

Understanding top-of-atmosphere flux bias in the AeroCom Phase III models: a clear-sky perspective

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Key Points:

- Model biases in clear-sky top-of-atmosphere reflected shortwave fluxes are linked to biases in aerosol optical depth and surface albedo
- Over ocean, aerosol optical depth bias contributes about 25% to the 60°S-60°N mean SW flux bias for the multi-model mean (MMM) result
- Over land, aerosol optical depth and land surface albedo biases contribute about 40% and 30%, respectively, to the 60°S-60°N mean SW flux bias for the MMM result

Abstract

Biases in aerosol optical depths (AOD) and land surface albedos in the AeroCom models are manifested in the top-of-atmosphere (TOA) clear-sky reflected shortwave (SW) fluxes. Biases in the SW fluxes from AeroCom models are quantitatively related to biases in AOD and land surface albedo by using their radiative kernels. Over ocean, AOD contributes about 25% to the 60°S-60°N mean SW flux bias for the multi-model mean (MMM) result. Over land, AOD and land surface albedo contribute about 40% and 30%, respectively, to the 60°S-60°N mean SW flux bias for the MMM result. Furthermore, the spatial patterns of the SW flux biases derived from the radiative kernels are very similar to those between models and CERES observation, with the correlation coefficient of 0.6 over ocean and 0.76 over land for MMM using data of 2010. Satellite data used in this evaluation are derived independently from each other, consistencies in their bias patterns when compared with model simulations suggest that these patterns are robust. This highlights the importance of evaluating related variables in a synergistic manner to provide an unambiguous assessment of the models, as results from single parameter assessments are often confounded by measurement uncertainty. We also compare the AOD trend from three models with the observation-based counterpart. These models reproduce all notable trends in AOD (i.e. decreasing trend over eastern United States and increasing trend over India) except the decreasing trend over eastern China and the adjacent oceanic regions due to limitations in the emission dataset.

Plain Language Summary

Aerosol optical depths (AOD) from satellite retrievals have been used to evaluate the AeroCom models. However, these evaluations are often non-conclusive due to uncertainties in the retrievals and the differences among many products. In this study, biases in top-of-atmosphere reflected shortwave fluxes are linked to biases in aerosol optical depth and surface albedo by using their respective radiative kernels. Over ocean, AOD contributes about 25% to the 60°S-60°N mean SW flux bias for the multi-model mean (MMM) result. Over land, AOD and land surface albedo contribute about 40% and 30%, respectively, to the 60°S-60°N mean SW flux bias for the MMM result. The spatial patterns of the SW flux biases derived from the radiative kernels are very similar to the model flux bias relative to satellite observations, with the correlation coefficient of 0.6 over ocean and 0.76 over land for MMM using data of 2010. This study highlights the importance of evaluating related variables in a synergistic manner to provide an unambiguous assessment of the models, as results from single parameter assessments are often confounded by measurement uncertainty.

1 Introduction

Atmospheric aerosols play important roles in the climate systems directly by scattering and absorbing the solar and terrestrial radiation, and indirectly by modifying the cloud properties. The direct aerosol radiative effect (DARE) and direct aerosol radiative forcing (DARF) are often used to quantify aerosols' impact on climate. DARE is defined as the mean radiative flux perturbation due to the presence of aerosols (both natural and anthropogenic), while DARF is the anthropogenic component of DARE. Many studies used satellite measurements to estimate the DARE (e.g., Loeb & Manalo-Smith, 2005; Remer & Kaufman, 2006; Yu et al., 2006; Su et al., 2013). However, determining DARF from satellite measurements is more challenging because current satellite sensors cannot discriminate anthropogenic aerosols from natural aerosols. Our current understanding of DARF relies mostly on the AeroCom model simulations (Schulz et al., 2006; Myhre et al., 2013), with a few studies estimating observational constrained DARF (Bellouin et al., 2005, 2008; Su et al., 2013; Paulot et al., 2018).

76 As the AeroCom models are vital in advancing our understanding on how aerosols
 77 are affecting the climate and its future projection, many aspects of the models have been
 78 examined under prescribed conditions. Stier et al. (2013) assessed the host model un-
 79 certainties on DArF by prescribing identical aerosol radiative properties in all models
 80 and found significant model diversity in simulated clear- and all-sky DArF. One of the
 81 variables that contributed to this diversity was surface albedo which had a global mean
 82 inter-model relative standard deviation of 4% and significantly larger variability on re-
 83 gional scale. Randles et al. (2013) examined the performance of radiative transfer schemes
 84 used in the models and found that diversity among models in the top-of-atmosphere (TOA)
 85 DArF depended on the solar zenith angle (SZA) and was the largest for purely scatter-
 86 ing aerosols at low SZAs ($\sim 20\%$). They also noted that models overestimated the TOA
 87 clear-sky flux under aerosol-free conditions by about 1.3 to 3.5% depending on the at-
 88 mospheric profiles and SZAs.

89 Aerosol properties from the AeroCom models have been compared against satel-
 90 lite retrievals and AEROSOL ROBOTIC NETwork (AERONET, Holben et al., 2001) mea-
 91 surements extensively (e.g., Kinne et al., 2006; Gliß et al., 2021). However, radiative fluxes
 92 from the AeroCom models have not been compared with the satellite observations. In
 93 this study, TOA fluxes from AeroCom phase III 2019 control experiment are compared
 94 with fluxes from the Clouds and the Earth’s Radiant Energy System (CERES) (Wielicki
 95 et al., 1996; Loeb et al., 2016). We will focus on clear-sky TOA shortwave (SW) flux com-
 96 parison, because of the large inter-model differences in cloud fraction (Stier et al., 2013).
 97 In order to understand the global and regional flux differences between CERES and the
 98 AeroCom models, differences in aerosol optical depth (AOD) and land surface albedo
 99 (α) are also presented, as these two variables contribute the most to TOA SW flux un-
 100 der clear-sky conditions. The observational-based SW flux is independently derived from
 101 AOD and land surface albedo datasets used in this study based on different satellite in-
 102 struments and algorithms. When the AOD and surface albedo differences can explain
 103 the SW flux differences, it is more likely that these differences are robust. Using differ-
 104 ent observations to evaluate the models synergistically can therefore be beneficial over
 105 using a single variable and provide more reliable diagnostics for model evaluation. Fur-
 106 thermore, relying on radiative kernels for AOD and surface albedo, one can tie the AOD
 107 and land surface albedo differences between models and observations to the flux differ-
 108 ences. The AeroCom models included in this study are briefly described in section 2. Satel-
 109 lite observations and datasets are in section 3, and the radiative kernels are described
 110 in section 4. Results on global and regional comparisons are presented in Section 5, and
 111 trend comparisons are in Section 6. Discussions and conclusions are in Section 7.

112 2 AeroCom Models

113 This study uses the AeroCom phase III 2019 control experiment ([https://wiki.met.
 114 no/aerocom/phase3-experiments](https://wiki.met.no/aerocom/phase3-experiments)). For this experiment, models use harmonized anthro-
 115 pogenic and biomass burning emissions from the Community Emission Data System (CEDS,
 116 Hoesly et al., 2018) for Coupled Model Intercomparison Project Phase 6 (CMIP6). Mod-
 117 eling centers are required to submit simulation results for at least 2010 and 1850, using
 118 2010 meteorology and prescribed sea-surface temperature from input4MIPS (Durack &
 119 Taylor, 2018). Among the models that participated in this control experiment, 9 mod-
 120 els provided all necessary variables (i.e. aerosol optical depth, TOA reflected SW flux
 121 under clear-sky conditions, surface upwelling and downwelling SW flux), and are included
 122 in this study. Table 1 lists these models, along with their spatial resolution and refer-
 123 ences describing the details of each model. All model outputs are linearly interpolated
 124 to a $1^\circ \times 1^\circ$ latitude-longitude grid to facilitate comparisons with satellite observational
 125 data. The global means calculated using the interpolated grid differ less than 0.01% from
 126 using the original spatial resolutions. A brief description of each model is given in Ap-
 127 pendix A.

Table 1. List of AeroCom models used in this study.

| Long name | Short name | lat \times long | Reference |
|-------------------------|-------------|----------------------------------|---------------------------|
| CAM5-ATRAS | CAM5 | $1.9^\circ \times 2.5^\circ$ | Matsui2017 |
| ECHAM6.3-HAM2.3-met2010 | ECHAM | $1.875^\circ \times 1.875^\circ$ | Tegen2019 |
| GFDL-AM4-met2010 | GFDLm | $1.00^\circ \times 1.25^\circ$ | Zhao2018 |
| GFDL-AM4-fsST | GFDLf | $1.00^\circ \times 1.25^\circ$ | Zhao2018 |
| GISS-ModelE2.1.1-OMA | GISS OMA | $2.0^\circ \times 2.5^\circ$ | Tsigaridis2013, Bauer2020 |
| GISS-ModelE2.1.1-MATRIX | GISS MATRIX | $2.0^\circ \times 2.5^\circ$ | Bauer2008 |
| INCA | INCA | $2.25^\circ \times 2.50^\circ$ | Balkanski2004, Schulz2009 |
| MIROC-SPRINTARS | SPRINTARS | $0.55^\circ \times 0.55^\circ$ | Takemura2005 |
| OsloCTM3v1.01-met2010 | Oslo | $2.25^\circ \times 2.25^\circ$ | Lund2018, Sovde2012 |

3 Satellite Observations

3.1 TOA Reflected Shortwave Flux

Clear-sky TOA radiative fluxes for $1^\circ \times 1^\circ$ latitude-longitude regions are from the CERES Energy Balanced and Filled (EBAF) Ed4.1 product (Loeb et al., 2018). EBAF data takes advantage of the many algorithm improvements that have been made in the Edition 4 CERES Level 1-3 data products and it is the only global dataset that can be used to study the variations of Earth radiation budget over a range of temporal and spatial scales. The Earth’s energy imbalance in the CERES EBAF is constrained to be consistent with ocean heat content (Johnson et al., 2016) by using an objective constraint algorithm to adjust SW and longwave (LW) TOA fluxes within their ranges of uncertainty to remove the inconsistency between average global net TOA flux and heat storage in the Earth-atmosphere system (Loeb et al., 2009, 2018). Additionally, because of the relatively coarse spatial resolution of the CERES instruments (~ 20 -km at nadir), the standard CERES Level-3 data products have many spatial gaps in monthly mean clear-sky TOA flux maps due to the absence of cloud-free areas occurring at the CERES footprint scale in some $1^\circ \times 1^\circ$ regions. In EBAF, this problem is mitigated by inferring clear-sky fluxes from both CERES and MODIS measurements to produce a new clear-sky TOA flux climatology for every $1^\circ \times 1^\circ$ grid box every month. The uncertainty for TOA clear-sky SW flux is estimated as 5 Wm^{-2} on the grid box level (Loeb et al., 2018). EBAF data has been widely used to evaluate global general circulation models (e.g., Pin-cus et al., 2008; Su et al., 2010; H. Wang & Su, 2013; Wild et al., 2013; Paulot et al., 2018; S. E. Bauer et al., 2020; Loeb et al., 2020).

3.2 Aerosol Optical Depth

Aerosol optical depths (AOD) retrieved from Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-Angle Imaging Spectroradiometer (MISR) are used in this study to compare with the model simulations and to help interpreting the SW flux biases. The Aqua MODIS collection 6.1 monthly gridded Dark Target and Deep Blue merged AOD product (Sayer et al., 2014) combines AODs retrieved from Dark Target over-water algorithm with Dark Target and Deep Blue over land algorithms to provide more complete AOD spatial coverage over snow-ice-free surfaces. Validations against AERONET and Maritime Aerosol Network (MAN) data indicate that MODIS dark target retrievals agree well with AERONET over land (high correlation and low bias) (Levy et al., 2013). Over ocean, Aqua MODIS AODs are also highly correlated with those from AERONET, but biased high at low AODs and the scatter for high AODs is significantly larger than

the expected error (Levy et al., 2013; Schutgens et al., 2020). Validations of the deep blue retrievals over land also indicate very low bias (Sayer et al., 2014; Schutgens et al., 2020).

MISR Level 3 gridded AOD product at $0.5^\circ \times 0.5^\circ$ spatial resolution is also used in this study. The MISR Level 3 product is aggregated from higher spatial resolution version 23 (V23) level 2 data (Garay et al., 2020). Compared to the V22 AOD retrieval, V23 implemented many changes which resulted in significant reduction in AOD over ocean. The 16-year mean AOD is reduced from 0.157 in V22 to 0.114 in V23 over ocean. However, the AODs over land agree well between V22 and V23. Validations against the AERONET measurements show modest improvement in V23 retrievals over land in comparison to V22, with the absolute bias decreases from -0.004 to -0.002 and the percent of retrievals that fall within the error envelope, defined as $\pm(0.03+10\%AOD)$, increases from 59.7% to 66.1%. However, V23 retrievals tend to overestimate AOD at low AODs (like MODIS, mentioned above) and underestimate AOD at high AODs. Validations against MAN AOD derived from handheld Microtops Sun photometers show significant improvement in V23, the absolute bias is reduce from 0.037 to 0.0 and the percent of retrievals that fall within the error envelope increases from 61% to 87% in comparison to V22.

3.3 Surface Albedo

The Terra and Aqua combined MODIS Bidirectional Reflectance Distribution Function (BRDF)/albedo product (MCD43C1, Version 6) provides the daily weighting parameters to calculate the directional hemispherical reflectance (black-sky albedo) and bihemispherical reflectance (white-sky albedo) at a spatial resolution of 0.05° over land. These parameters are used in polynomial albedo representations to estimate the black-sky albedo at any given SZA and the white-sky albedo (Li & Strahler, 1992; Lucht et al., 2000). The MODIS broadband surface albedos agree well with the in-situ ground-based and airborne measurements, the root mean square errors are less than 0.020 for forest during the dormant periods and 0.025 during the snow-covered periods, less than 0.030 for agriculture and grassland during the dormant periods and 0.050 during the snow-covered periods, and less than 0.047 for the snow covered tundra (Z. Wang et al., 2012, 2014; Cescatti et al., 2012; Roman et al., 2013). The MODIS broadband surface albedos also agree with other satellite products to within 0.01 (Taberner et al., 2010; Carrer et al., 2010).

The actual surface albedo is a combination of black-sky albedo and white-sky albedo from the following (Schaaf et al., 2002):

$$\alpha(\theta_0, AOD, \phi) = \alpha_{ws} \times f(\theta_0, AOD, \phi) + \alpha_{bs}(\theta_0) \times (1 - f(\theta_0, AOD, \phi)) \quad (1)$$

where α_{ws} is the white-sky albedo and α_{bs} is the black-sky albedo, f is the fraction of diffuse light and depends on the SZA (θ_0), AOD, and aerosol types (ϕ). A look-up table of f (available at https://www.umb.edu/spectralmass/terra_aqua_modis/modis_user_tools) is provided for continental and maritime aerosol types with AODs ranging from 0 to 0.98 for SZAs between 0° and 89° .

For a given month, actual surface albedo is calculated every 10 minutes to take into account that black-sky albedo is a function of SZA and the value of f is determined using monthly MODIS Dark Target and Deep Blue merged AOD assuming continental aerosol type. At every time step within every $1^\circ \times 1^\circ$ grid, surface albedo calculated at 0.05° resolution are converted to surface upwelling SW fluxes by using the clear-sky surface downwelling SW fluxes from CERES Edition 4 synoptic daily hourly product (SYN1deg-1Hour, Doelling et al., 2013), then average into the $1^\circ \times 1^\circ$ grid. These upwelling SW fluxes are averaged over the month, then divided by the monthly mean downwelling SW flux to produce the monthly mean surface albedo.

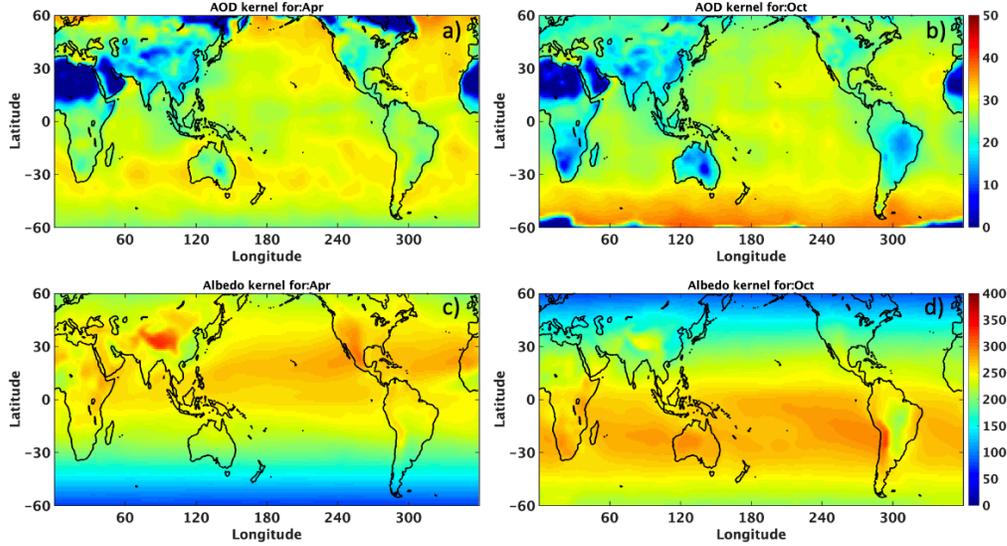


Figure 1. The monthly mean TOA shortwave flux radiative kernels (in Wm^{-2}) for aerosol optical depth (a) April, (b) October, and surface albedo (c) April, (d) October.

4 Radiative Kernels

209

210 Radiative kernels are partial derivatives of TOA SW flux derived by imposing small
 211 systematic perturbations to the base-state values. Thorsen et al. (2020) derived aerosol
 212 radiative kernels using 1 year (2007) of 3-hourly MERRA-2 data. Radiative kernels of
 213 aerosol optical depth, single-scattering albedo (SSA), aerosol asymmetry factor, and sur-
 214 face albedo are derived for each month. These kernels are able to reproduce the aerosol
 215 direct radiative effect to within 0.3 Wm^{-2} when compared to the true aerosol direct ra-
 216 diative effect calculation in MERRA-2. These kernel calculations are very computational
 217 costly, thus kernels are only calculated using 2007 data. Although it would be most ac-
 218 curate to apply these kernels to the same year of measurements and simulations, the im-
 219 pact of interannual variability on these kernels is expected to be small (Thorsen et al.,
 220 2020) and they are applied to the AeroCom simulations of 2010 (the output year selected
 221 by the AeroCom phase III experiment).

222 Figure 1 shows the AOD and surface albedo kernels for April and October (differ
 223 from the annual mean results presented in Thorsen et al. (2020)). Kernels are expressed
 224 in units of watts per meter squared per unit change in the respective variables. Figures
 225 1a and 1b show that the TOA SW flux would increase by more than 30 Wm^{-2} over the
 226 oceans off the west coast of Africa if the AOD were increased by 1, whereas the incre-
 227 ment is less than 10 Wm^{-2} over the Sahara desert because TOA SW flux has smaller
 228 sensitivity to AOD changes over bright surface than over dark surface. Aerosol compo-
 229 sition also affects the AOD kernels. For example, the AOD radiative kernel over South
 230 America is about 25 Wm^{-2} in April and is reduced to 15 Wm^{-2} in October (see Fig-
 231 ure 1b) during the biomass burning season. Figure 1c and 1d shows the regional TOA
 232 SW flux changes for unit increase in surface albedo, which are mostly sensitive to solar
 233 insolation and are also sensitive to the initial surface albedo. In this study, we use these
 234 kernels to assess the contributions of AOD and surface albedo biases to TOA SW flux
 235 biases in the AeroCom models.

236

The clear-sky TOA reflected SW flux bias in the model can be expressed as:

$$\Delta F = F_m - F_o = \Delta F_{AOD} + \Delta F_\alpha + \Delta F_\chi, \quad (2)$$

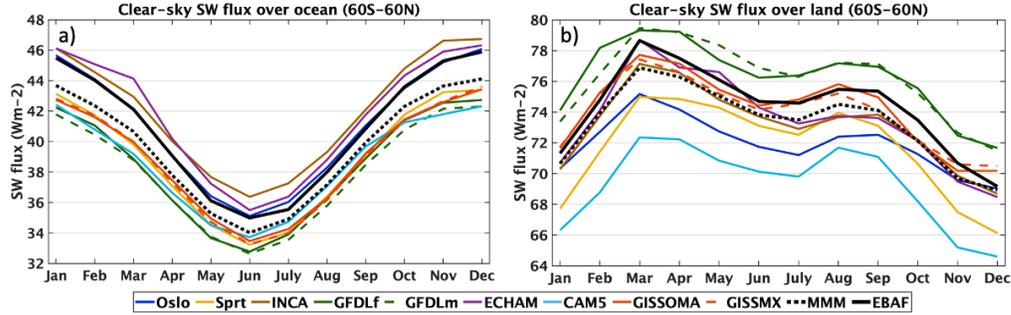


Figure 2. TOA clear-sky reflected SW flux comparisons between CERES EBAF, individual AeroCom models, and the multi-model mean (MMM) over ocean (a) and land (b) between 60°S and 60°N.

237 where F_m and F_o are the TOA reflected SW flux from models and CERES EBAF, ΔF_{AOD}
 238 and ΔF_α are the flux biases caused by biases in AOD and surface albedo in models rel-
 239 ative to MODIS retrievals calculated using their respective radiative kernels, and ΔF_χ
 240 is the residual in flux bias, which can be attributed to differences in aerosol composition
 241 and atmospheric state (i.e. water vapor, ozone, etc.), and also to uncertainties in radiative
 242 transfer calculations, satellite retrievals, and kernel calculations.

243 5 Comparisons Between Models and Observations

244 Figure 2 shows the seasonal cycle of monthly mean clear-sky SW flux calculated
 245 over ocean and land between 60°S and 60°N (60S-60N) from CERES EBAF (black line),
 246 nine AeroCom models, and the multi-model mean (MMM, black dashed line) result for
 247 2010. Figure 3 shows the seasonal cycle of monthly mean AOD over 60S-60N ocean and
 248 land from MODIS, MISR, nine AeroCom models, and the MMM. Over ocean, the sea-
 249 sonal cycle of SW flux is very similar to that of solar insolation as the AODs remain rel-
 250 atively constant throughout the year (Figure 3a). Clear-sky SW fluxes from INCA and
 251 ECHAM-HAM models are greater than those from CERES EBAF throughout the year,
 252 and AODs from these two models are also on the high end when compared with the satel-
 253 lite retrievals, consistent with the AOD assessment from Gliß et al. (2021). SW flux from
 254 Oslo agrees with EBAF almost perfectly, despite its AOD is on the low end when com-
 255 pared with satellite retrievals. Monthly mean clear-sky SW fluxes from all other mod-
 256 els are smaller than EBAF by about 2-3 Wm^{-2} , and their AODs are generally smaller
 257 than the MODIS retrievals as well. It is worth noting that even though AODs from SPRINT-
 258 ARS are much lower than all other models, its SW fluxes are actually greater than a cou-
 259 ple of other models because its aerosols are almost non-absorbing (see figure 10). The
 260 MMM fluxes are about 1-2 Wm^{-2} smaller than CERES EBAF and the MMM AODs
 261 lie in between MODIS and MISR AODs. Over land, the GFDL models produce greater
 262 clear-sky SW fluxes than EBAF, GISS models agree with EBAF fairly well, and all other
 263 models have low biases. This results in MMM being 1-2 Wm^{-2} smaller than CERES EBAF.
 264 AODs from all models are outside the boundaries of satellite retrievals. Thus the MMM
 265 AODs are smaller than both MODIS and MISR AODs except during the boreal sum-
 266 mer months. There are no correspondences between flux biases and AOD biases over land,
 267 as land surface albedo biases from these models also contribute to the flux biases.

268 The seasonal cycles of AODs from MODIS and MISR are very similar over both
 269 ocean and land (Figure 3). However, the AOD seasonal cycles from the AeroCom mod-
 270 els differ significantly from the observations and from each other, especially over land.
 271 The MISR AODs are smaller than the MODIS AODs. As mentioned in section 3.2, the

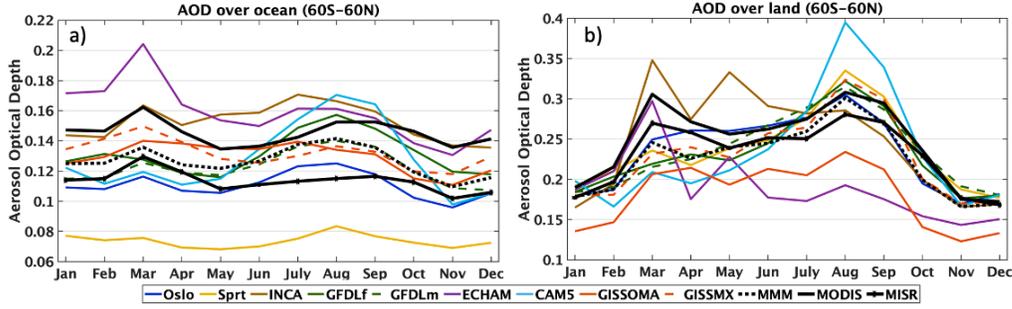


Figure 3. Aerosol optical depth comparisons between satellite retrievals (MODIS and MISR), individual AeroCom models, and the multi-model mean (MMM) over ocean (a) and land (b) between 60°S and 60°N.

Table 2. Annual mean SW flux biases of multi-model mean relative to CERES EBAF (ΔF) over land and ocean, flux biases due to AOD biases ΔF_{AOD} and land surface albedo biases ΔF_{α} , and the flux bias residues ΔF_{χ} .

| | Land | | Ocean | |
|---------------------|--------|----------|--------|----------|
| | Global | Tropical | Global | Tropical |
| ΔF | -1.0 | -1.7 | -1.7 | -2.8 |
| ΔF_{AOD} | -0.4 | -0.4 | -0.4 | -0.8 |
| ΔF_{α} | -0.3 | -0.9 | — | — |
| ΔF_{χ} | -0.3 | -0.4 | -1.3 | -2.0 |

272 mean AOD over ocean in the recently released MISR V23 is smaller than V22 by about
 273 0.04, while the mean AOD over land is nearly unchanged. Schutgens et al. (2020) inter-
 274 compared AOD retrievals from 14 satellite products (MISR was not included) and eval-
 275 uated them against the AERONET and MAN results. When collocating each individ-
 276 ual satellite product with either AERONET or MAN, they found that over land Aqua
 277 MODIS AODs show good agreement with AERONET (high correlation and low bias);
 278 Aqua MODIS AODs over ocean are also highly correlated with those from AERONET
 279 and MAN, but are biased high. They also argued that the satellite retrieval diversity can
 280 be used as a proxy for retrieval uncertainty. Judging from their analysis, it is reasonable
 281 to assume Aqua MODIS retrievals represent the high end of AODs whereas MISR re-
 282 trievals represent the low end over ocean. It is also notable that monthly gridded AODs
 283 from MISR often suffer spatial gaps and incoherent features because of MISR’s narrow
 284 swath, and the sampling difference also likely contributed to the global mean AOD dif-
 285 ference seen between MODIS and MISR. Hence, in the following discussion, we only present
 286 results using MODIS AODs, but the regional difference features with respect to mod-
 287 els remain the same when MISR AODs are used instead (see Appendix B).

288 To understand the contributions of AOD and land surface albedo biases to TOA
 289 SW flux biases, ΔF_{AOD} is calculated over ocean and land and ΔF_{α} is calculated over
 290 land using their respective radiative kernels. Figure 4a shows the monthly mean ΔF for
 291 MMM over 60S-60N and tropical (30°S-30°N) ocean (solid lines), and the flux residu-
 292 als (ΔF_{χ}) after considering the contribution of AOD biases to ΔF (dashed lines). Flux
 293 biases are reduced by up to 0.6 Wm^{-2} over 60S-60N ocean and by up to 1 Wm^{-2} over
 294 tropical ocean after accounting for ΔF_{AOD} . Figure 4b shows the monthly mean ΔF for
 295 MMM over 60S-60N and tropical land (solid line), and after accounting for the contri-

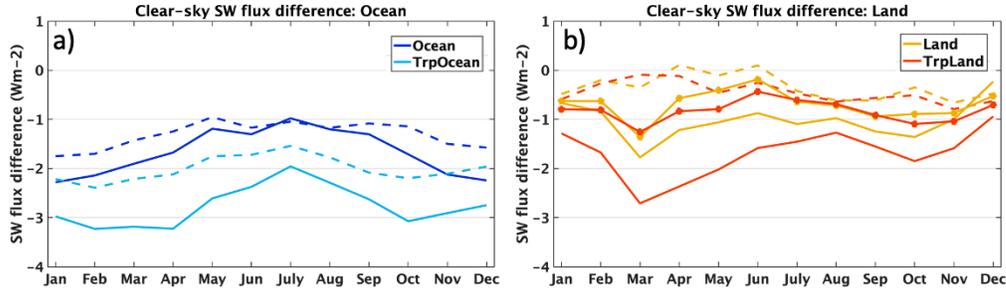


Figure 4. Monthly mean multi-model mean SW flux biases relative to CERES EBAF (solid lines) for 2010 over 60°S–60°N and tropical (30°S–30°N) ocean (a) and land (b). Over ocean, the dashed lines are the flux bias residuals (ΔF_χ) after accounting for the contributions of AOD biases to the flux biases. Over land, lines with circles represent ($\Delta F - \Delta F_\alpha$) and dashed lines are the flux bias residuals after accounting for the contributions of both AOD and surface albedo biases to the flux biases.

296 bution of ΔF_{AOD} (solid lines with circles) and after accounting for both ΔF_{AOD} and
 297 ΔF_α (dashed lines). Flux biases are reduced by up to 3 Wm^{-2} after accounting for bi-
 298 ases in AOD, and are reduced further by accounting for biases in surface albedo. Table
 299 2 summarizes the annual mean ΔF for MMM, and how much AOD and surface albedo
 300 (only over land) biases contribute to ΔF . Over ocean, accounting for the AOD bias re-
 301 duces the MMM flux bias by about 25%. Over land, accounting for the AOD and sur-
 302 face albedo biases reduces the MMM flux bias by about 70%. The residual in flux bias
 303 can be explained by differences in aerosol composition, radiative transfer calculation un-
 304 certainties of the models (Randles et al., 2013), and uncertainties in the radiative ker-
 305 nels and in satellite retrievals.

306 The monthly mean ΔF for AeroCom models and their respective ΔF_χ are show
 307 in Figure 5 for all models listed in Table 1. Over ocean, accounting for the biases in AOD
 308 reduces flux biases for almost all models, with larger impact over tropical oceans than
 309 over global oceans. Over land, accounting for the biases in AOD and surface albedo not
 310 only reduces the flux biases but also minimizes the seasonal dependence for Oslo, SPRINT-
 311 ARS, INCA and CAM5 models. For the GFDL models, accounting for the biases in AOD
 312 and surface albedo reverses the models from overestimating to underestimating the flux
 313 of about the same magnitude. For ECHAM model, ΔF_χ is slightly more negative than
 314 ΔF . For the two GISS models, accounting for the biases in AOD and surface albedo in-
 315 creases the flux biases by about 5 Wm^{-2} . This is largely due to the flux biases having
 316 little correspondence with the AOD and albedo biases, particularly over the Amazon,
 317 central Africa, and the Tibetan Plateau (see Figure 8).

318 On a $1^\circ \times 1^\circ$ latitude-longitude grid box level, the flux biases due to AOD and sur-
 319 face albedo biases derived from using the radiative kernels correlate very well with ΔF
 320 for MMM. Figure 6a shows the relationship between ΔF and ΔF_{AOD} over ocean using
 321 all grid boxes between 50°S–40°N of the entire year of 2010 (approximately 255,000 data
 322 points used), and the correlation coefficient is 0.6. MODIS retrievals produce some spu-
 323 riously large AODs over high latitude oceans (possibly due to the presence of sea ice)
 324 and are therefore excluded in the correlation analysis. Figure 6b shows the relationship
 325 between ΔF and $\Delta F_{AOD} + \Delta F_\alpha$ over land using all grid boxes between 60°S–60°N
 326 for the entire year of 2010 (approximately 142,000 data points used), and the correlation co-
 327 efficient is 0.76. Table 3 lists the correlation coefficients for MMM and nine AeroCom
 328 models between ΔF and ΔF_{AOD} over ocean and between ΔF and $\Delta F_{AOD} + \Delta F_\alpha$ over
 329 land for four seasonal months and the entire year. Over land, MMM has the highest cor-

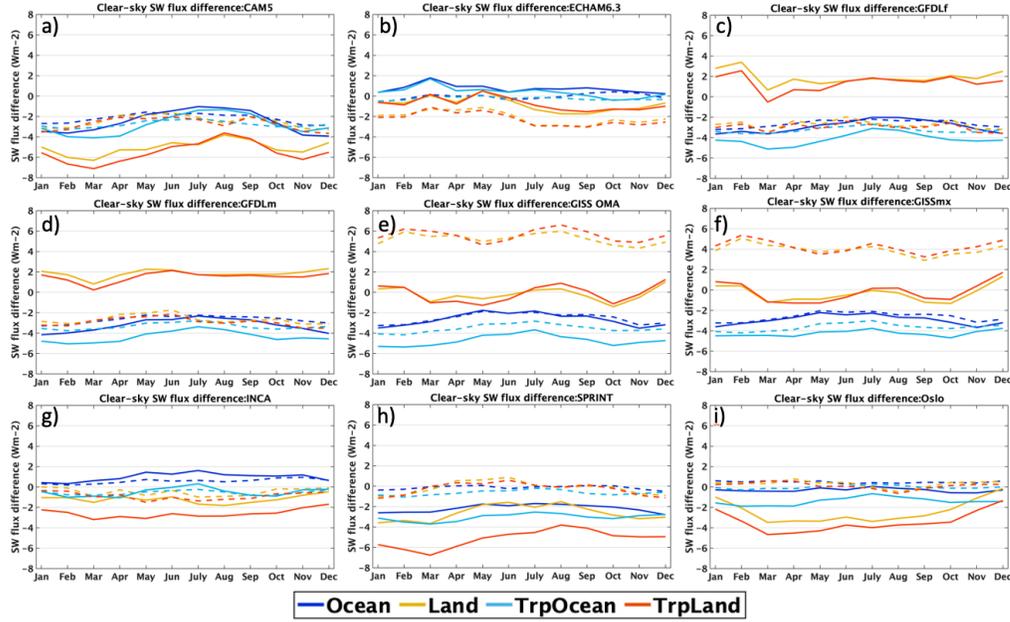


Figure 5. Monthly mean SW flux biases of the AeroCom models for 2010 over 60°S – 60°N ocean (solid blue lines) and land (solid orange lines), and over tropical (30°S – 30°N) ocean (solid cyan lines) and land (solid red lines). The dashed lines are the flux bias residuals (ΔF_{χ}) for the corresponding regions. (a) CAM5, (b) ECHAM, (c) GFDL, (d) GFDLm, (e) GISS OMA, (f) GISS MATRIX, (g) INCA, (h) SPRINTARS, and (i) Oslo.

330 relation in January and the lowest in October; Over ocean, MMM has the highest cor-
 331 relation in April and the lowest correlation in October. Correlation coefficients for in-
 332 dividual models vary across the seasons. Over land, the yearly correlation coefficient
 333 is the highest for SPRINT model ($r=0.94$) and lowest for GISS models ($r=0.78$). Over ocean,
 334 the yearly correlation coefficient is the highest for CAM5 and ECHAM-HAM models ($r=0.78$)
 335 and lowest for GISS MATRIX model ($r=0.52$).

336 Figure 7 shows the MMM regional TOA SW flux biases (a), SW flux biases due
 337 to biases in AOD and surface albedo calculated from their radiative kernels (b), AOD
 338 biases (c), and land surface albedo biases(d) for April 2010. For this month the 60S – 60N
 339 mean ΔF is -1.8 Wm^{-2} and -1.2 Wm^{-2} over ocean and land. The spatial patterns of
 340 flux bias derived from kernels correspond well with ΔF . Correlation coefficient is 0.66
 341 between ΔF and ΔF_{AOD} over ocean, and is 0.75 between ΔF and $\Delta F_{AOD} + \Delta F_{\alpha}$ over
 342 land. After accounting for the contributions of AOD and land surface albedo to ΔF , the
 343 60S – 60N mean flux bias (ΔF_{χ}) is reduced to -1.3 Wm^{-2} and 0.1 Wm^{-2} over ocean and
 344 land.

345 Good correspondence between ΔF and flux biases derived from radiative kernels
 346 is also observed for all models. Figure 8 shows the regional ΔF (left panels), and SW
 347 flux biases due to AOD and land surface albedo biases calculated from their radiative
 348 kernels (right panels) for April 2010. The spatial distributions between the two GFDL
 349 models are very similar; hence, only the GFDL (observed SST without wind nudging)
 350 simulation is shown. The GISS MATRIX model performs better than GISS OMA model
 351 in terms of simulating sulfate aerosols and AOD (S. E. Bauer et al., 2020). We also find
 352 that GISS MATRIX model agrees better than GISS OMA when compared to MODIS
 353 and MISR AODs. Here only results from GISS MATRIX model are shown.

Table 3. Correlation coefficients between ΔF and $\Delta F_{AOD} + \Delta F_{\alpha}$ over land, and between ΔF and ΔF_{AOD} over ocean for January, April, July, October and the entire year of 2010 for multi-model mean (MMM) result and the nine AeroCom models.

| Model | Land (60°S–60°N) | | | | | Ocean (50°S–40°N) | | | | |
|-------------|------------------|------|------|------|------|-------------------|------|------|------|------|
| | Jan | Apr | July | Oct | Year | Jan | Apr | July | Oct | Year |
| MMM | 0.82 | 0.75 | 0.73 | 0.72 | 0.76 | 0.65 | 0.66 | 0.53 | 0.48 | 0.60 |
| CAM5 | 0.88 | 0.81 | 0.73 | 0.70 | 0.81 | 0.78 | 0.77 | 0.79 | 0.70 | 0.78 |
| ECHAM | 0.84 | 0.86 | 0.69 | 0.71 | 0.81 | 0.78 | 0.78 | 0.72 | 0.68 | 0.78 |
| GFDLm | 0.83 | 0.83 | 0.72 | 0.71 | 0.79 | 0.62 | 0.71 | 0.53 | 0.54 | 0.61 |
| GFDLf | 0.89 | 0.86 | 0.73 | 0.70 | 0.84 | 0.58 | 0.76 | 0.53 | 0.69 | 0.68 |
| GISS OMA | 0.81 | 0.78 | 0.75 | 0.75 | 0.78 | 0.74 | 0.69 | 0.62 | 0.68 | 0.68 |
| GISS MATRIX | 0.80 | 0.78 | 0.76 | 0.75 | 0.78 | 0.62 | 0.52 | 0.44 | 0.45 | 0.52 |
| INCA | 0.85 | 0.87 | 0.84 | 0.85 | 0.85 | 0.70 | 0.72 | 0.65 | 0.59 | 0.67 |
| SPRINTARS | 0.93 | 0.94 | 0.95 | 0.94 | 0.94 | 0.55 | 0.66 | 0.56 | 0.54 | 0.59 |
| Oslo | 0.86 | 0.88 | 0.85 | 0.84 | 0.85 | 0.74 | 0.69 | 0.54 | 0.51 | 0.64 |

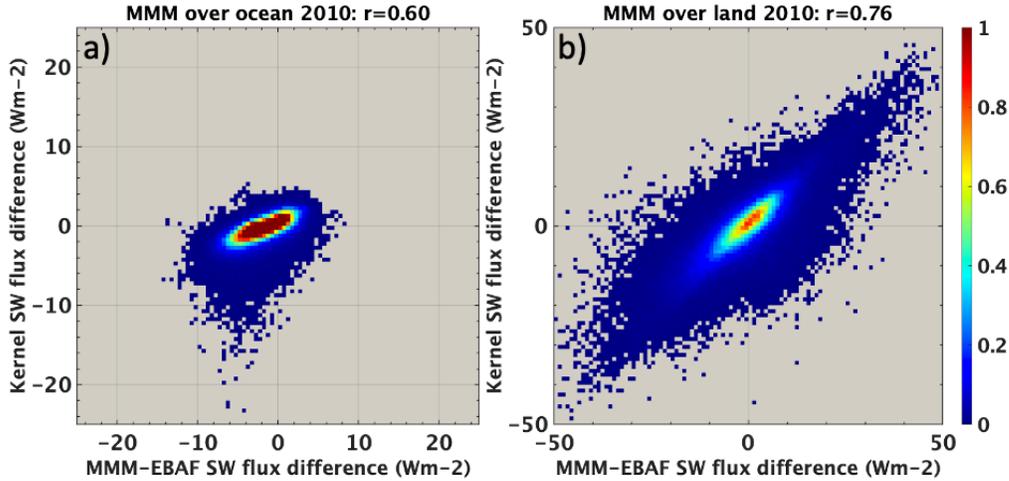


Figure 6. Relationship between SW flux biases of multi-model mean relative to CERES EBAF and kernel based flux biases using all grid boxes over ocean between 50°S–40°N using all 12 months of 2010 (a), and using all grid boxes over land between 60°S–60°N using all 12 months of 2010 (b). Over land, kernel SW flux biases are $\Delta F_{AOD} + \Delta F_{\alpha}$. Over ocean, kernel SW flux biases are ΔF_{AOD} . Color bar shows the relative number density.

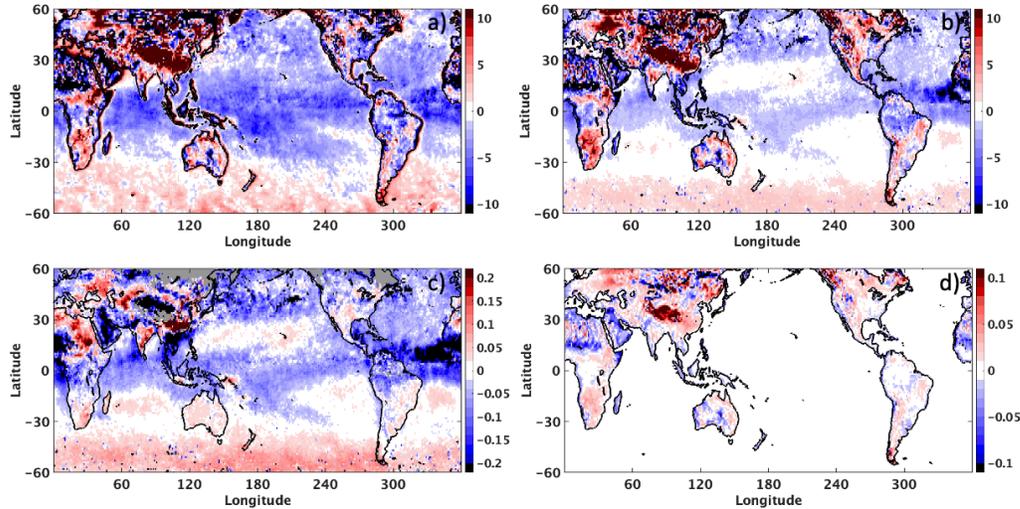


Figure 7. TOA clear-sky reflected SW flux biases of multi-model mean (MMM) (a), and TOA clear-sky reflected SW flux biases due to AOD biases (shown in c) and surface albedo biases (shown in d) calculated from radiative kernels (b), aerosol optical depth biases of MMM relative to MODIS (c), and land surface albedo biases of MMM relative to MODIS (d) for April 2010.

354 Over ocean, the spatial distribution of ΔF corresponds well with ΔF_{AOD} for all
 355 models. For April, ECHAM-HAM model has the highest correlation coefficient of 0.78
 356 and GISS MATRIX model has the lowest correlation coefficient of 0.52 (Table 3). A com-
 357 mon feature for all models is that they underestimate the dust outflow off the west coast
 358 of Africa (Figure 9), thus also underestimate the TOA SW flux. All models except CAM5
 359 and SPRINTARS overestimate AOD and SW flux over the Southern Ocean, with INCA
 360 model has the largest overestimation. It is worth pointing out that several models (i.e.
 361 ECHAM-HAM, GISS MATRIX, and SPRINTARS) significantly overestimate AOD and
 362 flux over New Caledonia and surroundings. Over land, the spatial distribution of ΔF
 363 and $\Delta F_{AOD} + \Delta F_{\alpha}$ are highly correlated, despite that there are large diversities among
 364 models in terms of AOD and land surface albedo (Figure 9). The highest correlation co-
 365 efficient is 0.94 for SPRINTARS model and the lowest is 0.78 for GISS models for April.
 366 The high correlations between ΔF and kernel-based flux biases indicate that AOD and
 367 land surface albedo biases can explain most of the SW flux biases. This finding is sig-
 368 nificant as the MODIS AOD and surface albedo, and the CERES clear-sky flux are de-
 369 rived independently from each other (i.e., they rely upon different measurements and al-
 370 gorithms). Consistencies in their biases indicate that the AOD biases shown in this study
 371 are robust and constraining the modeled AODs by satellite observations and correcting
 372 the land surface albedo used in the models will improve the SW flux agreement between
 373 models and CERES EBAF.

374 The consistency in spatial distributions between AOD/surface albedo biases and
 375 SW flux biases demonstrates that independently derived satellite products are valuable
 376 in diagnosing model deficiencies when used jointly. Consistency in regional features of
 377 these variables can be helpful in identifying the particular processes and/or parameter-
 378 izations that are responsible for these biases. For example, when comparing AODs off
 379 the west coast of Africa from MODIS with AODs from the models, it is unclear if the
 380 MODIS retrieval overestimates the dust outflow from Africa or if the models underes-
 381 timate the dust outflow. Only by comparing the clear-sky TOA SW fluxes from CERES
 382 with those from the models confirms that the models indeed underestimate the dust out-
 383 flow there which result in low biases of SW flux. One may argue that overestimating dust

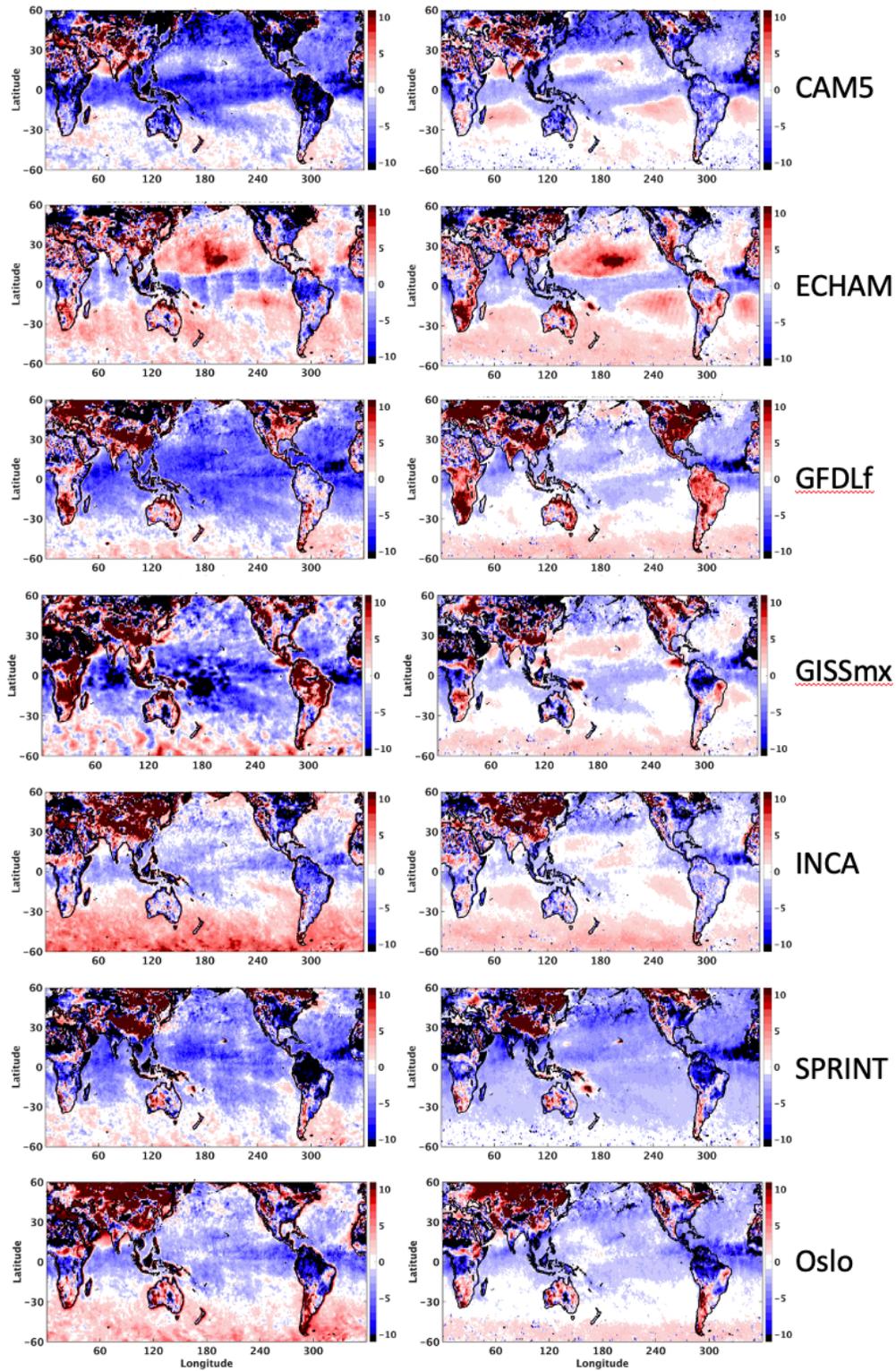


Figure 8. TOA clear-sky reflected SW flux biases for AeroCom models relative to CERES EBAF for April 2010 (left panels). Right panels use radiative kernel calculations to determine the flux biases associated with AOD and surface albedo biases.

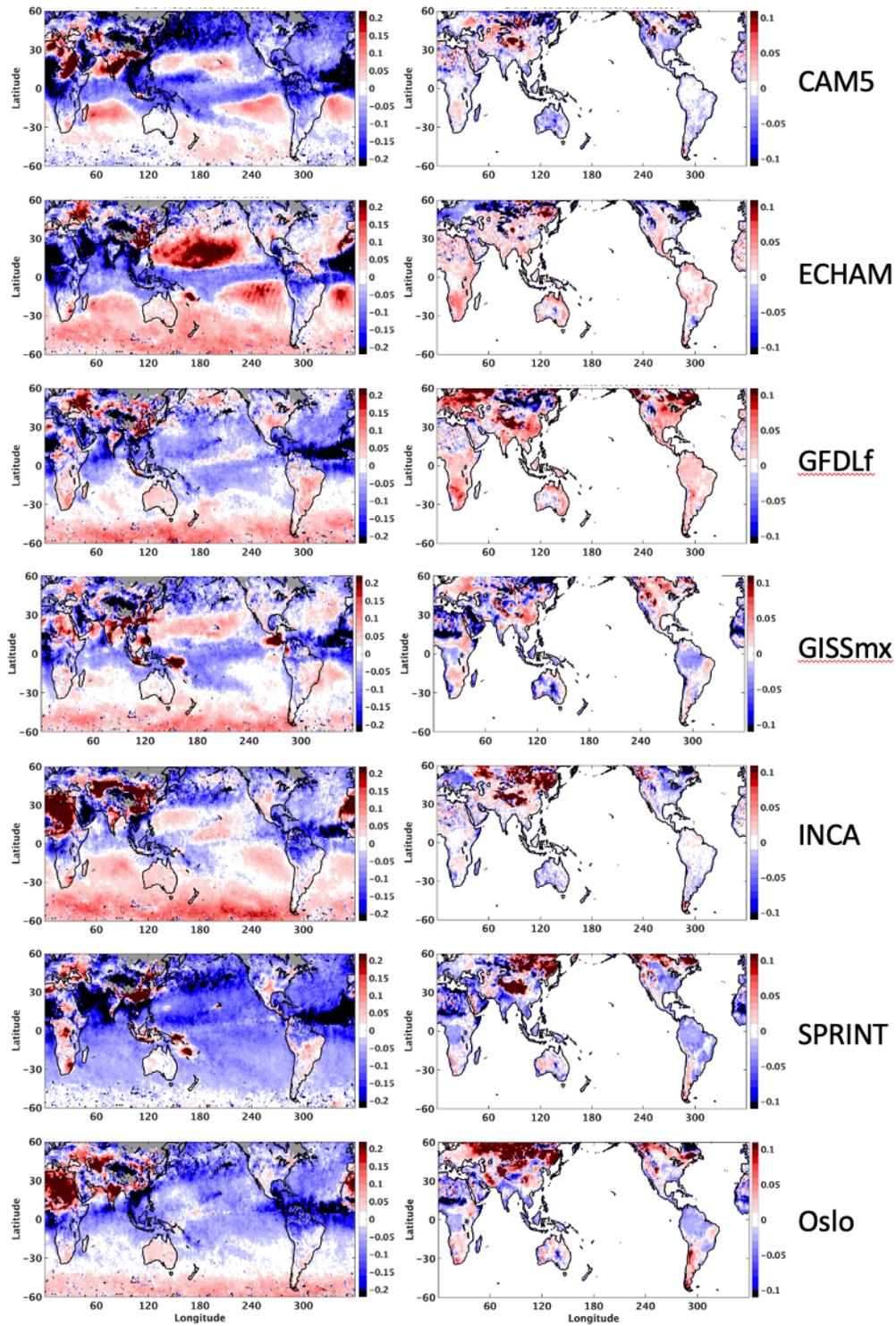


Figure 9. AOD biases (left panels) and land surface albedo biases (right panels) of AeroCom models relative to MODIS retrievals for April 2010.

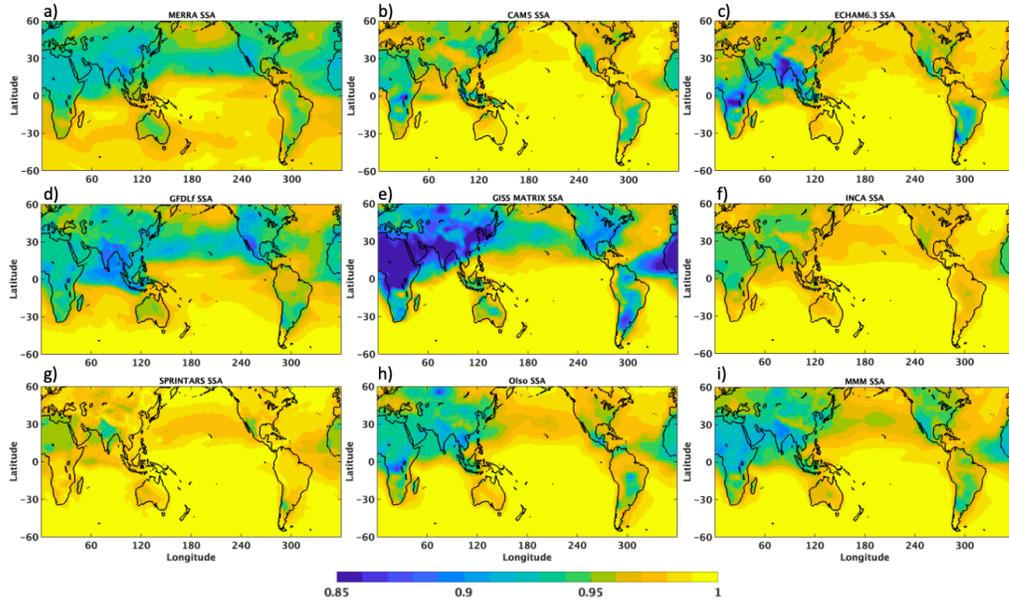


Figure 10. Single-scattering albedo for (a) MERRA-2, (b) CAM5, (c) ECHAM, (d) GFDL, (e) GISS MATRIX, (f) INCA, (g) SPRINTARS, (h) Oslo, and (i) multi-model mean for April. MERRA-2 result is for 2007, and all others are for 2010.

384 aerosol absorption can also contribute to the flux biases. However, to explain the mag-
 385 nitude of flux bias shown in Figure 8, biases of dust aerosol SSA need to be on the order
 386 of 0.25 based on the SSA radiative kernels from Thorsen et al. (2020). We thus con-
 387 clude that the flux biases over the west coast of Africa are mostly due to AOD biases.
 388 Additionally, accounting for the impact of AOD and surface albedo biases on SW flux
 389 using radiative kernels of these variables significantly improves the regional flux agree-
 390 ment between models and CERES EBAF. The agreements of monthly global and tropi-
 391 cal means over ocean and land are also improved for all models except for the GISS mod-
 392 els.

393 As mentioned before, differences in aerosol composition, radiative transfer calcu-
 394 lation uncertainties of the models (Randles et al., 2013), and uncertainties in the radi-
 395 ative kernels and in satellite retrievals can all contribute to ΔF_{χ} . The radiative kernels
 396 that we use in this study are based on MERRA-2 reanalysis, whose base-state aerosols
 397 are different from the AeroCom models. Over ocean, aerosols in MERRA-2 are gener-
 398 ally more absorbing than the AeroCom models (Figure 10). Thus the AOD kernels over
 399 ocean are less sensitive to changes in AOD than if an AeroCom model (less absorbing)
 400 is used as the base state. Over land, single scattering albedo (SSA) from many AeroCom
 401 models agree reasonable well with that from MERRA-2 (global mean SSA difference is
 402 about 0.02-0.03), except that GISS MATRIX model has much lower SSA than MERRA-
 403 2 (global mean SSA difference exceeds 0.06) whereas SPRINTARS model has much higher
 404 SSA than MERRA-2 (global mean SSA difference is about 0.04). The differences in aerosol
 405 composition affects the magnitude of kernel derived flux biases from AOD and surface
 406 albedo biases and are part of ΔF_{χ} . Although accounting for the biases in AOD and sur-
 407 face albedo between AeroCom models and satellite retrievals does not entirely eliminate
 408 the TOA SW flux bias, it certainly reduces the global mean biases (except for GISS mod-
 409 els over land potentially due to its aerosols are very absorbing) and mitigates large re-
 410 gional biases for all models. Currently no aerosol composition observations on the global
 411 scales are available for constraining the model simulations, but correcting the aerosol load-

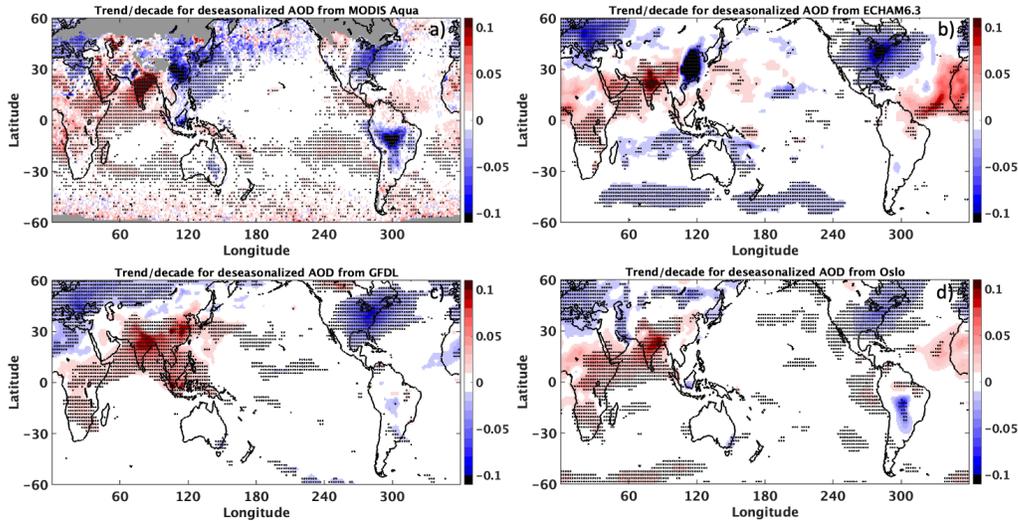


Figure 11. Aerosol optical depth trends (per decade) calculated using satellite retrievals from Aqua MODIS (a) and using ECHAM-HAM (b), GFDL (c), and Oslo (d) model simulations. Data from July 2002 to December 2017 are used here. All models use emissions from CEDS until 2014, but use different emissions between 2015 and 2017.

412 ing and land surface albedo in the models then adjusting the aerosol composition, size
 413 distribution, effective refractive indices, and aerosol hygroscopic growth to minimize the
 414 residual fluxes can further improve the TOA SW flux agreement between models and
 415 observation. Radiative kernels of each model can be developed to achieve maximum con-
 416 sistency to guide the model development.

417 6 Regional Trends

418 ECHAM-HAM, GFDL (AMIP run), and Oslo models also provide historical sim-
 419 ulations which can be used to calculate the regional trends in aerosol distributions. Here
 420 we compare the regional trends in AODs derived from these models with those from Aqua
 421 MODIS retrievals using the time period (July 2002 to December 2017) common to both.
 422 All three models use emissions from CEDS until 2014. However, each model handles the
 423 emissions between 2015 and 2017 differently. ECHAM-HAM used the CMIP6 Shared
 424 Socioeconomic Pathways (SSP) 3-7.0 emission scenario (Gidden et al., 2019). Oslo used
 425 the SSP2-4.5 middle of the road emission scenario (Fricko et al., 2017). GFDL used CEDS
 426 anthropogenic and biomass burning emissions of 2014 for 2015-2017, and the dust and
 427 sea-salt emissions are calculated using the actual wind speed produced by the model. Fig-
 428 ure 11 shows the AOD trend per decade calculated from Aqua MODIS and from model
 429 simulations, and areas with stipplings indicate the trend is significant at the 95% con-
 430 fidence interval. All three models reproduce the decreasing aerosol trends over Europe,
 431 eastern United States and the Atlantic Ocean, and the increasing trends over India, In-
 432 dian Ocean, Arabian Peninsular, and Central Africa. The most notable difference among
 433 the models is over eastern China and the adjacent oceanic regions where MODIS indi-
 434 cates a decreasing trend. The vastly different trends among the models are due to dif-
 435 ferent emission data used between 2015-2017 for the three models, and the fact that some
 436 pathway scenarios significantly underestimate the recent decline in anthropogenic aerosol
 437 emissions over China (Z. Wang et al., 2021).

The direct aerosol radiative effect (DARE) is the difference between the TOA SW flux for a pristine atmosphere with no aerosols and the actual atmosphere with aerosols (Loeb et al., 2021):

$$DARE = F(0, \alpha, X) - F(AOD, \alpha, X) \quad (3)$$

where α is the surface albedo and X represents other atmospheric variables that influence TOA SW flux. The deseasonalized anomaly in DARE is:

$$\delta DARE = \delta F(0, \alpha, X) - \delta F(AOD, \alpha, X) \quad (4)$$

Deseasonalized DARE anomaly and trend can be readily calculated from model outputs. However, it is more challenging to derive DARE from observations as pristine conditions cannot be observed directly. Assuming the contribution of other atmospheric variables to $\delta F(0, \alpha, X)$ is negligible, then $\delta DARE$ can be expressed as follows:

$$\delta DARE = \delta F(\alpha) - \delta F(AOD, \alpha, X) \quad (5)$$

Here $\delta F(\alpha)$ is the TOA flux anomaly due to surface albedo change, and can be derived from surface albedo anomaly and radiative kernel (Paulot et al., 2018). Over land, surface albedo is based on MCD43C1 described in section 3.3. Over ocean, surface albedo is specified from a look-up table based on the Coupled Ocean Atmosphere Radiation Transfer model (Jin et al., 2004; Rutan et al., 2009). We can derive $\delta F(AOD, \alpha, X)$ directly from EBAF data.

Figure 12 shows DARE trend (Wm^{-2} per decade) calculated from CERES EBAF and the surface albedo datasets, and from model simulations. Both observation-based and model simulations show increasing trends over eastern United States and the Atlantic Ocean (DARE becomes less negative), and decreasing trends over central Africa, India, and Indian Ocean (DARE becomes more negative). These DARE trends are consistent with the AOD trends over these regions. The increasing trend off the coast of eastern China in EBAF data is largely absent in the models, as is in the AOD trend. ECHAM-HAM model shows a strong increasing trend over eastern China, whereas trends from EBAF, GFDL, and Oslo are very muted. The observational-based DARE trend and AOD trends show good agreement over many regions, which adds confidence in both products.

Figure 13 further exams the deseasonalized anomalies of SW DARE over three land regions listed in Table 4. Over eastern China, DARE from EBAF shows a small increasing trend (Table 4), whereas model simulations show very different trends. The strong increasing trend from ECHAM-HAM is partly due to the large emission reduction introduced by SSP3-7.0 starting in 2015. Over India, both EBAF and model simulations show decreasing trends, with EBAF shows the sharpest decline. This could be partly due to the decreasing trend in surface albedo over India that results in larger radiative efficiency. Over eastern USA, anomalies and trends from EBAF and model simulations show excellent agreement.

7 Conclusions

AeroCom models have played an essential role in advancing our understanding of DARF, though large diversity still exists among the models. To improve the model performance and to understand the root causes of the large diversity among them, the models have been evaluated against in-site and satellite observations. To date, evaluations have been mainly focusing on aerosol optical depth. In this study, we evaluate TOA clear-sky reflected SW fluxes from the AeroCom models against the SW fluxes from CERES EBAF. Additionally, AODs and land surface albedo from AeroCom models are also evaluated against satellite retrievals in order to explain the SW flux biases.

To quantify how much the SW flux biases can be explained by the biases in AOD and land surface albedo, we use the radiative kernels of AOD and land surface albedo

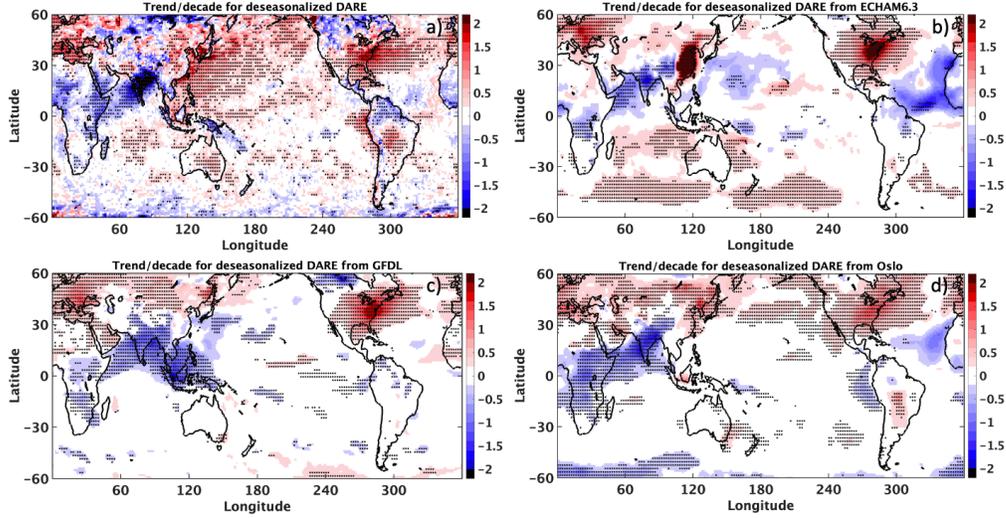


Figure 12. TOA SW direct aerosol radiative effect trends (Wm^{-2} per decade) calculated using CERES EBAF (a) and using ECHAM-HAM (b), GFDL (c), and Oslo (d) model simulations. Data from July 2002 to December 2017 are used here.

Table 4. Aerosol direct radiative effect trends (Wm^{-2} per decade) over China, India, and USA. Only land areas in the indicated latitude and longitude ranges are included in the regional trend calculation.

| | China (25-40°N, 110-120°) | India (7-22°N, 70-90°) | USA (30-42°N, 265-283°) |
|-------|------------------------------|---------------------------|----------------------------|
| EBAF | 0.4 ± 0.3 | -1.9 ± 0.3 | 1.2 ± 0.3 |
| ECHAM | 1.8 ± 0.8 | -0.7 ± 0.3 | 1.6 ± 0.3 |
| GFDL | 0.0 ± 0.3 | -0.8 ± 0.2 | 1.3 ± 0.2 |
| USA | 0.2 ± 0.2 | -1.1 ± 0.3 | 1.0 ± 0.2 |

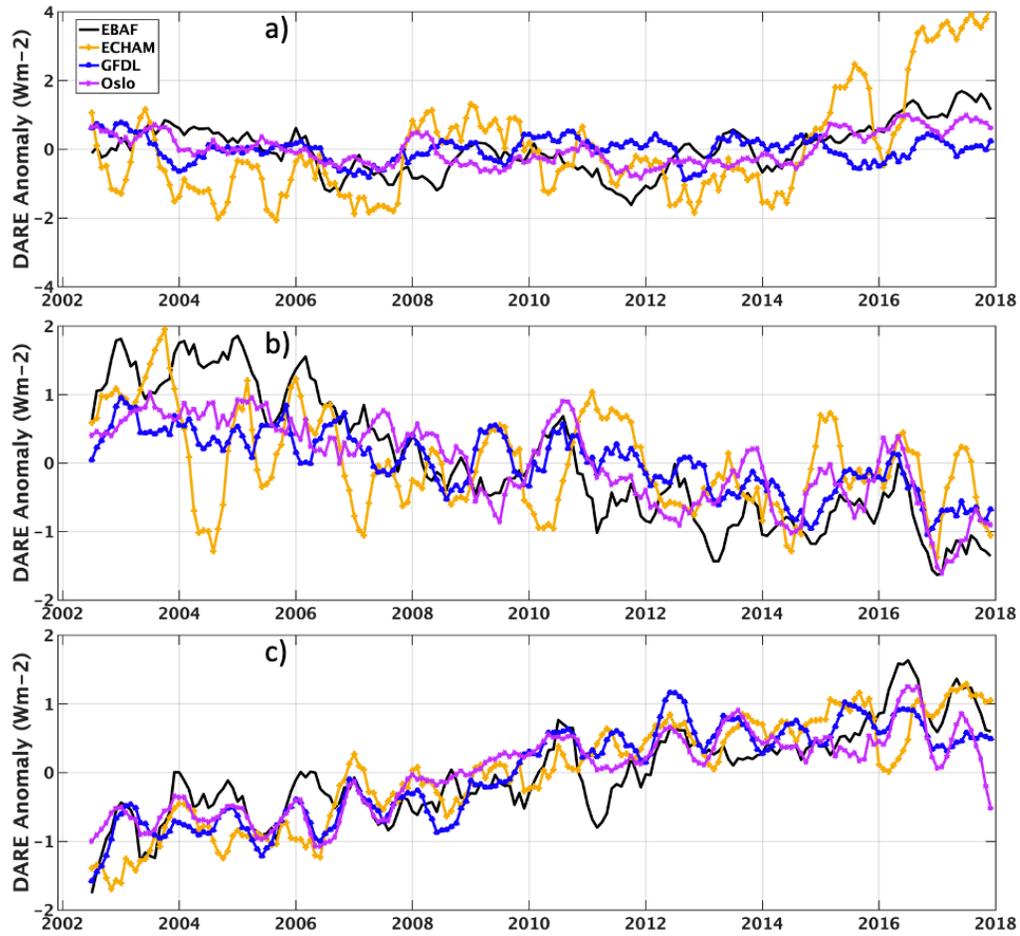


Figure 13. Deseasonalized anomalies (Wm^{-2}) of TOA SW direct aerosol radiative effect over eastern China (a), India (b), and eastern USA (c) calculated using CERES EBAF (black lines), ECHAM-HAM (orange line), GFDL (blue lines), and Oslo (purple) model simulations. Data from July 2002 to December 2017 are used here.

483 developed from MERRA-2 reanalysis to attribute their contributions to SW flux biases.
 484 Over ocean, the 60S-60N mean TOA SW flux bias in MMM is reduced by 25% after ac-
 485 counting for the contribution from AOD bias. Over land, the 60S-60N mean TOA SW
 486 flux bias in MMM is reduced by 70% after accounting for the contribution from biases
 487 in AOD and land surface albedo. Furthermore, the spatial patterns of the flux bias de-
 488 rived from the radiative kernels are very similar to those between models and CERES
 489 EBAF, with the correlation coefficient of 0.6 over ocean and 0.76 over land for MMM
 490 using data of 2010. The correlation coefficients for all models considered in this study
 491 are also high, ranging from 0.52 to 0.78 over ocean and from 0.78 to 0.95 over land. The
 492 high correlation indicates that most of the SW flux biases can be explained by the bi-
 493 ases in AOD and surface albedo between models and observations. Given that the CERES
 494 EBAF TOA SW flux is independently derived from MODIS AOD and land surface albedo,
 495 consistencies in their bias patterns when compared with model simulations suggest that
 496 these features are robust. In addition, the regional patterns of flux bias are unique to
 497 each model, which point to the deficiency in each model in simulating the specific aerosols
 498 in different source regions.

499 The AOD and DARE trends from ECHAM-HAM, GFDL, and Oslo model are com-
 500 pared with the observation-based counterparts. All three models reproduce the decreas-
 501 ing trends in MODIS AOD over Europe, eastern United States and the Atlantic Ocean,
 502 and central South America, and the increasing trends over India, Indian Ocean, Arabian
 503 Peninsular, and Central Africa. The models fail to reproduce the decreasing trend in AOD
 504 over eastern China and the adjacent oceanic regions due to limitations in the emission
 505 dataset.

506 Using independently derived satellite datasets (TOA reflected SW flux, AOD, and
 507 surface albedo) to assess the AeroCom models provide an opportunity to evaluate re-
 508 lated variables in a synergistic manner, thus provide an unambiguous assessment of the
 509 model performance and point to ways that can improve the aerosol simulations. Regional
 510 bias patterns in these variables, when they corroborate each other, offer a more convinc-
 511 ing assessment of the model performance and possibly the cause of the differences. Ra-
 512 diative kernels provide a convenient way to link the AOD and surface albedo biases to
 513 TOA SW flux biases, which can be used as a diagnostic tool for model development. All
 514 models should correct their land surface albedo by using satellite derived product as in-
 515 puts and constrain AODs using satellite retrievals. Implementing these changes will im-
 516 prove the global and regional SW flux agreement between models and satellite observa-
 517 tions, and reduce the diversity among the models.

518 Appendix A

519 A brief description of each model used in this paper is provided below.

520 A1 CAM5

521 The Community Atmosphere Model version 5 (CAM5) with the Aerosol Two-dimensional
 522 bin module for foRmation and Aging Simulation (ATRAS) uses a two-dimensional sec-
 523 tional aerosol representation with 12 particle size bins (from 1 to 10000 nm in dry di-
 524 ameter) and 8 black carbon mixing state bins (Matsui, 2017). The CAM5-ATRAS model
 525 considers the following atmospheric aerosol processes: emissions, new particle formation,
 526 condensation of sulfate, nitrate, and organic aerosols, coagulation, activation, aqueous-
 527 phase formation, dry and wet deposition, and aerosol-radiation-cloud interactions. Aerosol
 528 optical properties are calculated based on the Mie theory code (Bohren & Huffman, 1998),
 529 and radiative transfer for shortwave and longwave is calculated by the Rapid Radiative
 530 Transfer Method for GCMs (Iacono et al., 2008). CAM5-ATRAS aerosol simulations have
 531 been evaluated by surface, aircraft, and satellite observations in our previous studies (e.g.,
 532 Matsui & Mahowald, 2017; Matsui et al., 2018; Matsui & Moteki, 2020).

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A2 ECHAM6.3

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ECHAM6.3-HAM2.3 is the combination of the global climate model ECHAM6.3 (Mauritsen et al., 2019) and the Hamburg Aerosol Module (HAM2.3, Tegen et al., 2019). It uses the PSRad (Pincus & Stevens, 2013) two-stream radiative transfer scheme with 16 shortwave and 14 longwave bands. The Monte Carlo independent column approximation is used for cloud overlap. Aerosol radiative properties are pre-computed using Mie theory and read from a look-up table based on Mie size-parameter and volume weighted real and imaginary refractive index for seven aerosol modes containing up to five aerosol species (sulfate, black carbon, particulate organic matter, sea salt and dust) and aerosol water. Aerosol water uptake is based on kappa-Koehler theory (O’Donnell et al., 2011). In subtropical oceans where shallow convective clouds are prevalent, AOD is overestimated likely because precipitation from shallow convective clouds is only allowed if the clouds reach a certain thickness (Muench & Lohmann, 2020). Furthermore, black carbon and organic carbon concentrations are underestimated to some extent, which may be due to underestimated biomass burning emissions and cause to low AOD in biomass burning regions (Tegen et al., 2019).

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A3 GFDL

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The Geophysical Fluid Dynamics Laboratory Atmospheric Model version 4 (AM4) has cubed-sphere topology with 96×96 grid boxes per cube face (C96; approximately 100 km grid size) and 33 levels in the vertical, contains an aerosol bulk model that generates mass concentration of aerosol fields (sulphate, carbonaceous aerosols, sea salt and dust) from emissions and a “light” chemistry mechanism designed to support the aerosol model but with prescribed ozone and radicals (Zhao et al., 2018). Simulations up to the year 2014 are driven by time-varying boundary conditions, and natural and anthropogenic forcings developed in support of CMIP6 (Eyring et al., 2016), except for ship emission of SO₂ (black carbon ship emission is included). For the following simulated years, the anthropogenic emissions for 2014 are repeated. The dust emission is driven by the simulated winds from constant sources with their erodibility expressed as a function of surrounding topography (Ginoux et al., 2001). The sea salt emissions are based on Martensson et al. (2003) and Monahan et al. (1986) for fine and coarse mode particles, respectively. Aerosols are externally mixed except for black carbon, which is internally mixed with sulphate. The optical properties of the mixture are calculated by volume weighting of their refractive indices using a Mie code. The GFDL-AM4-met2010 (GFDLm) and GFDL-AM4-fSST (GFDLf) models are run with observed sea surface temperature and sea-ice distribution. In addition for GFDLm, the wind components are nudged, with a 6-hour relaxation time, towards the NCEP-NCAR re-analysis (Kalnay et al., 1996). The diagnostics are projected from the C96 cubed-sphere to equally spaced 1° latitude and 1.25° longitude grid using first order conservative method. In GFDL model, the aerosol effect is estimated by calling the radiative transfer scheme twice, with and without aerosols in the absence of clouds. The radiative time step is 1 hour for shortwave and 3 hour for longwave. The shortwave code is an update of the 18 band formulation of Freidenreich and Ramaswamy (2005). These updates are described in detail by Zhao et al. (2018). They are related to H₂O, CO₂, and O₂ formulations, and shortwave absorption by CH₄ and N₂O. In addition, the effects of the shortwave water self continuum and the O₂ and N₂ continua have been updated.

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A4 GISS

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The GISS model hosts two aerosol schemes, the GISS One-Moment Aerosol (OMA) and the Multiconfiguration Aerosol TRacker of mIXing state (MATRIX) models use the same aerosol emissions. Sea salt, dimethyl sulfide, isoprene, and dust emission fluxes are calculated interactively. Anthropogenic dust sources are not represented in ModelE2.1. Dust emissions vary spatially and temporally only with the evolution of climate variables

584 like wind speed and soil moisture (Miller et al., 2006). OMA is a mass-based scheme in-
 585 cluding sea salt, dust, sulfate, nitrate, ammonium, carbonaceous aerosols (BC and OC)
 586 aerosols (Tsigaridis et al., 2013). Aerosols are externally mixed and assumed to have a
 587 prescribed constant size distribution. Aerosol hydration in OMA is calculated in the ra-
 588 diation code following Tang and Munkelwitz (1994). MATRIX (S. Bauer et al., 2008)
 589 is an aerosol microphysics scheme that tracks aerosol mixing state, based on the quadra-
 590 ture method of moments, in which the amount of water in aerosol is calculated with the
 591 aerosol thermodynamics module EQSAM (Metzger et al., 2002), using the phase state
 592 of an ammonia-sulfate-nitrate-water inorganic aerosol (OA) in thermodynamic equilib-
 593 rium for metastable aerosols, except for sea salt where the Lewis parameterization is used
 594 (Lewis & Schwartz, 2013). As such, hygroscopic swelling of aerosol is already considered
 595 and does not need to be recalculated during the radiative calculations.

596 **A5 INCA**

597 INCA (INteraction with Chemistry and Aerosols) is a chemistry-aerosol model cou-
 598 pled to a land surface and a dynamical model. INCA simulates dust, sea salt, black car-
 599 bon (BC), NO₃, SO₄, SO₂, and organic aerosol (OA) with a combination of accumula-
 600 tion, coarse, and super-coarse modes, as well as soluble and insoluble components (Schulz
 601 et al., 2009). Because of the simplified chemistry scheme, DMS emissions are prescribed
 602 and not interactively calculated, and the secondary organic aerosols are not simulated
 603 therefore this specific run is underestimating the OA. In the current version BC solu-
 604 ble mode is internally mixed with sulphate (R. Wang et al., 2016), for which the refrac-
 605 tive index is estimated using the Maxwell-Garnett method, improving the accuracy of
 606 the BC optical absorption properties.

607 The radiative transfer model for the calculations with aerosols relies on the RRTM
 608 model as implemented by the European Centre for Medium-Range Weather Forecasts,
 609 a model that we used for SW and LW calculations. The number of spectral bands used
 610 for aerosols is 6 for SW and 16 for LW spectrum. The spectral dependence of optical prop-
 611 erties of each aerosol species has been estimated with Mie theory of spherical particles
 612 with log-normal distribution.

613 **A6 MIROC-SPRINTARS**

614 An aerosol climate model, Spectral Radiation Transport Model for Aerosol Species
 615 (SPRINTARS, Takemura et al., 2005, 2009), is incorporated into a coupled atmosphere-
 616 ocean general circulation model, MIROC6 (Tatebe et al., 2019). The horizontal and ver-
 617 tical resolutions are T213 ($\sim 0.5625^\circ \times 0.5625^\circ$ in longitude and latitude) and L56, re-
 618 spectively. SPRINTARS calculates the aerosol-radiation and aerosol-cloud interactions
 619 by coupling the radiation and cloud-precipitation schemes, respectively as well as aerosol
 620 transport processes. The radiative transfer scheme, mstrnX, adopt a two-stream discrete-
 621 ordinate method with a correlated k-distribution method (Sekiguchi & Nakajima, 2008).
 622 Scattering and absorption of solar and terrestrial radiation by aerosols are calculated as-
 623 suming the Mie theory with refractive indices of dry aerosols and water from d’Almeida
 624 et al. (1991). The volume-weighted refractive indices are assumed for internally-mixed
 625 particles between black carbon and organic aerosols as well as aerosols and water.

626 **A7 Oslo**

627 The OsloCTM3 is a global, offline chemical transport model (CTM) driven by 3-
 628 hourly meteorological data from the European Centre for Medium Range Weather Fore-
 629 cast (ECMWF) Integrated Forecast System (IFS) model (Lund et al., 2018; Søvde et al.,
 630 2012). The model is run in a $2.25^\circ \times 2.25^\circ$ horizontal resolution, with 60 vertical lev-
 631 els (the uppermost centered at 0.1 hPa). The treatment of transport and scavenging, as
 632 well as individual aerosol modules, is described in detail in Lund et al. (2018) and ref-

633 erences therein. The aerosol optical properties in OsloCTM3 are described in Myhre et
 634 al. (2007) with some recent updates, where the BC mass absorption coefficient (MAC)
 635 is following the formula in Zanatta et al. (2016) and a weak absorption implemented for
 636 OA (Lund et al., 2018).

637 Appendix B

638 Figure B1 shows the regional AOD biases of the AeroCom models relative to MISR
 639 retrievals (left panels) and the regional SW flux biases due to AOD biases (relative to
 640 MISR retrievals) and land surface albedo biases (relative to MODIS retrievals) calcu-
 641 lated from their radiative kernels (right panels) for April 2010. Many of the regional AOD
 642 bias patterns shown here are very similar to the AOD biases shown in Figure 9. The SW
 643 flux biases calculated from the radiative kernels using MISR AODs also resemble those
 644 shown in Figure 8. However, the biases over the tropical oceans are much muted when
 645 MISR AOD is used. The correlation coefficients between ΔF and $\Delta F_{AOD} + \Delta F_{\alpha}$ range
 646 from 0.79 to 0.94 over land, which is very similar to those derived when MODIS AOD
 647 is used. The correlation coefficients between ΔF and ΔF_{AOD} range from 0.26 to 0.63
 648 over ocean, not as high as when MODIS AOD is used. The reduced correlation over ocean
 649 is partly due to retrieval differences between MODIS and MISR, but largely due to MISR
 650 sampling issue as evident in the stripping features of the AOD bias plots.

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 653 CERES EBAF Ed4.1 data were obtained from <https://ceres.larc.nasa.gov/data/>. MODIS
 654 495 MYD08_M3.0_6.1 550 nm AOD Dark Target+Deep Blue Combined data were ob-
 655 tained from the Giovanni online data system, developed and maintained by the NASA
 656 GES DISC. The V6 MODIS Bidirectional Reflectance Distribution Func- tion (BRDF)/albedo
 657 products (MCD43C1) were obtained from the Land Processes Distributed Active Archive
 658 Center (LP DAAC) through <https://lpdaac.usgs.gov/products/mcd43c1v006/>.

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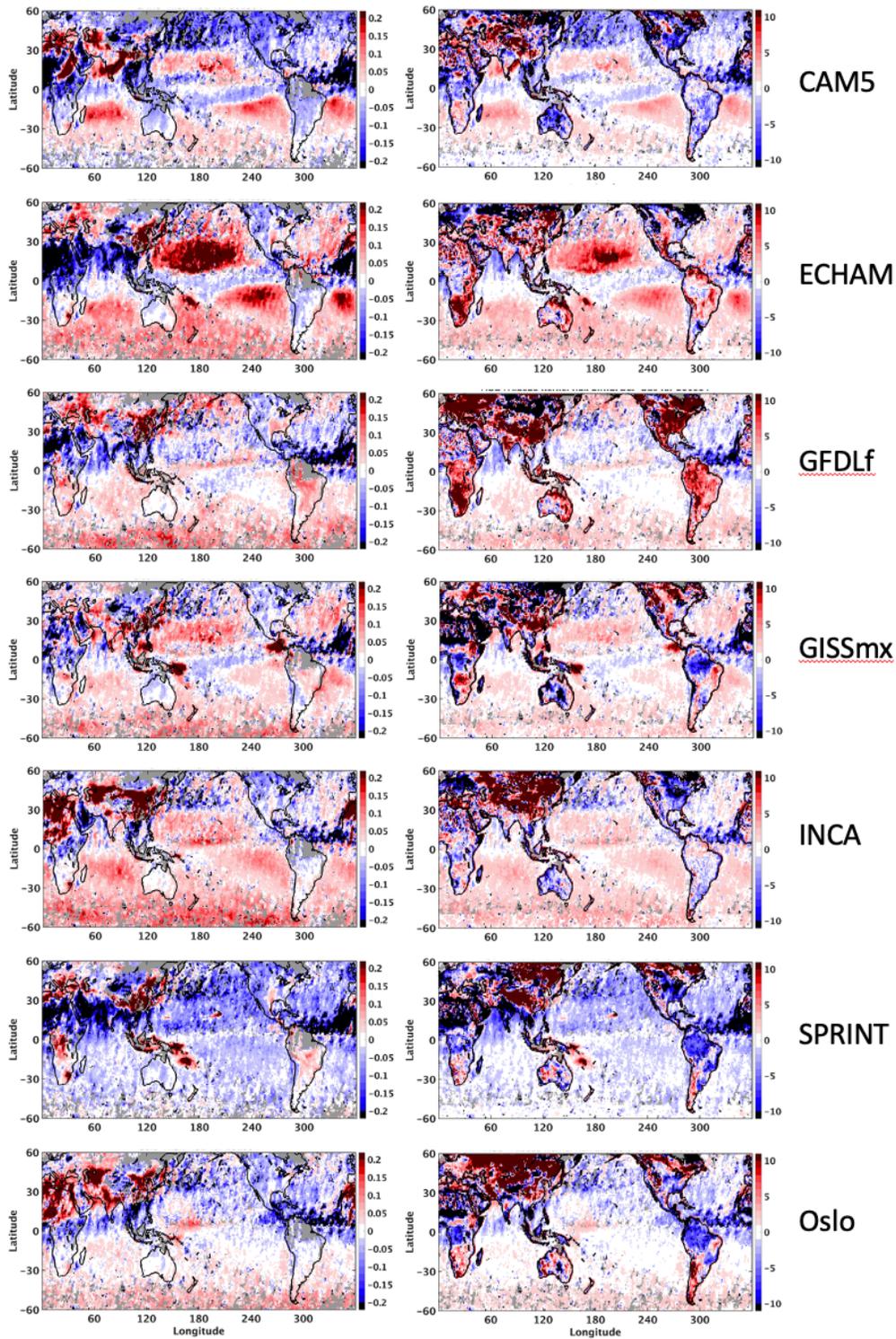


Figure B1. The monthly mean aerosol optical depth biases of AeroCom models relative to MISR retrieval (left panels), TOA SW flux biases due to AOD biases (models - MISR) and land surface albedo biases (models - MODIS) calculated from their respective radiative kernels (right panels) for April 2010.

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