

# 1 Droplet collection efficiencies inferred from satellite retrievals 2 constrain effective radiative forcing of aerosol-cloud interactions

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10

11 **Abstract.** Process-oriented observational constraints for the anthropogenic effective radiative forcing due to aerosol-  
12 cloud-interactions (ERF<sub>aci</sub>) are highly desirable because the uncertainty associated with ERF<sub>aci</sub> poses a significant  
13 challenge to climate prediction. The Contoured Frequency by Optical Depth Diagrams (CFODD) analysis supports  
14 evaluation of model representation of cloud liquid to rain conversion processes because the slope of a CFODD,  
15 generated from joint MODerate Resolution Imaging Spectroradiometer (MODIS)-CloudSat cloud retrievals, provides  
16 an estimate of cloud droplet collection efficiency in single-layer warm liquid clouds. Here we present an updated  
17 CFODD analysis as an observational constraint for the ERF<sub>aci</sub> due to warm rain processes and apply it to the U.S.  
18 Department of Energy's Energy Exascale Earth System Model version 2 (E3SMv2). A series of sensitivity  
19 experiments shows that E3SMv2 droplet collection efficiencies and ERF<sub>aci</sub> are highly sensitive to autoconversion,  
20 the rate of mass transfer from cloud liquid to rain, yielding a strong correlation between the CFODD slope and the  
21 shortwave component of ERF<sub>aci</sub> (ERF<sub>aci,sw</sub>; Pearson's R = -0.91). E3SMv2's CFODD slope ( $0.20 \pm 0.04$ ) is in  
22 agreement with observations ( $0.20 \pm 0.03$ ). The strong sensitivity of ERF<sub>aci,sw</sub> to the CFODD slope provides a useful  
23 constraint on highly uncertain warm rain processes, whereby ERF<sub>aci,sw</sub>, constrained by MODIS-CloudSat, is  
24 estimated by calculating the intercept of the linear association between the ERF<sub>aci,sw</sub> and the CFODD slopes, using  
25 the MODIS-CloudSat CFODD slope as a reference.

26

## 27 1 Introduction

28 Single-layer, low-level marine warm clouds cover 25% of the ocean surface (Charlson et al., 1987) and exert a strong  
29 cooling effect on climate due to their reflectivity (Hartmann et al., 1992; Hartmann and Short, 1980; Ramanathan et  
30 al., 1989). Aerosols modulate multiple radiative properties of low warm clouds, including droplet number

31 concentration ( $N_d$ ), liquid water path (LWP), geometric , cloud fraction, and lifetime, and their net impact on the cloud  
32 radiative forcing is the most uncertain component of the climate system (e.g., Stevens and Feingold, 2009;  
33 Christensen et al., 2020; Glassmeier et al., 2021). Though aerosols also exert a significant influence on ice and mixed-  
34 phase clouds, aerosol-cloud interactions (ACI) make their largest contribution to global radiative forcing via liquid  
35 water clouds (Bellouin et al., 2020).

36 In marine warm cloud regimes, an increase in aerosol concentrations typically leads to increasing  $N_d$ . Given constant  
37 condensed water content, enhanced aerosol concentrations increase cloud albedo due to higher concentrations of  
38 smaller cloud droplets through the so-called “Twomey effect” (Twomey, 1974). However, the cooling effect of  
39 increased  $N_d$  can be offset or enhanced by competing aerosol-mediated cloud properties such as cloud fraction and  
40 LWP. For example, increased numbers of smaller droplets can diminish cloud fraction by reducing cloud droplet  
41 sedimentation (Bretherton et al., 2007) and increasing cloud-top evaporation and dry air entrainment (Wang et al.,  
42 2003). On the other hand, aerosols can also increase cloud fraction and vertical extent by suppressing precipitation  
43 (Albrecht, 1989; Pincus and Baker, 1994). Christensen et al. (2020) demonstrated that the impact of aerosol on low-  
44 level cloud areal coverage depends on the stability of the atmosphere: in thermodynamically stable lower tropospheric  
45 conditions, increased aerosol results in increased cloud fraction, lifetime and  $N_d$ , whereas in unstable conditions,  
46 entrainment and evaporation offset Twomey effects, resulting in relatively smaller changes to cloud radiative  
47 properties.

48 Earth Systems Models (ESMs) are relied upon for estimating the global Effective Radiative Forcing of Aerosol-Cloud  
49 Interactions (ERFaci) due to the dearth of observations from the pre-industrial era. Yet ESM estimates are challenged  
50 by the lack of observational constraints on ERFaci and the cloud processes that modulate ERFaci, which must be  
51 parameterized due to the computational expense of explicitly resolving them. Mülmenstädt et al. (2020) proposed a  
52 renewed focus on process-oriented observational constraints as a solution to “equifinality”, whereby differing  
53 representations of cloud processes can reproduce observed state variables such as LWP and cloud fraction. The  
54 problem of equifinality renders many global long-term observations of state variables useless for constraining ERFaci  
55 on their own. Mülmenstädt et al. (2020) argues that constraints based on cloud process observations are thus highly  
56 desirable as an alternative approach to state variable-based constraints because mitigating bias in a cloud process  
57 representation will improve estimates of the response of the process to aerosols. Process-oriented constraints on

58 ERFaci are useful for quantifying the sensitivity of ERFaci to a specific process or constraining the component of  
59 ERFaci that is affected by a process, rather than for constraining ERFaci overall (Mülmenstädt and Feingold, 2018).  
60 Recent examples of process-based diagnostics include the Earth System Model Aerosol-Cloud Diagnostics Package  
61 (ESMAC Diags) (Tang et al., 2022; Tang et al., 2023), which supports evaluation of aerosol activation processes, and  
62 Varble et al. (2023) which demonstrated multiple model-observations comparison approaches that target processes  
63 affecting cloud albedo susceptibility using geostationary satellite data and surface-based observations. Christensen et  
64 al. (2023) applied ground-based measurements, satellite retrievals and meteorological reanalysis products in a  
65 Lagrangian framework to evaluate multiple aerosol-cloud processes in E3SM, including cloud condensation nuclei  
66 deposition via precipitation and the temporal response in  $N_d$  to aerosol perturbations.

67 In response to the demand for process-oriented constraints on warm liquid cloud processes, we present a constraint on  
68 the shortwave component of ERFaci ( $ERF_{aci_{sw}}$ ) due to autoconversion, a parameterization representing the transfer  
69 of liquid mass and number from the cloud to rain category, based on satellite cloud retrievals. For the past 12 years,  
70 prior studies have applied the Contoured Frequency by Optical Depth Diagrams (CFODD) analysis (Nakajima et al.  
71 2010; Suzuki et al. 2010) to evaluate model representation of warm rain processes because the slopes of CFODDs,  
72 generated from spaceborne radar reflectivity profiles (CloudSat) (e.g. Marchand et al., 2008) and cloud property  
73 retrievals from the Moderate Resolution Imaging Spectroradiometer (MODIS) (e.g. Platnick et al., 2017), provide an  
74 estimate of cloud droplet collection efficiency in warm liquid clouds (Suzuki et al. 2010). Here we demonstrate how  
75 an updated CFODD analysis can be applied to constrain ERFaci due to autoconversion using the U.S. Department of  
76 Energy's Energy Exascale Earth System Model version 2 (E3SMv2) and the relationship between CFODD slopes and  
77  $ERF_{aci_{sw}}$  in SLWCs.

78 To support the application of CFODD analysis as a constraint on  $ERF_{aci_{sw}}$ , we modified the Warm Rain Diagnostics  
79 (WRDs) subroutine (Michibata et al. 2019) that was recently implemented in the Cloud Feedback Model  
80 Intercomparison Project (CFMIP) Observations Simulator Package (COSPV2.0), a software package that supports  
81 climate model evaluation against satellite observations (Michibata et al., 2019; Swales et al., 2018). The WRDs  
82 support evaluation of model warm rain processes in single-layer warm liquid clouds (SLWCs) based on joint statistics  
83 from MODIS and CloudSat. The first diagnostic provides the fractional occurrence of SLWCs, classified as non-  
84 precipitating, drizzling, or raining clouds based on CloudSat column maximum radar reflectivity. The second

85 diagnostic is the CFODD, which is the probability density function (PDF) of radar reflectivity as a function of in-  
86 cloud optical depth (ICOD), where ICOD is the optical depth integrated from the cloud top downward to each vertical  
87 layer and represents an in-cloud vertical coordinate (Nakajima et al., 2010; Suzuki et al., 2010). The CFODD shows  
88 how vertical cloud microphysical structures transition from non-precipitating to precipitating as a function of cloud-  
89 top effective radius ( $R_e$ ), and the slope of reflectivity change with ICOD provides an estimate of droplet collection  
90 efficiency factor (Suzuki et al., 2010). Previous studies have used CFODDs to demonstrate that pollution decreases  
91 droplet collection efficiency, suppressing rainfall near the cloud base (Mangla et al., 2020; Michibata et al., 2014;  
92 Suzuki et al., 2013), and to evaluate model cloud liquid to rain conversion processes against satellite observations  
93 (Suzuki et al., 2015; Jing et al. 2019; Michibata and Suzuki, 2020). Takahashi et al. (2021) proposed an updated  
94 CFODD analysis in which  $R_e$  thresholds are defined by quartile distributions of SLWC samples rather than the  
95 traditional CFODD  $R_e$  thresholds to focus evaluation on warm rain process representation rather than the bias in  $R_e$   
96 distribution. Modifications to the WRDs in the present study include additional diagnostics that provide SLWC  
97 sampling statistics to illuminate how sample size affects CFODD results, the implementation of a CloudSat ground-  
98 clutter mask in the simulated WRDs and updates to SLWC selection criteria for better consistency between  
99 observations and satellite simulators. The updated CFODD analysis is demonstrated here as a constraint on the  
100 component of  $ER_{Faci_{SW}}$  that is affected by droplet collection efficiency due to autoconversion.

## 101 **2 Warm Rain Diagnostics Overview**

102 The WRDs and their implementation in COSPv2.0 were described in Michibata et al. (2019). The WRDs are designed  
103 to run online with the host model, accumulating time step statistics on warm rain cloud processes for subcolumns to  
104 mitigate the risk of data-processing bottlenecks associated with outputting large data volumes. COSPv2.0 generates  
105 ensembles of stochastic subcolumns from model gridbox-mean variables to emulate model subgrid variability and to  
106 resolve discrepancies in spatial resolution between observations and the model grid (Swales et al., 2018).

107 To generate observational reference data for model evaluation, Michibata et al. (2019) used the MODIS and CloudSat  
108 products 2B-TAU R04 (Polonsky, 2008) and 2B-GEOPROF R04 (Mace et al., 2007; Marchand et al., 2008),  
109 respectively, for SLWC detection between June 2006 and April 2011. The SLWC detection are described in  
110 Supplement Table S1 and include CloudSat reflectivity  $\geq -30$  dBZ, MODIS liquid COT  $> 0.3$ , and cloud top  
111 temperature  $\geq 273$  K. Model-simulated SLWCs are detected using COSPv2.0 simulated CloudSat reflectivity and

112 multiple MODIS cloud properties, including ice water path (IWP), liquid water path (LWP), cloud-top effective radius  
 113 ( $R_e$ ), and cloud optical thickness (COT) (Table S1). For the SLWC fractional occurrence (frequency) diagnostic,  
 114 SLWCs are binned by precipitation intensity according to the maximum column CloudSat reflectivity ( $Z_{max}$ ), where  
 115 non-precipitating, drizzling and raining SLWCs correspond to  $Z_{max} < -15 \text{ dBZ}_e$ ,  $-15 \text{ dBZ}_e \leq Z_{max} < 0 \text{ dBZ}_e$ ,  
 116 and  $Z_{max} \geq 0 \text{ dBZ}_e$ , respectively. The SLWC fractional occurrence diagnostic features frequency of each  
 117 precipitation type relative to the total SLWC population.

118 To support evaluation of liquid cloud collection efficiencies and cloud to rain transition processes, CFODDs are  
 119 constructed from the PDFs of CloudSat reflectivity profiles binned by ICOD. ICOD ( $\tau_d$ ) is parameterized as a function  
 120 of MODIS COT ( $\tau_c$ ) by invoking the adiabatic condensation growth model to vertically slice the column COT into  
 121 each layer (Suzuki et al., 2010). The relationship between  $\tau_d$  and  $\tau_c$  is as follows:

$$122 \quad \tau_d(h) = \tau_c \left\{ 1 - \left( \frac{h}{H} \right)^{5/3} \right\} \quad (1)$$

123 where  $h$  is height and  $H$  is the geometric height of the cloud. The detailed derivation of the ICOD coordinate is  
 124 provided in Suzuki et al. (2010). The slope of the resulting 2D-PDF diagnostic is modulated by droplet collection  
 125 efficiency, with steeper slope implying higher efficiency. The CFODD shows where, with ICOD on the y-axis as a  
 126 vertical coordinate, the droplet collection efficiency increases, and where the transition from non-precipitating to  
 127 drizzling and raining occurs, using the radar reflectivity as a proxy for the precipitation rate as described above (e.g.,  
 128 Muhlbaeuer et al., 2014). CFODDs are also typically binned by  $R_e$  to reveal how droplet collection efficiency changes  
 129 with droplet size (Suzuki et al., 2010; Takahashi et al., 2021; Jing et al., 2017).

130 In this study, CFODD slopes are estimated using RANdom SAMple Consensus (RANSAC) robust linear regression  
 131 (Fischler et al., 1987). RANSAC was chosen for performing linear regression due to the right-skewed distribution of  
 132 CFODD datasets. The regression was applied to the MODIS-CloudSat profiles and E3SMv2 output at  $4 \leq \text{ICOD} \leq 20$   
 133 and  $Z < 20 \text{ dBZ}$ . For E3SMv2 output, the regression was applied to approximated source CloudSat reflectivity and  
 134 ICOD data that was estimated from time-mean CFODD frequencies. The reflectivity and ICOD thresholds were  
 135 chosen to reduce the effect of the Mie scattering regime where the radar reflectivity can be saturated and to restrict  
 136 analysis to profiles where the uncertainty of MODIS COT retrievals is lower as error can be higher in optically thin  
 137 liquid clouds (e.g.,  $\text{COT} < 4$ ) (Platnick et al., 2017). The uncertainty in the RANSAC slope calculation is estimated

138 by “bootstrapping”, repeatedly performing RANSAC regressions on 1000 random subsamples of 80% the CFODD  
139 dataset to generate a distribution of slope estimates. The 1-sigma error and 95% confidence intervals were calculated  
140 from this distribution. The residual threshold applied for RANSAC outlier detection was 0.1 and  $0.5 \times$  median absolute  
141 error (MAE) for MODIS-CloudSat and E3SMv2, respectively. Data points with MAE exceeding the residual threshold  
142 are excluded from the linear regression in RANSAC.

## 143 **2.1 E3SMv2**

144 Several updates to the WRDs are described in Sect. 2.2, the impacts of which are demonstrated through an application  
145 of the updated WRDs to the U.S. Department of Energy’s Energy Exascale Earth System Model v2 (E3SMv2). The  
146 atmosphere component of the model, E3SM Atmosphere Model v2 (EAMv2), is described in detail in Golaz et al.  
147 (2022). Like its predecessor EAMv1, EAMv2 predicts stratiform and shallow cumulus cloud macrophysics through  
148 the Cloud Layers Unified by Binormals (CLUBB) parameterization, which unifies the treatment of planetary boundary  
149 layer turbulence, shallow convection, and cloud macrophysics through a higher-order turbulence closure scheme  
150 (Bogenschutz et al., 2013; J. C. Golaz et al., 2002; Larson, 2017; Larson & Golaz, 2005). CLUBB diagnoses cloud  
151 fraction and cloud liquid water from a joint double-Gaussian PDF. Ice and liquid cloud fractions in CLUBB are  
152 analytically diagnosed by integrating saturated proportions of the joint PDF (Guo et al. 2015).

153 Cloud microphysics is represented with the “Morrison and Gettelman version 2” (MG2) scheme (Gettelman and  
154 Morrison, 2015). MG2 prognoses the mass mixing ratios and number concentrations of cloud liquid, ice and  
155 precipitation hydrometeors. The coupled MG2 cloud microphysics and CLUBB higher-order turbulence  
156 parametrization explicitly provides values for hydrometer mass and number mixing ratios as well as cloud fraction.  
157 Deep convection is represented by the Zhang and McFarlane (1995) (ZM) scheme. As convective cloud fraction is  
158 not parameterized in the mass-flux based ZM scheme, it is diagnosed from the cloud mass flux for cloud radiation  
159 calculation (Hack et al., 1993). The total cloud fraction in EAMv2 combines CLUBB, deep convective cloud fractions  
160 and ice cloud fraction following (Park et al., 2014). The four-mode version of the Modal Aerosol Module (MAM4) is  
161 used to predict aerosol properties and processes (Liu et al., 2012, 2016; H. Wang et al., 2020).

162 EAMv2 runs on 72 vertical atmospheric levels with a top at 0.1h Pa (Rasch et al., 2019; Xie et al., 2018). However,  
163 distinct from its predecessor EAMv1, EAMv2 has two separate parameterized physics and dynamics grids (Hannah  
164 et al., 2021), with average horizontal grid spacings of  $\sim 165$  km and  $\sim 110$  km, respectively.

165 A six-year E3SMv2 simulation with transient, present-day forcing was run between 2006 and 2011 with online  
166 COSPv2.0 for comparison with A-Train observations of SLWCs, allowing one additional year (2005) for model spin-  
167 up. To facilitate comparison with observations, large-scale winds were constrained via the “nudging” technique (Lin  
168 et al., 2016; Ma et al., 2014; Zhang et al., 2014), in which horizontal and vertical winds are relaxed toward the Modern  
169 Era-Retrospective Analysis for Research and Applications, Version 2 (MERRA2) reanalysis data (Gelaro et al., 2017)  
170 with a 6-hour time-scale. MERRA2 data are read in every 3 hours and linearly interpolated to model times. COSPv2.0  
171 is called at every time step (0.5 h) and run with 10 subcolumns. We observed little change in CFODD results for  
172 increased numbers of subcolumns of 20 to 50.

## 173 **2.2 COSPv2.0**

174 Cloud-observing instrument simulators support evaluation of model cloud representation by translating gridbox-mean  
175 model variables (e.g., cloud fraction, hydrometeor mass mixing ratio, precipitation) into quantities that are measured  
176 by a cloud sensor (e.g., reflectivity). COSPv2.0 includes multiple cloud-observing satellite simulators and has been  
177 used extensively to diagnose issues in model cloud representation (Cesana & Chepfer, 2012; Kay et al., 2016; Song  
178 et al., 2018a; Y. Zhang et al., 2010). Recently, M. Zhang et al. (2022) used the COSPv2.0 CALIPSO simulator to  
179 demonstrate that changes to the Wegener-Bergeron-Findeisen process in EAMv2 decreased an ice cloud fraction low  
180 bias in the Arctic compared to EAMv1 but did not correct excesses of supercooled liquid.

181 There are known limitations to COSPv2.0 that affect its application to E3SM for cloud representation evaluation. The  
182 subgrid distribution of cloud variables generated by COSPv2.0 does not match E3SM subgrid variability.  
183 Hydrometeor species are distributed homogeneously across the subcolumns generated by COSPv2.0 via the  
184 subcolumn generator SCOPS (Subcolumn Cloud Overlap Profile Sampler) (Klein and Jakob, 1999), such that the  
185 ensemble of subcolumns reproduces the gridbox cloud fraction but not the subgrid distribution of liquid and ice within  
186 the simulated clouds (Dewald, 2021). Song et al., (2018b) demonstrated that the default “homogeneous hydrometeor  
187 scheme” from SCOPS results in overestimation of radar reflectivity in warm liquid clouds, thus overestimating  
188 precipitating clouds since maximum column reflectivity is often used to distinguish precipitating clouds (as in the  
189 WRDs). Errors in simulated satellite retrievals have also been attributed to SCOPS overlap assumptions (Hillman et  
190 al., 2018). Such a bias from SCOPS can result in unfair observational evaluation of a host model such as E3SMv2.  
191 Inconsistencies in microphysical assumptions between the host model and COSP pose another challenge. While many

192 microphysical assumptions in COSPv2.0 can be configured for agreement with E3SMv2 microphysics (MG2), some  
193 inconsistencies remain, including gamma distribution shape parameters for hydrometeor size distributions and  
194 hydrometeor vertical overlap assumptions (J. Wang et al., 2021). Next-generation E3SM development includes efforts  
195 to improve agreement in the subgrid variability and microphysical assumptions involved in forward simulating  
196 satellite retrievals. Other issues include the simplified treatment of satellite cloud detection in simulators. For example,  
197 the CloudSat Cloud Profiling Radar (CPR) cloud mask value threshold  $\geq 30$  is applied for cloud detection in the  
198 WRDs' A-Train analysis to indicate "good" or "strong" echo with high confidence detection (see next section and  
199 Supplement Table 1). The CPR cloud mask confidence levels consider signal-to-noise ratios, horizontal averaging,  
200 and spatial continuity (Marchand et al., 2008), but as this cloud mask is not available in COSPv2.0, CloudSat cloud  
201 detection is simulated by applying a reflectivity threshold  $-30 \leq Z_e \leq 20$  dBZ.

202 The WRDs rely on COSPv2.0 simulated MODIS and CloudSat retrievals. The WRDs in COSPv2.0 work as  
203 follows: First, COSPv2.0 takes in model atmospheric state and cloud variables including temperature, pressure,  
204 water vapor and hydrometeor mass mixing ratios, hydrometeor  $R_e$ , large-scale stratiform cloud fraction, convective  
205 cloud fraction and precipitation rate. The COSPv2.0 subcolumn generator SCOPS then produces subgrid  
206 distributions of clouds and precipitation for better comparison with smaller scale satellite pixel measurements.  
207 SCOPS subcolumns are homogenous, discrete samples generated such that a sufficiently large ensemble reproduces  
208 the model column profile of bulk cloud properties (Webb et al., 2001; Swales et al., 2018). SCOPS assigns each  
209 subcolumn a type (large-scale stratiform, convective or clear-sky) according to the host model's convective and  
210 large-scale stratiform cloud fraction. Cloud properties such as hydrometeor mass mixing ratios and  $R_e$  are distributed  
211 homogeneously across the subcolumns by cloud type (i.e., all stratiform cloud subcolumns are assigned the same  
212 stratiform ice and liquid mixing ratios as SCOPS only takes total convective and stratiform cloud fraction as input,  
213 and does not consider stratiform liquid and ice cloud fraction in its default configuration. "Maximum-random"  
214 cloud overlap is applied to subcolumns, consistent with the model parameterizations. The MODIS and CloudSat  
215 simulators apply simplified versions of their respective retrieval algorithms to each subcolumn, emulating MODIS  
216 retrievals of cloud properties, radar reflectivity and lidar backscatter, respectively. Gridbox-mean values are  
217 estimated from accumulated subcolumn statistics. The WRDs take as inputs gridbox-mean simulated MODIS  
218 retrievals of LWP, IWP, COT and  $R_e$ , as well as subcolumn CloudSat reflectivity profiles. The simulated MODIS  
219 COT represents in-cloud mean, as do the other MODIS variables used in the WRDs (e.g., LWP,  $R_e$ ). For example,



220 the MODIS liquid COT is computed by averaging the MODIS liquid COT in cloudy subcolumns across the grid-  
221 box. In E3SMv2-COSP, the same in-cloud stratiform COT value from the E3SMv2 radiative transfer module is  
222 distributed across all the subcolumns designated as stratiform cloud by SCOPS, as described above. These values  
223 and cloud/clear-sky designations for each subcolumn are used as input to the MODIS simulator to calculate the in-  
224 cloud MODIS liquid COT. Subcolumn-level SLWC reflectivity profiles are used as input to the WRDs, also with  
225 cloud properties homogenously distributed across the subcolumns of a given classification. Thus, in E3SM-COSP,  
226 the SLWC samples within a gridbox that have the same subcolumn classification (i.e., stratiform liquid or stratiform  
227 rain) will have the same simulated MODIS COT and CloudSat reflectivity profile.

228 Deviations from the original WRDs implemented in COSPv2.0 (Michibata et al., 2019b) include the application of  
229 the simulated CloudSat ground-clutter filter (available in COSPv2.0, but not applied to the WRDs previously) for  
230 better comparison with CloudSat retrievals, and the elimination of the “fracout” input used in the SLWC detection  
231 scheme from SCOPS. “Fracout” is the subcolumn-level cloud classification by vertical level from SCOPS, where each  
232 level of each subcolumn is designated as large-scale stratiform, convective, or clear-sky. This input was removed from  
233 the WRDs’ SLWC detection algorithm because of the lack of comparable cloud-type designation in the observations  
234 and CloudSat simulator and because “fracout” vertical cloud profiles were observed to deviate significantly from  
235 CloudSat reflectivity profiles (i.e., fracout indicates clear-sky where CloudSat reflectivity indicates cloud, or vice  
236 versa).

### 237 **2.3 Satellite data**

238 The MOD06-1KM-AUX R05 product (Platnick et al., 2017), which provides MODIS collection 6 retrievals at 1 km  
239 resolution along the CloudSat footprint, supplied the 6 MODIS cloud retrievals required for the SLWC detection  
240 described in Suzuki et al. (2010): LWP, IWP,  $R_e$ , COT, cloud top pressure and cloud layer number. Standard MODIS  
241 products from the 2.1  $\mu\text{m}$  channel were used for  $R_e$ , consistent with the simulated MODIS  $R_e$  used in the WRDs.  
242 Atmospheric temperature profiles were obtained from ECMWF-AUX R05 (Partain and Cronk, 2017), which includes  
243 temperature profiles from the European Centre for Medium-Range Weather Forecast (ECMWF) model (Dee et al.,  
244 2011) interpolated to the CloudSat footprint. 2B-GEOPROF R05 provided the CloudSat reflectivity profiles, the Cloud  
245 Profiling Radar (CPR) cloud mask and echo top characterization at 1.8 km resolution (Marchand et al., 2008). The  
246 detection of SLWCs and CFODD analysis in the present study follows Suzuki et al. (2010) (see Supplement Table 1

247 for details) with one exception: a COT threshold was decreased from 15 to 0.3; this had a substantial impact on cloud  
 248 occurrence (Figure 1; described next) and is consistent with the COT threshold implemented in the COSPv2.0 WRDs.  
 249 The decreased COT threshold also increases the weight of optically thin SLWCs, as the linear regression is applied to  
 250 the CFODD source data directly (i.e., the ICOD and reflectivity profiles).

#### 251 **2.4 Autoconversion sensitivity experiments and ERFaci**

252 The autoconversion parameterization in E3SMv2 is a modified Khairoutdinov & Kogan (2000) scheme (hereafter,  
 253 KK2000), in which coefficients were updated in response to large uncertainties in different cloud regimes and to  
 254 improve fidelity in climate simulations. The KK2000 autoconversion scheme is  $\frac{\delta q_r}{\delta t_{auto}} = A Q_c^\alpha N_d^\beta$ , where  $q_r$  is  
 255 the rainwater mixing ratio,  $Q_c$  is the cloud water mixing ratio,  $N_d$  is the cloud droplet number concentration, and  $A$ ,  $\alpha$   
 256 and  $\beta$  are the modified coefficients.

257 To develop a constraint on the ERFaci due to autoconversion, we performed multiple pairs of simulations featuring  
 258 preindustrial (PI) and present-day (PD) aerosol emissions. In each pair of simulations, one of the three coefficients  
 259 ( $A$ ,  $\alpha$  or  $\beta$ ) was modified to its KK2000 value, a value reported by Wood (2005), a value from Kogan (2013) or a value  
 260 within a range bounded by the three studies. The Kogan (2013) coefficient values were derived from a large-eddy  
 261 simulation (LES) with bin resolved microphysics for cumulus clouds, whereas the focus of Wood (2005) and KK2000  
 262 was stratocumulus clouds from observational and LES perspectives, respectively. One additional experiment on the  
 263 KK2000 parameterization for the accretion rate was performed, the formulation of which is  $\frac{\delta q_r}{\delta t_{accre}} =$   
 264  $F_1 F_2 67 (Q_c Q_r)^{1.15} \rho^{-1.3}$ , where  $Q_r$  is the rain water mixing ratio,  $F_1$  represents subgrid  $Q_c$  variability,  $\rho$  is air density,  
 265 and  $F_2$  is an accretion rate enhancement factor.  $F_2$  was increased by a factor of  $\sim 3$  in the accretion sensitivity  
 266 experiment.  $F_2$  is considered a tunable parameter in E3SM (Ma et al., 2022). The experiment details are provided in  
 267 Table 1.

268 Table 1. KK2000 coefficient and accretion enhancement factor values applied in 12 sensitivity experiments. Dash (“-”  
 269 “) indicates the coefficient value was unchanged from the default E3SMv2 parameterization (equal to the “CNTL”  
 270 simulation value).

Name	A	$\alpha$	$\beta$	accre
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CNTL	3.05E4	3.19	-1.4	1.75
alpha01	-	4.22	-	-
beta01	-	-	-1.0	-
acoef100x	3.05E6	-	-	-
alpha02	-	2.47	-	-
acoef0.05x	1.35E3	-	-	-
alpha03	-	3.00	-	-
beta03	-	-	-1.79	-
beta04	-	-	-3.01	-
acoef10x	3.05E5	-	-	-
acoef5x	1.53E5	-	-	-
acoef50x	1.53E6	-	-	-
accre01	-	-	-	5

271

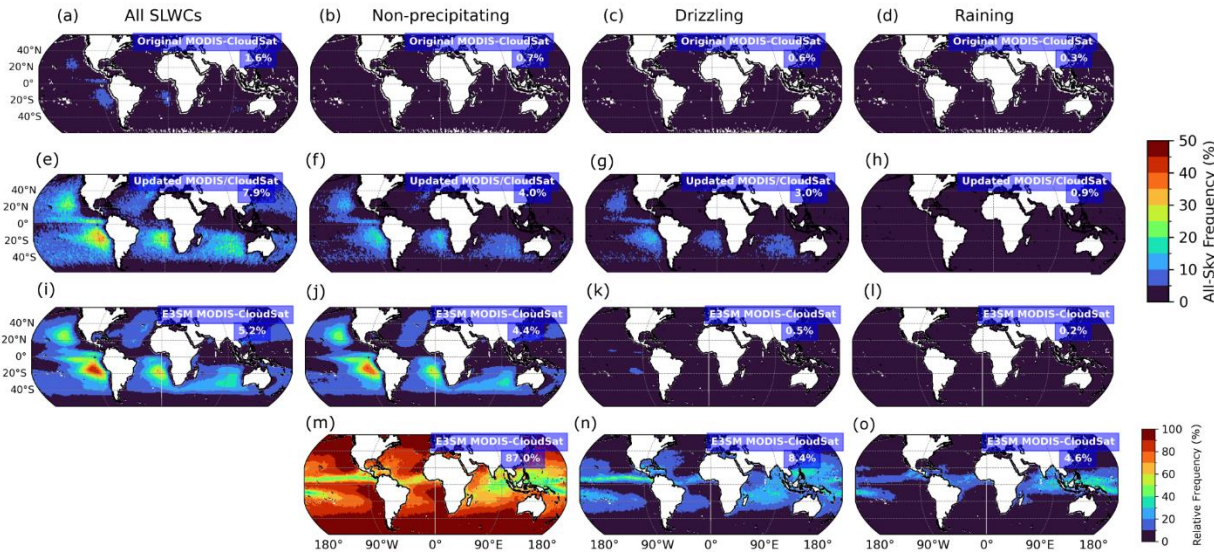
272 ERFaci for each pair of simulations was calculated following the Ghan (2013) method, where  $ERFaci = \Delta(F_{clean} -$   
273  $F_{clear, clean})$ .  $F_{clean}$  is the radiative flux at the top-of-atmosphere (TOA) neglecting the absorption and scattering of  
274 aerosols, and  $F_{clear, clean}$  is the radiative flux at the TOA neglecting both clouds and the absorption and scattering of  
275 aerosols. The  $\Delta$  indicates the PD – PI difference. While the PD-PI approach is a common strategy for estimating  
276 ERFaci, Christensen et al. (2023) demonstrated that it may yield a different estimate than the PD approach, where  
277 components of ERFaci (LWP adjustment,  $N_d$  adjustment, cloud fraction adjustment) are estimated by regressions of  
278 cloud properties multiplied by the anthropogenic aerosol fraction. We calculate ERFaci for SLWCs only, binned by  
279 the MODIS  $R_e$  range corresponding to the CFODD analysis.

280 A constraint on  $ERFaci_{sw}$  was calculated from the linear regression between E3SMv2 CFODD slopes and  $ERFaci_{sw}$ ,  
281 using the MODIS-CloudSat CFODD slope as a reference. A 95% confidence interval for the linear fit was estimated  
282 by bootstrapping the linear regression within the uncertainty of the CFODD slopes. CFODD slope values were  
283 randomly sampled 1000 times within their 1-sigma error and repeatedly regressed with  $ERFaci_{sw}$ . The original data  
284 (i.e., RANSAC CFODD slope values and corresponding  $ERFaci_{sw}$  values) were additionally resampled with

285 replacement to generate a distribution of coefficients for the ordinary least squares (OLS) regression. The 95%  
 286 confidence interval for the linear fit was then calculated from the combined linear regression coefficient distributions  
 287 to reflect uncertainty from both the OLS fit and the CFODD slopes.

### 288 3 Updates to MODIS and CloudSat SLWC analysis and reference data

289 The first diagnostic in the original WRDs featured relative frequencies of SLWCs by precipitation intensity in both  
 290 the A-Train reference data and the COSPv2.0 output (e.g., Fig. 1 m-o). We have updated this diagnostic with all-sky  
 291 frequencies and by decreasing the lower MODIS COT threshold from 15 to 0.3, for consistency with the WRDs  
 292 implemented in COSPv2.0 (Fig. 1 a-l). SLWCs featured in Fig. 1 and all following figures and analyses are ocean-  
 293 only due to higher uncertainties in MODIS retrievals over land (Platnick et al., 2017).



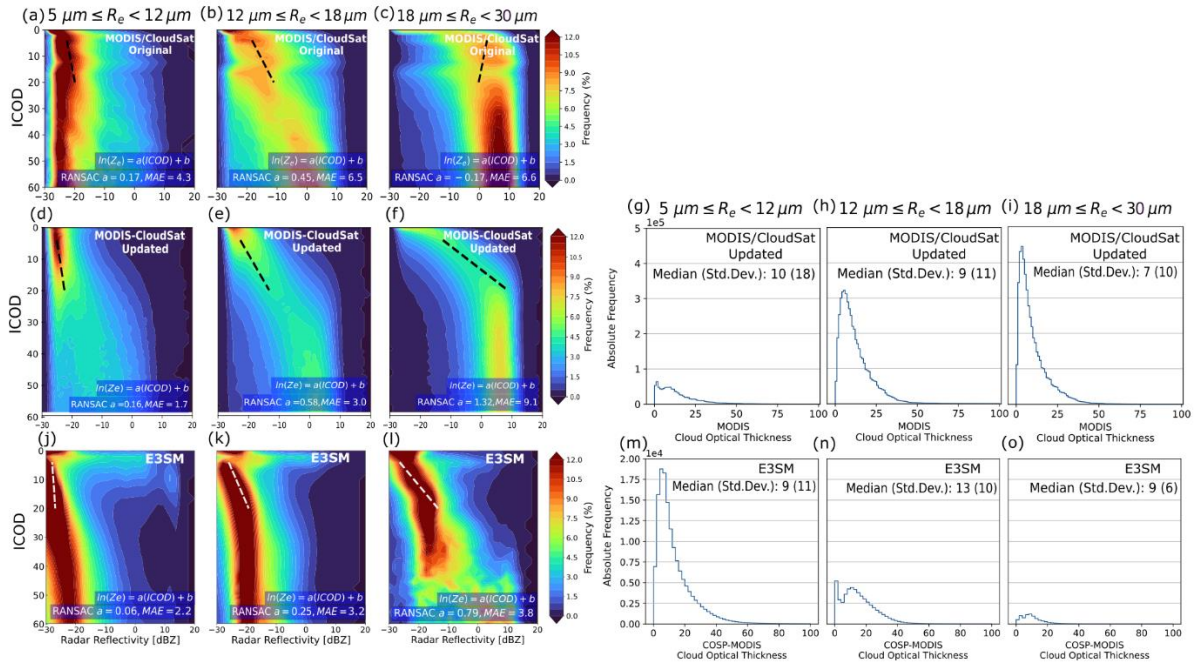
294  
 295 **Figure 1.** All-sky frequencies of total SLWCs June 2006 – Apr 2011, non-precipitating ( $Z_{max} < -15 \text{ dBZ}_e$ ), drizzling  
 296 ( $-15 \text{ dBZ}_e \leq Z_{max} < 0 \text{ dBZ}_e$ ) and raining ( $Z_{max} \geq 0 \text{ dBZ}_e$ ) ocean-only SLWCs according to original reference analysis of  
 297 MODIS and CloudSat observations (Michibata et al., 2019a, 2019b) (a-d), updated reference MODIS and CloudSat analysis (e-h)  
 298 and E3SMv2-COSPv2.0 (i-l). Figures m-o show frequencies of non-precipitating, drizzling and raining SLWCs relative to the total  
 299 SLWCs simulated by E3SMv2. Values in blue boxes indicate global ocean-only grid-weighted mean frequency. SLWCs were  
 300 undersampled in original reference A-Train analysis by a factor of  $\sim 5$ . Compared to the original reference A-Train data, the updated  
 301 analysis demonstrates that E3SM underrepresents rather than overrepresents total SLWC frequency and that precipitating SLWCs  
 302 are underrepresented by a factor of 6 compared to observations.

303 Figure 1 also shows that decreasing the lower MODIS COT threshold from 15 to 0.3 in the updated A-Train analysis  
304 (Sect. 2.3) increased total SLWC sampling by 5-fold (global ocean mean, see Sect. 2.3) compared to the original  
305 CFODD analysis in Michibata et al. (2019a) and Michibata et al. (2019b). The increase in SLWC sampling in the  
306 reference data affects multiple outcomes of the model evaluation in this case: E3SMv2 underrepresents, rather than  
307 overrepresents, total SLWCs, and the SLWCs that are missing from E3SMv2 are entirely the precipitating SLWC  
308 populations. The underrepresentation of precipitating SLWCs in E3SMv2-COSP indicates that any bias from SCOPS  
309 towards increased precipitation in warm liquid clouds is relatively minor (Sect. 2.2; Song et al. (2018)). Not all the  
310 differences between the original and updated reference data can be explained by the change in COT threshold,  
311 however, as we were unable to reproduce the original CFODD data with the updated satellite products used in this  
312 study. Fig. S1 and S2 show that increasing the lower COT threshold from 0.3 to 15 yields SLWC frequencies that are  
313 much closer to the original reference data (+25%) than the updated reference data, but significant differences remain  
314 in the CFODDs.

315 The effects of the increased SLWC sampling in the A-Train reference data also significantly affected the CFODDs  
316 and thus the comparison between A-Train and E3SMv2 droplet collection efficiencies. Figure 2 shows CloudSat  
317 reflectivity frequency binned by ICOD for the original A-Train reference data (Fig. 2 a-c), the updated A-Train  
318 reference data (d-f) and E3SMv2 (j-l), and RANSAC robust linear regression slopes at  $4 \leq \text{ICOD} \leq 20$ . In comparisons  
319 with various other linear regression techniques, we found that RANSAC best supported the comparison of CFODD  
320 slopes between E3SMv2 and observations because of the right-skewed distribution of CloudSat reflectivities at  $0 \leq$   
321  $\text{ICOD} \leq 20$  in E3SMv2 CFODDs (Figs. 2 j-l). RANSAC minimizes the median absolute error (MAE) and is less  
322 sensitive to strong outliers in the dimension of the predicted variable ( $Z_e$  in this case) compared to other linear  
323 regression techniques.

324 The updated A-Train CFODD distributions are significantly different than the original CFODD distributions (2D-  
325 Kolmogorov-Smirnov test,  $p \ll 0.05$ ). Compared to updated A-Train CFODDs, the E3SMv2 CFODDs show  
326 decreased droplet collection efficiencies and an increased range of reflectivities near the cloud top in all size bins,  
327 indicating that regardless of  $R_e$ , SLWCs are drizzling and raining near the cloud top with significantly higher frequency  
328 than SLWCs in observations but have decreased collection efficiency below cloud top compared to MODIS-CloudSat.

329



330

331 **Figure 2.** Contoured frequency by optical depth diagrams (CFODDs) for SLWCs June 2006 – April 2011 binned by MODIS cloud  
 332 top effective radius ( $R_e$ ) from original reference MODIS-CloudSat observations analysis (a-c), updated reference MODIS-CloudSat  
 333 observations analysis (d-f), and E3SMv2 (j-l). Random Sample Consensus (RANSAC) linear regressions were applied to the  
 334 CFODD at  $4 \leq \text{ICOD} \leq 20$  to estimate droplet collection efficiencies. RANSAC slopes and Median Absolute Error (MAE) values  
 335 are shown in blue boxes. Droplet collection efficiencies increase with MODIS  $R_e$  as expected, except for the largest  $R_e$  size bin in  
 336 the original reference data (Fig. s2c). Figs. g-i and m-o show absolute frequencies of SLWCs by MODIS COT, demonstrating that  
 337 E3SMv2 overrepresents SLWCs with small  $R_e$  relative to medium and large  $R_e$ , compared to observations.

338 The high reflectivities near the cloud top are pronounced in the subset of E3SMv2 SLWCs with  $4 < \text{MODIS COT} <$   
 339  $20$  (Fig. S3), indicating that the high reflectivity at low ICOD in Figs. 2 (j-l) is not just a product of a subset of  
 340 precipitating, optically thin SLWCs, but that layers near the cloud top in deeper SLWCs are also precipitating. also  
 341 contribute. The reflectivity profiles used to generate the CFODD come from the CloudSat simulator, which was not  
 342 modified for this study. Examples of simulated CloudSat reflectivity profiles in SLWCs with  $Z_e > 0$  dBZ near cloud  
 343 top are shown in Fig. S4. The source of this issue and its implications for E3SMv2 representation of liquid cloud  
 344 properties warrant further investigation that is beyond the scope of the present study.

345 Figure 2 shows absolute frequencies of SLWCs binned by MODIS COT in each CFODD  $R_e$  bin for the updated A-  
 346 Train analysis (Fig. 2 g-i) and E3SMv2 only (Fig. 2 m-o). Note, this information was unavailable in the original

347 reference data (Michibata et al., 2019a). Compared to COT distributions in the updated A-Train analysis, E3SMv2  
348 shows decreasing SLWC frequency with  $R_e$  and an underrepresentation of SLWCs with large  $R_e$ , which aligns with  
349 the underrepresentation of precipitating SLWCs in Fig. 1. Fig. 2o also shows that few SLWCs with large  $R_e$  have a  
350 COT > 20, indicating that the CFODD reflectivity profile in Fig. 2l at ICOD > 20 is comprised of few samples. The  
351 SLWC COT PDFs have been implemented in the WRDs to support the interpretation of CFODD statistics.

## 352 **4 Results and Discussion**

### 353 **4.1 CFODD analysis to constrain ERF<sub>aci</sub> due to warm rain processes**

354 To demonstrate the potential of the CFODD analysis described above for constraining ERF<sub>aci<sub>SW</sub></sub> due to warm rain  
355 processes, we performed 12 experiments featuring variations of E3SMv2's autoconversion and accretion  
356 parameterizations, computing ERF<sub>aci<sub>SW</sub></sub> for the SLWC samples represented in each CFODD and the corresponding  
357  $R_e$  bin (hereafter, "ERF<sub>aci<sub>SW</sub></sub><sub>SLWCs</sub>") following Ghan (2013; see Sect. 2.4). In each experiment, a single coefficient of  
358 either the KK2000 autoconversion or accretion parameterization was perturbed, each of which is treated as a tunable  
359 parameter in E3SMv2. The uncertain KK2000 coefficients, coupled with parameterization simplifications (e.g., bulk  
360 moments and assumed droplet size distributions), result in uncertainties and biases in the model representation of  
361 raindrop formation and growth. The experiments are described in Table 1, and the CFODDs for each experiment are  
362 shown in Fig. S5.

363 Figure 3 shows a strong negative correlation between E3SMv2 ERF<sub>aci<sub>SW</sub></sub><sub>SLWCs</sub> with "small" or "medium"  $R_e$  (i.e.,  $5$   
364  $\leq R_e < 18 \mu\text{m}$ ) and the corresponding combined  $5 \leq R_e < 18 \mu\text{m}$  CFODD slope (Pearson's  $R = -0.91$ ). SLWCs with  
365 large  $R_e$  ( $18 \leq R_e < 30 \mu\text{m}$ ) were excluded from the analysis in Fig. 3 because this population represents a negligible  
366 fraction of total SLWCs in E3SMv2 (see Fig. S6), resulting in poor sampling statistics and larger regression  
367 uncertainties. As CFODD slopes represent an estimate of droplet collection efficiency, Fig. 3 demonstrates that  
368 ERF<sub>aci<sub>SW</sub></sub> strengthens (increases in magnitude) with increasing droplet collection efficiency in E3SMv2 SLWCs with  
369  $R_e$  between 5 and 18  $\mu\text{m}$ . One possible physical explanation for the relationship between autoconversion, droplet  
370 collection efficiency, and ERF<sub>aci<sub>SW</sub></sub> is that increased autoconversion rates increase the susceptibility of clouds to  
371 precipitation suppression by aerosols. For a given optical depth, SLWCs with lower LWP and/or higher  $N_d$  will  
372 precipitate more when the autoconversion rate is increased. A larger population of precipitating SLWCs results in  
373 increased susceptibility to precipitation suppression by aerosols overall. When aerosols suppress precipitation (e.g.,

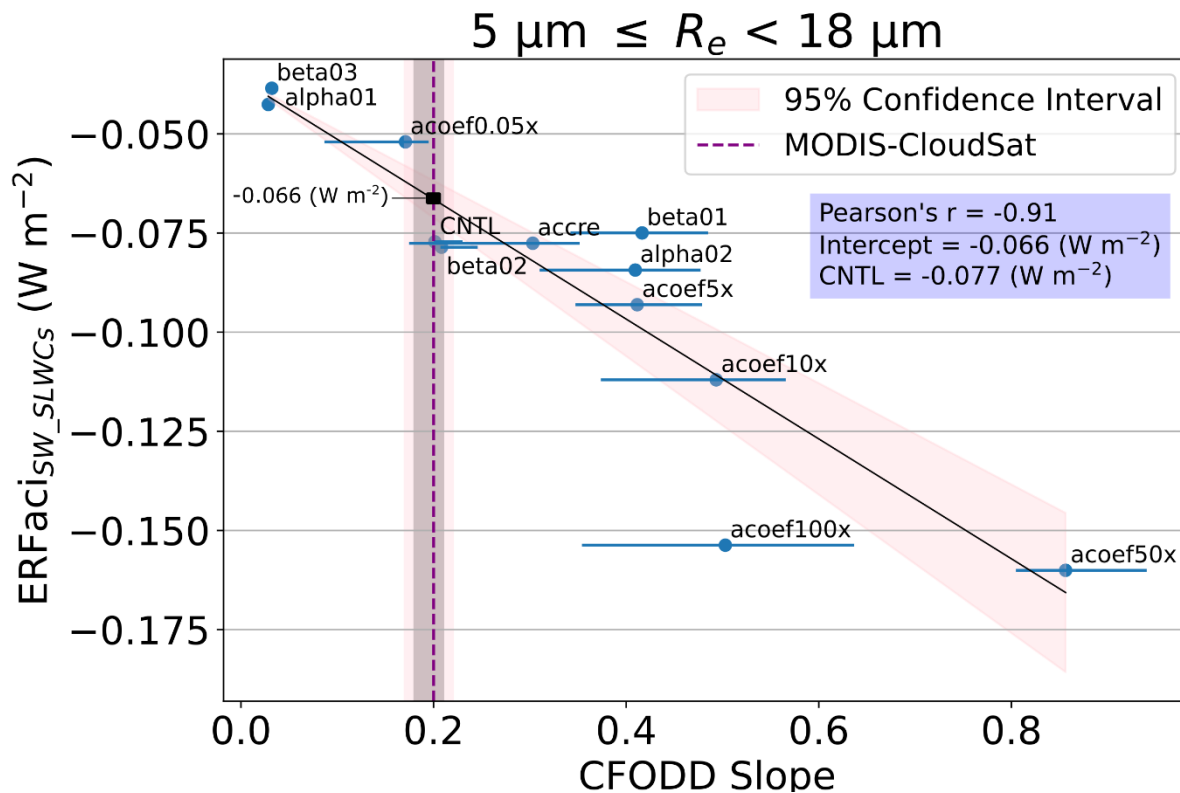
374 Suzuki et al., 2013), LWP and/or cloud fraction may be enhanced, resulting in brighter clouds and stronger  $ERFac_{iSW}$ .  
 375 The relationship between aerosols, LWP and cloud fraction (Albrecht, 1989), however, is highly uncertain, varies  
 376 regionally (Sato et al., 2018), and is influenced by processes that are buffered over multiple spatiotemporal scales  
 377 (Stevens and Feingold, 2009). Additionally, E3SMv2's CFODD slope ("CNTL" simulation) agrees with MODIS-  
 378 CloudSat within uncertainty, indicating that droplet collection efficiency is well-represented according to CFODD  
 379 analysis.

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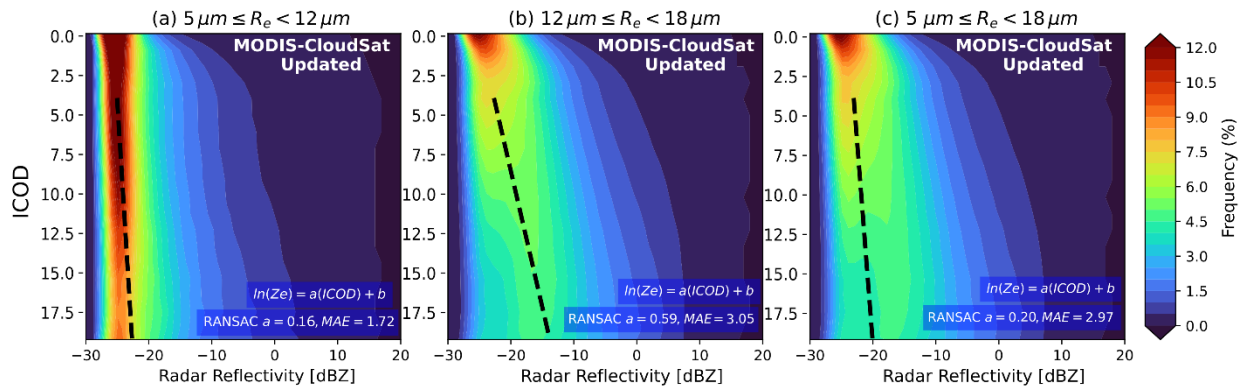
385 **Figure 3.** Linear regression between E3SMv2  $ERFac_{iSW\_SLWCs}$  and CFODD slopes, generated from SLWCs with MODIS  $R_e$

386 between  $5$  and  $18\ \mu\text{m}$ , in 12 PD autoconversion and accretion sensitivity experiments.  $ERFac_{iSW\_SLWCs}$  values reflect the SLWCs



387 represented in the corresponding CFODD (i.e., with  $R_e$  corresponding to the CFODD  $R_e$  bin). Results show a strong negative  
 388 correlation between E3SMv2  $ER_{Fac_{iSW\_SLWCs}}$  and CFODD slopes. We constrain the  $ER_{Fac_{iSW}}$  by predicting the  $ER_{Fac_{iSW\_SLWCs}}$   
 389 value at the reference MODIS-CloudSat  $5 \leq R_e < 18 \mu m$  CFODD slope (purple dashed line) from the linear regression (intercept  
 390 shown in blue box). Error bars represent 1-sigma error estimated from RANSAC-fit bootstrapping (Sect. 2). Grey and pink shaded  
 391 regions indicate the 68 and 95% confidence intervals for the MODIS-CloudSat CFODD slope, respectively. Labels indicate the  
 392 sensitivity experiment names (Table 1).

393 In Figure 3, we constrain  $ER_{Fac_{iSW}}$  due to autoconversion uncertainties using the linear regression between the  
 394 simulated CFODD slopes and  $ER_{Fac_{iSW\_SLWCs}}$ .  $ER_{Fac_{iSW}}$  and  $ER_{Fac_{iSW\_SLWCs}}$  values were calculated following Ghan  
 395 et al. (2013), which considers the difference in TOA radiative flux between the PD and PI experiments, neglecting  
 396 direct forcing of aerosols (see Sect. 2.4 for details). The constrained value of  $ER_{Fac_{iSW\_SLWCs}}$  is estimated at the  
 397 intercept of the linear relationship with the observed MODIS-CloudSat CFODD slope (Fig. 4). We find that the  
 398  $ER_{Fac_{iSW\_SLWCs}}$  predicted by the linear regression at the MODIS-CloudSat slope value ( $-0.066 W m^{-2} \pm 0.06 W m^{-2}$ )  
 399 approaches agreement with the  $ER_{Fac_{iSW\_SLWCs}}$  value predicted by the E3SMv2 CNTL simulation ( $-0.077 W m^{-2}$ ),  
 400 particularly considering the additional uncertainties imposed by the limited number of sensitivity experiments that are  
 401 not represented in the regression's 95% confidence interval. The agreement between the constrained and predicted  
 402 value of  $ER_{Fac_{iSW\_SLWCs}}$  indicates that the  $ER_{Fac_{iSW}}$  due to autoconversion is well-represented in E3SMv2 according  
 403 to CFODD analysis..

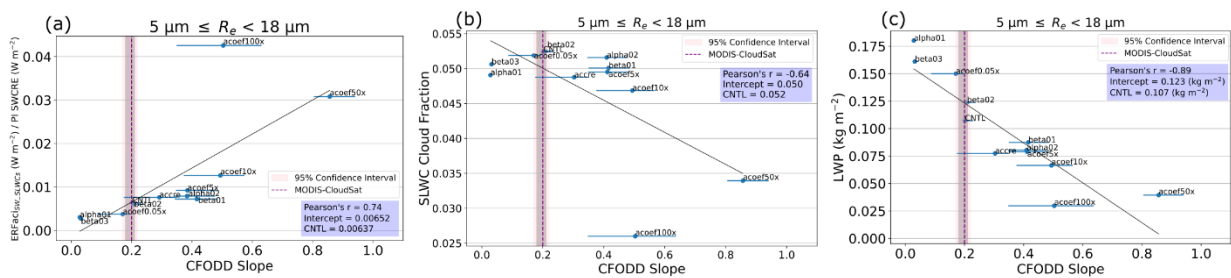


404  
 405 **Figure 4.** CFODDs for subset of SLWCs with max CloudSat reflectivity < 20 dBZ and COT < 20, June 2006 – April 2011, binned  
 406 by MODIS  $R_e$  from updated reference MODIS-CloudSat observations analysis (a-b), and with combined “small” and “medium”  
 407  $R_e$  SLWCs in (c). RANSAC linear regressions were applied to the CFODD at  $4 \leq ICOD \leq 20$  to estimate droplet collection  
 408 efficiencies. RANSAC slopes and Median Absolute Error (MAE) values are shown in blue boxes.

409 As  $ERF_{\text{Fac}_{\text{SW}}}$  is the result of many cloud processes, the updated CFODD analysis should be interpreted as a constraint  
410 on the component of  $ERF_{\text{Fac}_{\text{SW}}}$  that is modulated by droplet collection efficiency due to autoconversion. In other  
411 words, the updated CFODD analysis shows the change in  $ERF_{\text{Fac}_{\text{SW}}}$  one would expect if the bias in  $ERF_{\text{Fac}_{\text{SW}}}$  due to  
412 a specific process representation affecting droplet collection efficiency were eliminated. Base cloud processes that are  
413 independent of aerosol also contribute significantly to  $ERF_{\text{Fac}}$  estimates (Mülmenstädt et al., 2020). Autoconversion  
414 perturbations affect base cloud state (e.g., LWP, cloud fraction) and could, for example, cause stronger  $ERF_{\text{Fac}}$  by  
415 increasing cloud amount rather than increasing the impact of ACI on SW radiative forcing. Jing et al. (2019) evaluated  
416 different autoconversion parameterization schemes in an ESM using the CFODD analysis described in Michibata et  
417 al. (2019b) and found that the autoconversion scheme that yielded the best warm rain representation predicted a  
418 significantly stronger  $ERF_{\text{Fac}}$  that exceeded the uncertainty range of the IPCC AR5 and canceled out much of the  
419 warming trend of the last century. The conflict between process representation and  $ERF_{\text{Fac}}$  predictions in Jing et al.  
420 (2019) underscore a challenge with process-based constraints: improving the representation of a process can result in  
421 adverse outcomes to climate prediction due to compensating biases in the model. This challenge is particularly  
422 troublesome for constraints on processes like autoconversion that affect the base cloud state because decreasing  
423 autoconversion rates can increase total cloud amount, which can yield stronger  $ERF_{\text{Fac}}$ . Thus, a decreased  
424 autoconversion rate may improve precipitation outcomes in an ESM that presents the common “too frequent” warm  
425 rain bias (e.g., Stephens et al., 2010), yet cause improbably strong  $ERF_{\text{Fac}}$ . Our results show, however, that decreased  
426 autoconversion rates result in weaker  $ERF_{\text{Fac}_{\text{SW\_SLWCs}}}$  (Fig. 3), demonstrating that the base cloud state issue presented  
427 in prior studies of autoconversion is not a dominant factor contributing to the  $ERF_{\text{Fac}_{\text{SW}}}$  of warm rain processes in  
428 E3SMv2.

429 Figure 5a shows the linear relationship between  $ERF_{\text{Fac}_{\text{SW\_SLWCs}}}$  normalized by the PI SW Cloud Radiative Effect  
430 (SWCRE), which represents the fraction of  $ERF_{\text{Fac}}$  that is independent of base cloud state changes, and CFODD slope.  
431 The correlation coefficient in Fig. 5a (Pearson’s  $R = 0.74$ ) is decreased compared to Fig. 3 (Pearson’s  $R = -0.91$ ).  
432 However, comparing the negative correlations between CFODD slope and PI SLWC cloud fraction (Fig. 5b; Pearson’s  
433  $R = -0.64$ ) and LWP (Fig. 5c; Pearson’s  $R = -0.89$ ) with Fig. 3, the  $ERF_{\text{Fac}_{\text{SW\_SLWCs}}}$  increases in magnitude as LWP  
434 and cloud fraction decrease, further demonstrating that the contribution of base cloud state to  $ERF_{\text{Fac}_{\text{SW\_SLWCs}}}$  is  
435 relatively minor. The decreased correlation coefficient in Fig. 5a could also be influenced by poor sampling statistics  
436 in the “acoef100x” experiment. The acoef100x was the only one of six experiments involving perturbations of the “A”

437 coefficient in KK2000 (Table 1; Sect. 2.4) in which the CFODD slope did not increase with an increase in magnitude  
 438 of the “A” coefficient. Given the significant decrease in SLWC cloud fraction in this experiment compared to the  
 439 others (Fig. 5b, Table S2), the CFODD slope result may be affected by insufficient sample size, an additional  
 440 uncertainty of the CFODD linear regression that is not reflected in the bootstrapping-based uncertainty estimate (Sect.  
 441 2).



442  
 443 **Figure 5.** Linear regression between (a) E3SMv2 ERFaci<sub>SW\_SLWCs</sub> normalized by SWCRE, (b) SLWC cloud fraction, (c) SLWC  
 444 LWP and CFODD slopes in 12 PD autoconversion and accretion sensitivity experiments, calculated for SLWCs with MODIS  $R_e$   
 445 between 5 and 18  $\mu\text{m}$ . ERFaci<sub>SW\_SLWCs</sub> values reflect the SLWCs represented in the corresponding CFODD (i.e., with  $R_e$   
 446 corresponding to  $5 < R_e < 18 \mu\text{m}$ ). Error bars represent 1-sigma error estimated from RANSAC-fit bootstrapping (Sect. 2). Grey  
 447 and pink shaded regions indicate the 68 and 95% confidence intervals for the MODIS-CloudSat CFODD slope, respectively. Labels  
 448 indicate the sensitivity experiment names (Table 1).

449 While we derive a constraint for ERFaci<sub>SW</sub> using the combined small and medium  $R_e$  CFODDs, when the  $R_e$  subsets  
 450 are considered individually, they show distinct contributions to ERFaci<sub>SW\_SLWCs</sub>. Fig. S7 shows that SLWCs with small  
 451  $R_e$  have a negative ERFaci<sub>SW\_SLWCs</sub>, but that SLWCs in the medium and large  $R_e$  subsets have positive ERFaci<sub>SW\_SLWCs</sub>  
 452 values. This indicates that the dominant effect of aerosols on shortwave radiative forcing in the medium and large  
 453 SLWC subsets is decreased cloud fraction, which is reflected in the decreased SLWC sample sizes in the PD  
 454 simulations compared to PI (Fig. S8, S9). The negative linear relationship between ERFaci<sub>SW\_SLWCs</sub> and CFODD slope  
 455 in the medium and large  $R_e$  subsets indicates that increasing droplet collection efficiency partially counteracts the  
 456 decrease in cloud fraction due to aerosol. The small  $R_e$  SLWCs, however, show a positive correlation between  
 457 ERFaci<sub>SW</sub> and CFODD slope, indicating that ERFaci<sub>SW</sub> weakens as autoconversion rates increase, likely due to  
 458 decreased precipitation suppression susceptibility in this subset. The combined small and medium CFODD and  
 459 ERFaci<sub>SW\_SLWCs</sub>, therefore, represent the convolution of two populations with differing ERFaci<sub>SW</sub> sensitivities to  
 460 autoconversion perturbations. We chose to constrain ERFaci<sub>SW</sub> using the combined small and medium CFODD and

461  $ERF_{\text{Fac}_{\text{SW\_SLWCs}}}$  due the correlation performance and the dearth of large  $R_e$  SLWCs in E3SMv2. However, constraints  
462 for  $ERF_{\text{Fac}_{\text{SW}}}$  could potentially be derived for each individual  $R_e$  subset or various combinations thereof, depending  
463 on the distribution of SLWCs among the  $R_e$  size bins and their contribution to the host model's  $ERF_{\text{Fac}}$ . Considering  
464 that constrained  $ERF_{\text{Fac}_{\text{SW}}}$  increases in magnitude with increasing  $R_e$  in Fig. S7, that the shortwave component of  
465  $ERF_{\text{Fac}}$  is significantly larger than the longwave in CMIP6 models (Smith et al., 2020), and that E3SMv2's total  
466  $ERF_{\text{Fac}}$  ( $-1.50 \text{ W m}^{-2}$ ) is relatively strong compared to the IPCC AR6 'very likely' range ( $-1.0 \pm 0.7 \text{ W m}^{-2}$ ) (Forster  
467 et al., 2021), the underrepresentation of SLWCs with large  $R_e$  in E3SMv2 represents a compensating bias, without  
468 which the total  $ERF_{\text{Fac}}$  would be even stronger compared to IPCC AR6.

#### 469 **4.2 Limitations of CFODD-based constraint on $ERF_{\text{Fac}}$**

470 There are multiple limitations to the CFODD analysis that should be considered in its application as a constraint for  
471  $ERF_{\text{Fac}}$ . First, droplet collection is not explicitly represented in ESMs with bulk microphysical schemes such as  
472 E3SMv2, but is instead implicit in an amalgamation of process and drop size distribution parameterizations controlling  
473 the evolution of the cloud and precipitation. Delving into the impact of these individual processes on CFODD-based  
474 constraint of  $ERF_{\text{Fac}}$  is a good target of future work, while autoconversion modulation of  $ERF_{\text{Fac}}$  was the primary  
475 focus here. Furthermore, simulated radar reflectivity is highly sensitive to particle size distribution assumptions in the  
476 forward simulator (e.g., Bodas-Salcedo et al., 2011; J. Wang et al., 2021). The host model, therefore, could represent  
477 warm rain microphysical processes with high fidelity but still produce biased CFODD profiles when compared with  
478 observations. In COSPv2.0, the CloudSat simulator calculates size distributions from an assumed distribution (e.g.,  
479 log-normal, gamma, exponential) as well as mass-mixing ratios, precipitation fluxes, and gridbox-mean  $R_e$  from the  
480 host model. Default COSPv2.0 size distributions were used in this study: log-normal for large-scale stratiform and  
481 convective cloud liquid, and exponential for large-scale stratiform and convective cloud rain. The CFODD analysis  
482 itself is subject to multiple uncertainties, including the use of simple adiabatic and condensational growth assumptions  
483 to scale MODIS COT to ICOD. These assumptions result in a vertical distribution of optical depth, mass  
484 concentrations and particle size that may not reflect reality. For example, in the CFODD, particle size and mass  
485 concentration are assumed to monotonically increase with height, yet in the real cloud, particle sizes may decrease  
486 near the cloud top due to evaporation and entrainment (Suzuki et al., 2010). The uncertainties from assumed  
487 hydrometeor size distributions and CFODD construction should be carefully considered when using the CFODD to

488 evaluate model droplet collection efficiencies against observations and in the application as an ERFaci constraint.  
489 Simulated reflectivity biases affect the evaluation of the model CFODD slope against the observational CFODD slope  
490 and thus affect the estimation of ERFaci bias.

491 A few additional limitations on CFODD analysis are imposed by biases in E3SMv2 SLWC representation. The ERFaci  
492 constraint is restricted to the small and medium  $R_e$  CFODDs because of the underrepresentation of SLWCs with large  
493  $R_e$ . SLWCs with medium  $R_e$  are also underrepresented in E3SMv2, further limiting the CFODD analysis of E3SMv2  
494 ERFaci because process perturbations are limited to the extent that they do not significantly reduce the number of  
495 SLWCs with medium  $R_e$ . The high reflectivity near cloud top at  $ICOD < 4$  in E3SMv2 CFODDs presents another  
496 limitation. SLWCs with  $COT < 4$  represent a significant fraction of the SLWC population in both A-Train and  
497 E3SMv2 (Fig. 2), so including optically thin SLWCs in the linear regression would likely affect the CFODD slope  
498 and droplet collection efficiency estimates.

499 Despite these limitations and the uncertainty associated with estimates of droplet collection efficiency from simulated  
500 radar reflectivity, CFODD analysis offers a highly desired process-oriented constraint on ERFaci due to warm rain  
501 processes. In E3SMv2, the CFODD slope exhibits the expected behavior in response to autoconversion perturbations:  
502 slope increases with perturbations that increase the autoconversion rate and decreases with perturbations that decrease  
503 the autoconversion rate. Our results also show that the model  $ERFaci_{sw}$  is highly sensitive to the processes that the  
504 CFODD represents, enabling the constraint of  $ERFaci_{sw}$  against the CFODD slope derived from MODIS-CloudSat  
505 cloud retrievals. Prior studies have demonstrated that radar reflectivity biases can be partially mitigated by bringing  
506 the forward simulator into better agreement with the host model's microphysics parameterization and subgrid  
507 variability (Song et al., 2018b; J. Wang et al., 2021). Modified versions of COSP featuring improved consistency with  
508 E3SM are to be implemented in future E3SM model versions, which will decrease the uncertainties associated with  
509 CFODD analysis of E3SM.

## 510 **5 Summary**

511 In this study, we present an updated CFODD analysis and demonstrate how it can be applied to ESMs as a process-  
512 oriented constraint on ERFaci and find that E3SMv2's  $ERFaci_{sw}$  agrees with the MODIS-CloudSat constrained value  
513 within uncertainty. Demonstrated here as a constraint on the component of  $ERFaci_{sw}$  that is modulated by  
514 autoconversion, CFODD analysis represents a highly desirable constraint on a process, circumventing the equifinality

515 issue that bedevils atmospheric state variable-based approaches (Mülmenstädt et al., 2020). Limitations of CFODD-  
516 based constraint of ERFaci include the implicit representation of droplet collection efficiency in many ESMs,  
517 including E3SMv2, the sensitivity of simulated radar reflectivity to droplet size distribution representations and  
518 simplifying assumptions applied to construct the CFODD (e.g., adiabatic-condensational growth). While this study  
519 focuses on autoconversion, future studies should apply CFODD analysis to other microphysical processes that affect  
520 droplet collection efficiency (e.g., accretion, droplet breakup, evaporation) to generate additional ERFaci constraints.

521 Several updates to the WRDs package in COSPv2.0 were made to support the application of CFODD analysis to  
522 ESMs. In addition to the original WRDs diagnostics featuring relative frequencies of SLWCs by precipitation intensity  
523 and the CFODD by  $R_e$ , we have implemented additional diagnostics in the WRDs that include all-sky SLWC  
524 frequency maps and MODIS SLWC COT distributions for CFODD sampling statistics. Other updates include the  
525 estimation of CFODD slopes using Random Sample Consensus robust linear regression and changes to the SLWC  
526 detection schemes for better comparison between observations and satellite simulators.

527 In addition to the modifications of the WRDs described above, the MODIS and CloudSat observational reference data  
528 has been updated for consistency with COSPv2.0 SLWC detection. SLWC detection is increased 5-fold in the updated  
529 reference data. The increase in SLWC sampling also significantly affected the CFODD distributions and consequently,  
530 the A-Train reference droplet collection efficiency at large  $R_e$  ( $18 \mu\text{m} \leq R_e < 30 \mu\text{m}$ ). The updated WRDs showed that  
531 droplet collection efficiencies in E3SMv2 are decreased compared to observations and SLWCs with small MODIS  $R_e$   
532 ( $5 \mu\text{m} \geq R_e > 12 \mu\text{m}$ ) are overrepresented. The E3SMv2 CFODD results also show reflectivities exceeding 0 dBZ near  
533 cloud top at  $2 < \text{ICOD} < 4$  yet relatively low reflectivities at  $\text{ICOD} > 5$ . The unreasonably high reflectivities near cloud  
534 top may indicate artifacts due to inconsistencies between E3SMv2 outputs and COSPv2.0 inputs to the CloudSat  
535 simulator. This issue motivates further investigation in future studies involving applications of the CloudSat simulator  
536 to E3SM. The updates described herein have increased the WRDs' utility for evaluating model warm rain process  
537 representation and support the analysis needed to derive a constraint on ERFaci from CFODD analysis. Through an  
538 evaluation of E3SMv2, we demonstrate that the updated WRDs illuminate specific biases in SLWC representation  
539 and provide contextual sampling statistics that are critical for interpreting CFODD results and thus, for future  
540 applications of this observational constraint on ERFaci.

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542 *Code and Data Availability:* The CloudSat and MODIS data products are available from the CloudSat Data Processing  
543 Center at CIRA/Colorado State University (<https://www.cloudsat.cira.colostate.edu/>; last access: June 28, 2023). The  
544 reference A-Train data used in this study is available here: <https://doi.org/10.5281/zenodo.8384180>. The modified  
545 source code of COSPv2.0 is available here: <https://doi.org/10.5281/zenodo.8371120> and the E3SMv2 source code is  
546 available here: <https://github.com/E3SM-Project/E3SM> (last access: September 27, 2023). The python package for  
547 the two-dimensional Kolmogorov-Smirnov test applied in this study is available here  
548 (<https://github.com/syrte/ndtest/tree/master>; last access: June 28, 2023). The python package scikit-learn was used for  
549 robust linear regression analysis (<https://scikit-learn.org/stable/>; last access: June 28, 2023).

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552 analysis. MWC led the A-Train observations analysis and provided guidance on additional WRDs diagnostics  
553 development. AV provided input on CFODD analysis applications. JM provided guidance on ERFaci analysis. TM  
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