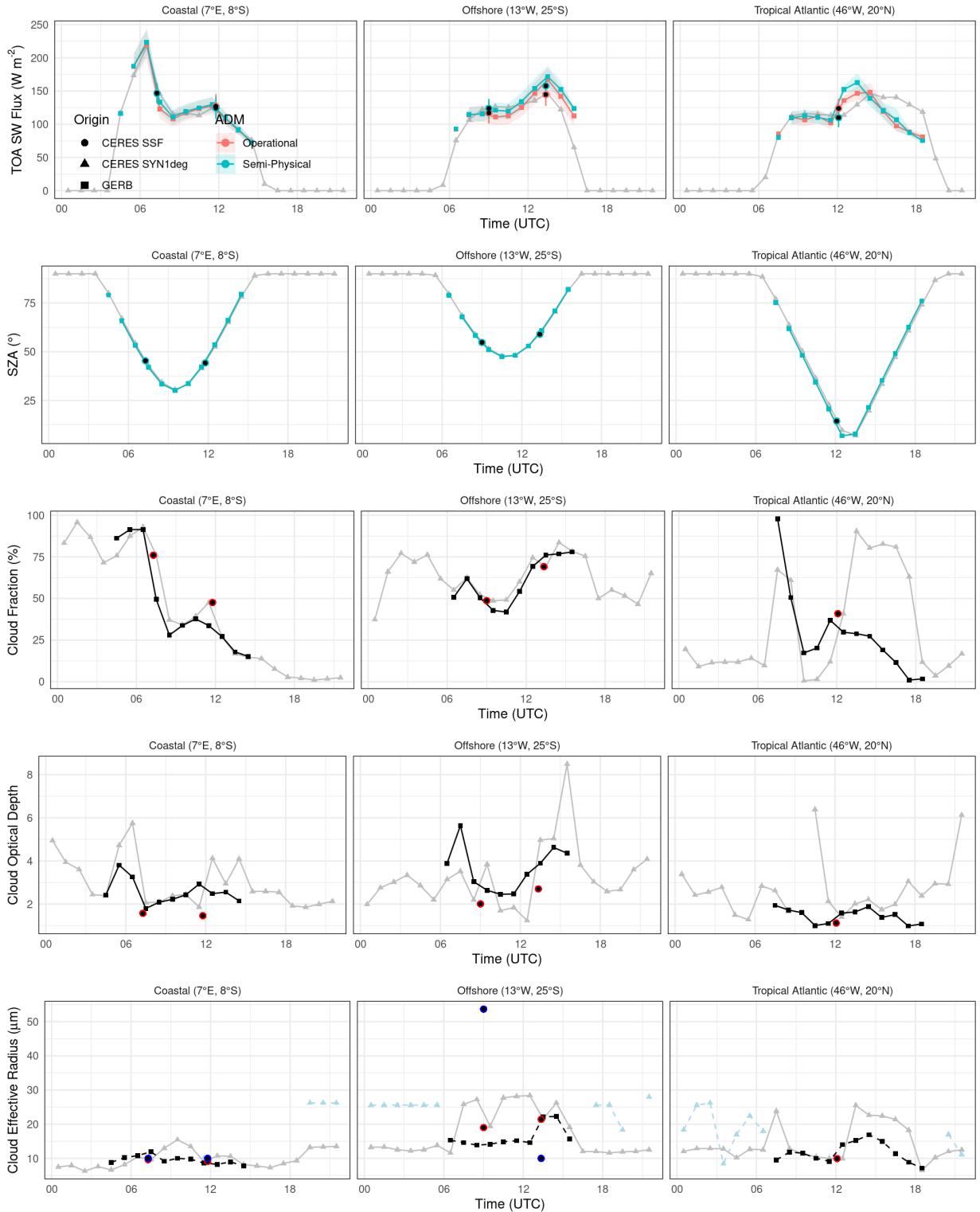
Magnitude of Changing Fluxes under Semi-Physical ADMs
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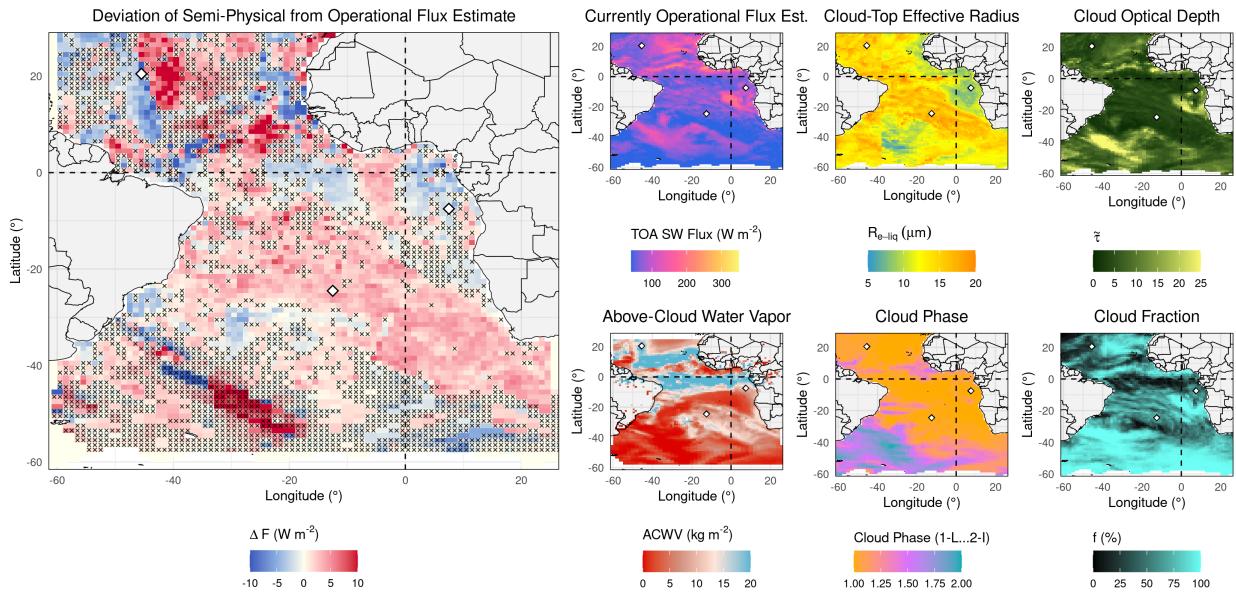
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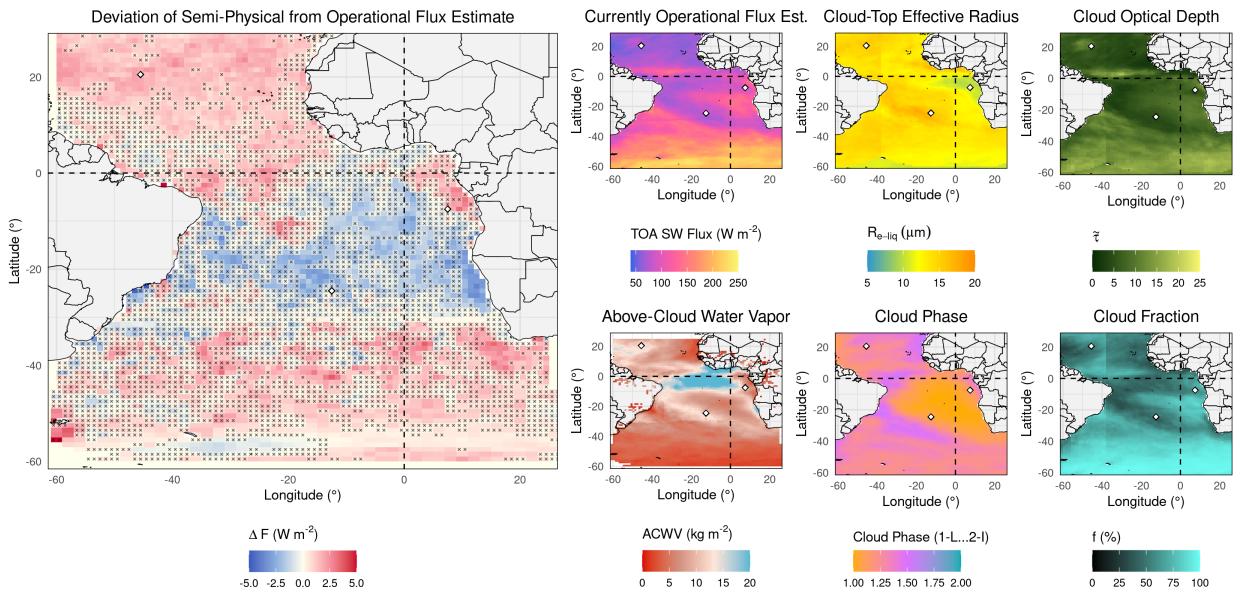
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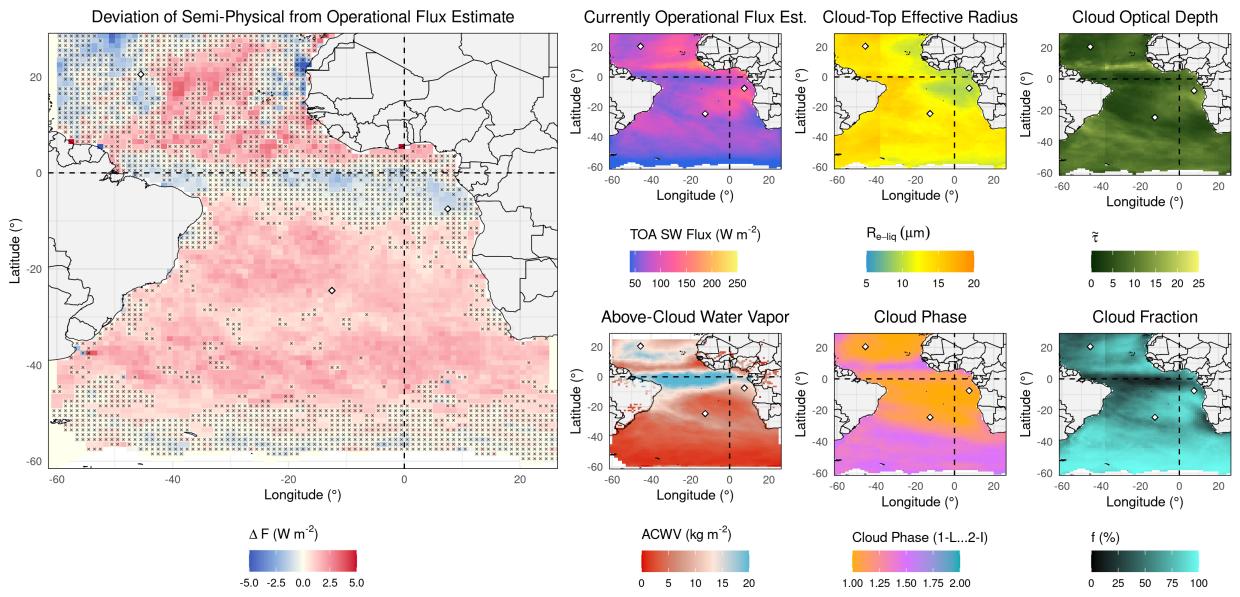
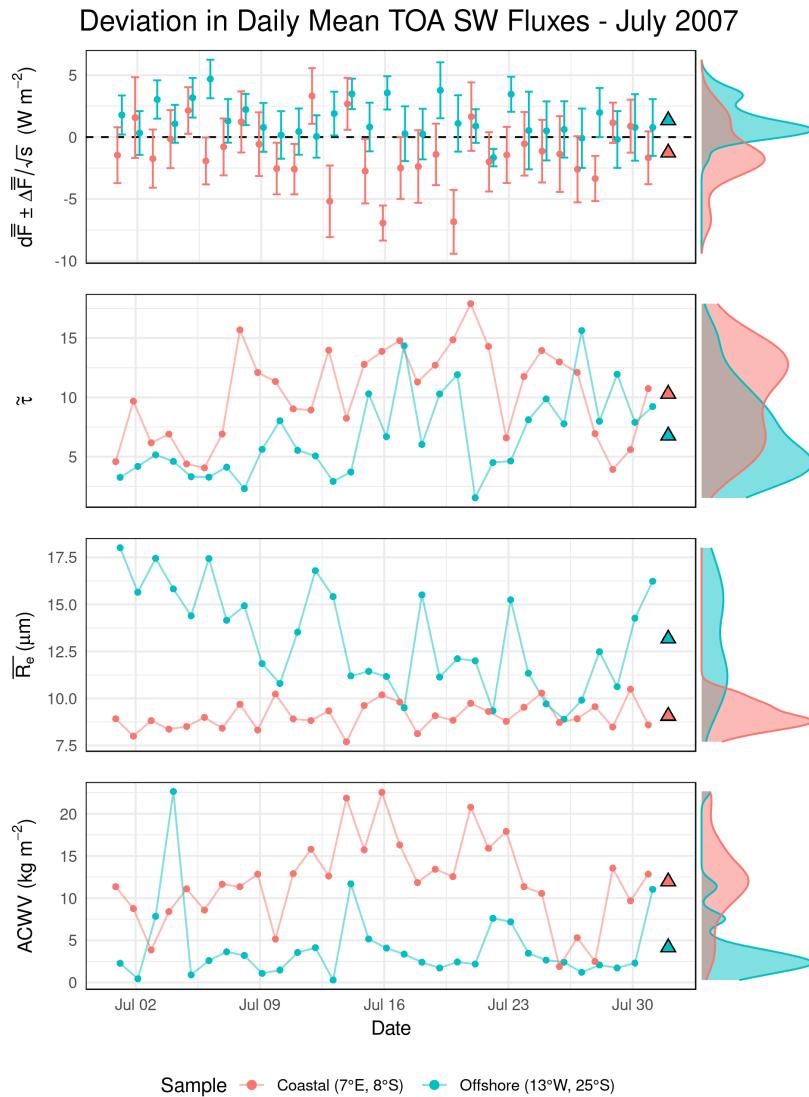


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795 FIG. 10. Daily averages during the month of July 2007 for two selected pixels (separated by color). We show
 796 differences in daily mean fluxes (errorbars show the standard error), and daily mean $\bar{\tau}$, \bar{R}_c , and ACWV (from top
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