Earth System Model Aerosol-Cloud Diagnostics Package (ESMAC Diags) Version 2: Assessments of Aerosols, Clouds and Aerosol-Cloud Interactions Through Field Campaign and Long-Term Observations

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Abstract.

Poor representations of aerosols, clouds and aerosol-cloud interactions (ACI) in Earth System Models (ESMs) have long been the largest uncertainties in predicting global climate change. Huge efforts have been made to improve the representation of these processes in ESMs, and key to these efforts is evaluation of ESM simulations with observations. Most well-established ESM diagnostics packages focus on the climatological features; however, they are lacking process-level understanding and representations of aerosols, clouds, and ACI. In this study, we developed an ESM aerosol-cloud diagnostics package (ESMAC Diags) to facilitate routine evaluation of aerosols, clouds and aerosol-cloud interactions simulated by the Department of Energy's (DOE) Energy Exascale Earth System Model (E3SM). This paper documents its version 2 functionality (ESMAC Diags v2), which has substantial updates from its version 1 (Tang et al., 2022a). The simulated aerosol and cloud properties have been extensively compared with in-situ and remote-sensing measurements from aircraft, ship, surface and satellite platforms in ESMAC Diags v2. It currently includes six field campaigns and two permanent sites covering four geographical regions: Eastern North Atlantic, Central U.S., Northeastern Pacific and Southern Ocean, where frequent liquid or mixed-phase clouds are present and extensive measurements are available from the DOE Atmospheric Radiation Measurement user facility and other agencies. ESMAC Diags v2 generates various types of single-variable and multi-variable diagnostics, including percentiles, histograms, joint histograms and heatmaps, to evaluate model representation of aerosols, clouds, and ACI. Select examples highlighting ESMAC Diags capabilities are shown using E3SM version 2 (E3SMv2). E3SMv2 in general can reasonably reproduces many observed aerosol and cloud properties, with biases in some variables such as aerosol particle and cloud droplet sizes and number concentrations. The coupling of aerosol and cloud number concentrations may be too strong in E3SMv2, possibly indicating a bias in processes that control aerosol activation. Furthermore, the liquid water path response to perturbed cloud droplet number concentration behaves differently in E3SMv2 and observations, which warrants a further study to improve the cloud microphysics parameterizations in E3SMv2.

1. Introduction

Poor representations of aerosols, clouds and aerosol-cloud interactions (ACI) in Earth System Models (ESMs) have long been the largest uncertainties in predicting global climate change (IPCC, 2021). Challenges come from several aspects: first, there are many aerosol properties (e.g., number, size, phase, shape, composition) and cloud micro- and macro-physical properties (e.g., fraction, water content, number and size of liquid and ice hydrometeors) that affect Earth's climate. Coincident measurements of these properties remain largely under-sampled due to substantial spatiotemporal variability and logistical difficulties for making such measurements. Second, there are complex interactive processes between aerosols, clouds, and ambient meteorological conditions, many of which are not fully understood, but are critical to properly interpreting relationships between observable properties. Third, many ACI processes are nonlinear, multi-scale processes that involve feedbacks depending on cloud types and meteorological regimes, which also shift in space and time, presenting challenges for assessing causal effect and representing such processes in ESMs.

Huge efforts have been made to improve the representation of aerosols, clouds and ACI in ESMs. Key to these efforts is evaluation of ESM simulations with observations. Many modeling centers have developed standardized diagnostics packages to document ESM performance. For aerosol and cloud properties, most diagnostic packages rely heavily on satellite measurements as evaluation data (e.g., AMWG, 2021; E3SM, 2021; Eyring et al., 2016; Gleckler et al., 2016; Maloney et al., 2019; Myhre et al., 2013; Schulz et al., 2006). Satellite remote sensing measurements have global or near global coverage but limited spatial and temporal resolution. They are also facing many challenges to retrieve some variables, especially for aerosol properties such as number concentration, size distribution, chemical composition etc. Some recent studies (e.g., Choudhury and Tesche, 2022) have retrieved cloud condensation nuclei (CCN) number concentration from satellite measurements, which provides a great addition to investigate ACI in global scale. However, large uncertainties exist in satellite retrievals, even for more sophisticated retrieved cloud microphysical properties such as droplet number concentration (e.g., Grosvenor et al., 2018). This limits their application to robustly quantify aerosols, clouds and ACI processes. In-situ measurements from ground, aircraft or ship platforms from field campaigns are also used in a few projects to evaluate ESMs (e.g., Reddington et al., 2017; Watson-Parris et al., 2019; Tang et al., 2022a; Zhang et al., 2020). Some of these field campaigns were conducted over remote or poorly sampled locations, which are highly valuable for model evaluation despite limited spatial coverage and time periods. Moreover, the U.S. Department of Energy (DOE) Atmospheric Radiation Measurement (ARM) user facility has conducted continuous field measurements at a few sites for multiple years. These long-term high-resolution field measurements have also been demonstrated to be valuable for evaluating ESMs (e.g., Zhang et al., 2020).

In response to the need for more ESM diagnostics for evaluating ACI processes, Tang et al. (2022a) developed an ESM aerosol-cloud diagnostics package (ESMAC Diags) to facilitate the routine evaluation of aerosols, clouds and ACI simulated by the Department of Energy's (DOE) Energy Exascale Earth System Model (E3SM, Golaz et al., 2019). It includes diagnostics that leverage in-situ measurements from multiple platforms during six field campaigns since 2013, which are not included in previous diagnostics tools (e.g., Reddington et al., 2017). Version 1 of ESMAC Diags (ESMAC Diags v1, Tang et al., 2022a) mainly focuses on aerosol properties. We present here version 2 of ESMAC Diags (ESMAC Diags v2) that is a direct extension of ESMAC Diags v1 with two major additions:

80   1. measurements from satellite and long-term diagnostics at the ARM Southern Great Plains
81 (SGP) and Eastern North Atlantic (ENA) sites.

82   2. diagnostics for cloud properties and aerosol-cloud interactions.

83 The new measurements, as well as major data quality controls are introduced in Section 2. Additional
84 discussions on retrieval uncertainties of cloud microphysical properties are performed in Section 3.
85 Details of the code structure of ESMAC Diags v2, which is substantially changed since version 1, are
86 described in Section 4. Section 5 provides selected examples of single-variable and multi-variable
87 diagnostics using ESMAC Diags v2 to highlight its capabilities. Lastly, Section 6 provides a summary.

2. Aerosol and cloud measurements from ground, aircraft, ship and satellite platforms

89 Following the initial development in version 1, ESMAC Diags v2 continues to focus on six field
90 campaigns conducted in four geographical regions: the Central U.S. (CUS, where the ARM Southern
91 Great Plains (SGP) site is located), Eastern North Atlantic (ENA), Northeastern Pacific (NEP), and
92 Southern Ocean (SO). Information on the six field campaigns is shown in Table 1 and their locations are
93 shown in Figure 1, each reproduced from Table 1 and Figure 3 in Tang et al. (2022a).

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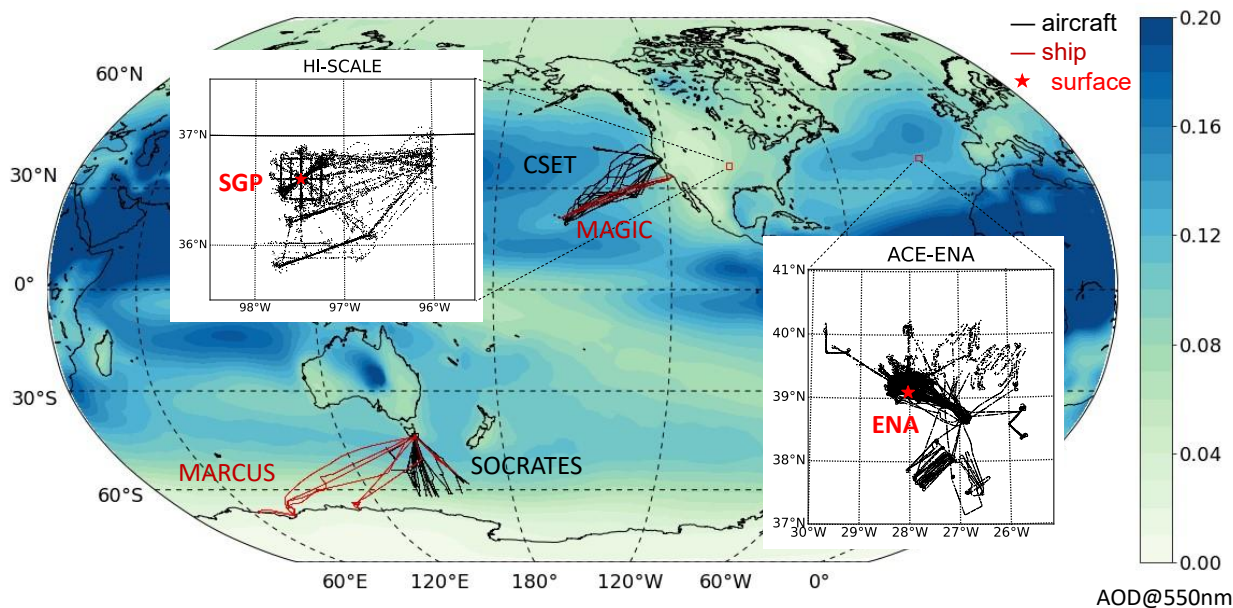
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96 Figure 1. Aircraft (black) and ship (red) tracks for the six field campaigns. Red stars in
97 the enlarged map indicate two ARM fixed sites: SGP and ENA, that have long-term
98 measurements available for model diagnostics. Overlaid is aerosol optical depth at 550nm
99 averaged from 2014 to 2018 simulated in E3SMv1. (Reproduced from Figure 3 in Tang et
100 al., 2022a)

101 Table 1. Descriptions of the field campaigns used in this study. (Reproduced from Table 1
102 in Tang et al., 2022a)

Campaign*	Period	Platform	Typical Conditions	Reference

3

HI-SCALE	IOP1: 24 Apr – 21 May 2016 IOP2: 28 Aug – 24 Sep 2016	Ground, aircraft (IOP1: 17 flights, IOP2: 21 flights)	Continental cumulus with high aerosol loading	(Fast et al., 2019)
ACE-ENA	IOP1: 21 Jun – 20 Jul 2017 IOP2: 15 Jan – 18 Feb 2018	Ground, aircraft (IOP1: 20 flights, IOP2: 19 flights)	Marine stratocumulus with low aerosol loading	(Wang et al., 2021)
MAGIC	Oct 2012 – Sep 2013	Ship (18 legs)	Marine stratocumulus to cumulus transition with low aerosol loading	(Lewis and Teixeira, 2015; Zhou et al., 2015)
CSET	1 Jul – 15 Aug 2015	Aircraft (16 flights)	Same as above	(Albrecht et al., 2019)
MARCUS	Oct 2017 – Apr 2018	Ship (4 legs)	Marine liquid and mixed phase clouds with low aerosol loading	(McFarquhar et al., 2021)
SOCRATES	15 Jan – 24 Feb, 2018	Aircraft (14 flights)	Same as above	(McFarquhar et al., 2021)

103 * Full names of the listed field campaigns:
104 HI-SCALE: Holistic Interactions of Shallow Clouds, Aerosols and Land Ecosystems
105 ACE-ENA: Aerosol and Cloud Experiments in the Eastern North Atlantic
106 MAGIC: Marine ARM GCSS Pacific Cross-section Intercomparison (GPCI) Investigation of Clouds
107 CSET: Cloud System Evolution in the Trades
108 MARCUS: Measurements of Aerosols, Radiation and Clouds over the Southern Ocean
109 SOCRATES: Southern Ocean Cloud Radiation and Aerosol Transport Experimental Study
110

111 The collection and processing of observations are the most time-consuming part of developing ESMAC
112 Diags, which also impacts the reliability of conclusions drawn from the model diagnostics. In this section,
113 we introduce the data used in ESMAC Diags v2, existing quality issues in some datasets, and treatments
114 to address these quality issues. Some variables are difficult to directly measure or have limited in-situ
115 sampling and thus must be derived from remote sensing measurements using retrieval algorithms. In
116 Section 3, we further discuss the uncertainty and reliability of some cloud retrieval products via
117 comparisons with in-situ aircraft measurements.

118 2.1. Data availability

119 All measurements, instruments, and data products used in the six field campaigns and two long-term sites
120 in ESMAC Diags v2 are shown in Table 2. Further details of the measurements, data product names, and
121 DOIs are given in Tables S1 to S6 (for field campaigns) and Tables S7 and S8 (for SGP and ENA sites) in
122 the supplementary material. To allow maximum overlapping of key measurements while also ensuring a
123 long enough period for statistical evaluation, we select the periods of 1 Jan 2011 – 31 Dec 2020 for SGP
124 and 1 Jan 2016 – 31 Dec 2018 for ENA for long-term analyses. In addition to the aerosol measurements
125 discussed in Tang et al. (2022a), we incorporate more cloud and radiation measurements, as well as
126 geostationary satellite retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST)
127 (Minnis et al., 2008; Minnis et al., 2011) algorithm. The VISST products archived by ARM cover
128 approximately 10° by 10° regions in 0.5° by 0.5° resolution centered over ARM sites. Moreover, ARM
129 recently released products consisting of merged aerosol particle and cloud droplet size distributions from

130 aircraft measurements for HI-SCALE and ACE-ENA campaigns. These data are now used in ESMAC
131 Diags v2.

132 Table 2: List of instruments and measurements used in ESMAC Diags v2.

Platform	Measurements	Instruments / data products	Available campaigns
Ground	Surface temperature, relative humidity, wind, pressure, precipitation; upper-level temperature, relative humidity, wind	Surface meteorological station (MET), ARM best estimate (ARMBE) products	HI-SCALE, ACE-ENA, SGP, ENA
	Longwave and shortwave radiation, cloud fraction	ARM best estimate (ARMBE) products	HI-SCALE, ACE-ENA, SGP, ENA
	Aerosol number concentration	Condensation particle counter (CPC), Condensation particle counter – fine (CPCF), Condensation particle counter – ultrafine (CPCU), Ultra-high sensitivity aerosol spectrometer (UHSAS), Scanning mobility particle sizer (SMPS)	HI-SCALE, ACE-ENA, SGP, ENA
	Aerosol size distribution	Ultra-high sensitivity aerosol spectrometer (UHSAS), Scanning mobility particle sizer (SMPS), Nano scanning mobility particle sizer (nanoSMPS)	HI-SCALE, ACE-ENA, SGP, ENA
	Aerosol composition	Aerosol chemical speciation monitor (ACSM)	HI-SCALE, ACE-ENA, SGP, ENA
	CCN number concentration	Cloud condensation nuclei (CCN) counter	HI-SCALE, ACE-ENA, SGP, ENA
	Cloud optical depth	Multifilter rotating shadowband radiometer (MFRSR)	HI-SCALE, ACE-ENA, SGP, ENA
	Cloud droplet number concentration	Cloud droplet number concentration retrieval (Ndrop), cloud retrieval from Wu et al. (2020)	HI-SCALE, ACE-ENA, SGP, ENA
	Cloud droplet effective radius	Multifilter rotating shadowband radiometer (MFRSR), cloud retrieval from Wu et al. (2020)	HI-SCALE, ACE-ENA, SGP, ENA
	Cloud liquid water path	Microwave radiometer (MWR), ARM best estimate (ARMBE) products	HI-SCALE, ACE-ENA, SGP, ENA
	Cloud base height, cloud top height	Active remote sensing of clouds (ARSCL)	HI-SCALE, ACE-ENA, SGP, ENA
Satellite	TOA shortwave and longwave radiation	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA
	cloud fraction; height, pressure and temperature at cloud top	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA
	liquid water path; cloud optical depth; droplet effective radius	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA
	Cloud droplet number concentration	Retrieved from VISST data using the algorithm in Bennartz (2007)	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA
Aircraft	Navigation information and meteorological parameters	Interagency working group for airborne data and telemetry systems (IWG)	HI-SCALE, ACE-ENA
	Aerosol number concentration	Condensation particle counter (CPC), Condensation particle counter – ultrafine (CPCU), Condensation nuclei counter (CNC), Ultra-high sensitivity aerosol spectrometer (UHSAS), Passive cavity aerosol spectrometer (PCASP)	HI-SCALE, ACE-ENA, CSET, SOCRATES

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	Aerosol size distribution	Ultra-high sensitivity aerosol spectrometer (UHSAS), Fast integrated mobility spectrometer (FIMS), Passive cavity aerosol spectrometer (PCASP), Best estimate aerosol size distribution (BEASD)	HI-SCALE, ACE-ENA, CSET, SOCRATES
	Aerosol composition	High-resolution time-of-flight aerosol mass spectrometer (AMS)	HI-SCALE, ACE-ENA
	CCN number concentration	Cloud condensation nuclei (CCN) counter	HI-SCALE, ACE-ENA, SOCRATES
	Cloud liquid water content	Water content measuring system (WCM), PMS-King Liquid Water Content (LWC)	HI-SCALE, ACE-ENA, CSET, SOCRATES
	Cloud droplet number size distribution	1DC, 2DC, 2DS, CDP, Cloud probe merged size distribution (mergedSD)	HI-SCALE, ACE-ENA, CSET, SOCRATES
Ship	Navigation information and meteorological parameters	Meteorological station (MET)	MAGIC, MARCUS
	Aerosol number concentration	Condensation particle counter (CPC), Ultra-high sensitivity aerosol spectrometer (UHSAS)	MAGIC, MARCUS
	Aerosol size distribution	Ultra-high sensitivity aerosol spectrometer (UHSAS)	MAGIC, MARCUS
	CCN number concentration	Cloud condensation nuclei (CCN) counter	MAGIC, MARCUS
	Cloud liquid water path	Microwave radiometer (MWR)	MAGIC, MARCUS
	Cloud droplet number concentration, cloud effective radius	Cloud retrieval from Wu et al. (2020)	MAGIC

133

134 All the observational data are quality controlled with their time resolution re-scaled to that suitable for
135 evaluating E3SM, and the rescale resolution can be adjusted to fit for different model output frequencies.
136 Currently, ground, ship and satellite measurements are re-scaled to a 1-hour frequency to be consistent
137 with current E3SM output frequency. Rescaling consists of computing either the median, mean or
138 interpolated value depending on the original data frequency and variable properties. For most aerosol and
139 cloud microphysics measurements, the median value is computed to remove occasional spikes or zeros
140 resulting from data contamination or measurement error. For some bulk cloud properties (e.g., cloud
141 fraction, liquid water path (LWP)), the mean value is computed to be consistent with grid-mean E3SM
142 output. Interpolation is only used when the input frequency is equal to or coarser than the frequency of
143 model output. For aircraft measurements, 1-minute resolution is used to retain high variability and allow
144 matching samples of aerosol and cloud at the same time. To compare with high-frequency aircraft data,
145 E3SM output is interpolated to the same resolution using the nearest grid cell and time slice. Although the
146 current 1-hour, 1-degree E3SM output could not capture the high variability of the aircraft measurements,
147 we are targeting the exascale E3SM version planned in the next few years. In kilometer scale resolution
148 ESM simulations, the high variability in aircraft measurements will be better captured. In the current
149 diagnostics we only focus on the statistics for the entire campaign. As seen later in Section 5.1, coarse-
150 resolution model outputs show similar percentile ranges with the high-resolution aircraft measurements,
151 indicating that for simple percentiles, large-scale variabilities dominate over subgrid variabilities over
152 month-long field campaign periods. Further analysis is needed to understand the importance of other
153 statistics (variance, covariance, etc.) of subgrid scale variabilities. . All processed data are saved in a
154 standardized NetCDF format (NETCDF, 2022) and available for downloading (see data availability
155 section) and direct use.

156 2.2 Data quality issues and treatments

157 Many observation datasets used in ESMAC Diags are ARM level-b (quality-controlled) or level-c (value-
158 added) products, which include quality control (QC) flags to indicate data quality issues. For most
159 datasets, a QC treatment is applied to remove all data with questionable flags. However, there are certain
160 datasets or circumstances in which a QC flag is overly strict (too many good data are removed) or not
161 strict enough (some bad data are not removed). Here we document some of these situations and how we
162 handle them in our data processing.

2.2.1 ARM Condensation Particle Counter (CPC) measurements

164 ARM CPC data have several QC values representing failure of different quality checks. One of them
165 checks if the concentration is greater than a maximum allowable value, which is set to 8,000 cm^{-3} for
166 model 3010 (CPC, size detection limit 10 nm), 10,000 cm^{-3} for model 3772 (CPCF, size detection limit 10
167 nm), and 50,000 cm^{-3} for model 3776 (CPCU, size detection limit 3 nm). At SGP, new particle formation
168 (NPF) events occur frequently when CPC and CPCF measurements can exceed 30,000 cm^{-3}. This is much
169 higher than the maximum allowable value but physically reasonable. Simply removing these large values
170 results in an underestimation of aerosol number concentration and produces unrealistic diurnal cycle since
171 they usually occur during the daytime (Tang et al., 2022a). By consulting with the ARM instrument
172 mentor, we only remove data with critical QC flags, but keep data with this QC flag that is overly
173 restrictive.

2.2.2 NCAR research flight aerosol number concentration (CN) measurements

175 NCAR research flight (RF) data used in ESMAC Diags do not include QC flags but occasionally show
176 suspiciously large or negative aerosol counts. The following minimum and maximum thresholds are
177 applied to remove suspicious data:

- 178 Total CN from a Condensation Nucleation Counter (CNC, reported as CONCN): minimum = 0,
  179 maximum = 25,000 cm^{-3}.
- 180 Total CN from an Ultra-High-Sensitivity Aerosol Spectrometer (UHSAS, reported as
  181 UHSAS100): minimum = 0, maximum = 5,000 cm^{-3}.
- 182 Aerosol number size distribution from an UHSAS (reported as CUHSAS_RWOOU or
  183 CUHSAS_LWII): minimum = 0, maximum = 500 cm^{-3} per size bin.

2.2.3 Ship-measured aerosol properties

185 Aerosol instruments on ships are occasionally contaminated by ship emissions, which present as large
186 spikes in aerosol and CCN number concentrations. For ARM MARCUS measurements, Humphries
187 (2020) published reprocessed CN and CCN data to remove ship exhaust contamination using method
188 described in Humphries et al. (2019). This data is used in this diagnostics package. For MAGIC, we could
189 not find any ship exhaust contamination information. By visually examining the dataset, a simple
190 maximum threshold (25,000 cm^{-3} for CPC, 5,000 cm^{-3} for UHSAS100, 2,000 cm^{-3} for CCN at 0.1%
191 supersaturation and 4,000 cm^{-3} for CCN at 0.5% supersaturation) is applied to remove likely
192 contamination from ship emissions.

2.2.4 CCN measurements

194 There are different supersaturation (SS) setting strategies for CCN measurements. Some aircraft
195 campaigns measured CCN with constant SS (ACE-ENA, HI-SCALE). Some other campaigns measured
196 CCN with time-varying (scanning) SS (SOCRATES, surface CCN counters at SGP and ENA). However,
197 the actual SS in a scanning strategy has fluctuations that are different than the target SS. For the latter,
198 CCN for each SS (0.1%, 0.2%, 0.3% and 0.5%) are obtained by selecting CCN measured within $\pm 0.05\%$
199 of the SS target.

200 For long-term measurements at SGP and ENA, near-hourly CCN spectra data are available, and a
201 quadratic polynomial is fit to the spectra such that CCN number concentration can be estimated at any SS
202 between the measured minimum and maximum SS values. We calculate and output CCN number
203 concentration from these fits at three target supersaturations (0.1%, 0.2% and 0.5%). The fitted spectra
204 data provides CCN number concentration at the exact target supersaturations, but the sample number is
205 slightly smaller due to occasional failure of polynomial fitting.

206 2.2.5 Contaminated surface aerosol measurements at ENA

207 The ARM ENA site is located at a local airport. Aerosol measurements at ENA are sometimes
208 contaminated by aircraft and vehicle emissions, rendering the measurements not representative of the
209 background environment. Gallo et al. (2020) identified periods when CPC measurements were likely
210 contaminated from localized emissions (Figure 2a). Their aerosol mask data has 1-min resolution. When
211 we rescale the data to 1-hr resolution and apply the mask on other coarse time-resolution aerosol
212 measurements (e.g., ACSM, Figure 2c), we mask hours in which more than half of the hour is flagged by
213 the aerosol mask. The masking slightly increases the occurrence fraction of small values due to removing
214 many large values, but it does not change the overall distribution (Figure 2b and 2d). A sensitivity
215 analysis was performed, showing that 50% is a reasonable threshold to balance removal of contamination
216 with keeping reasonable data (not shown).
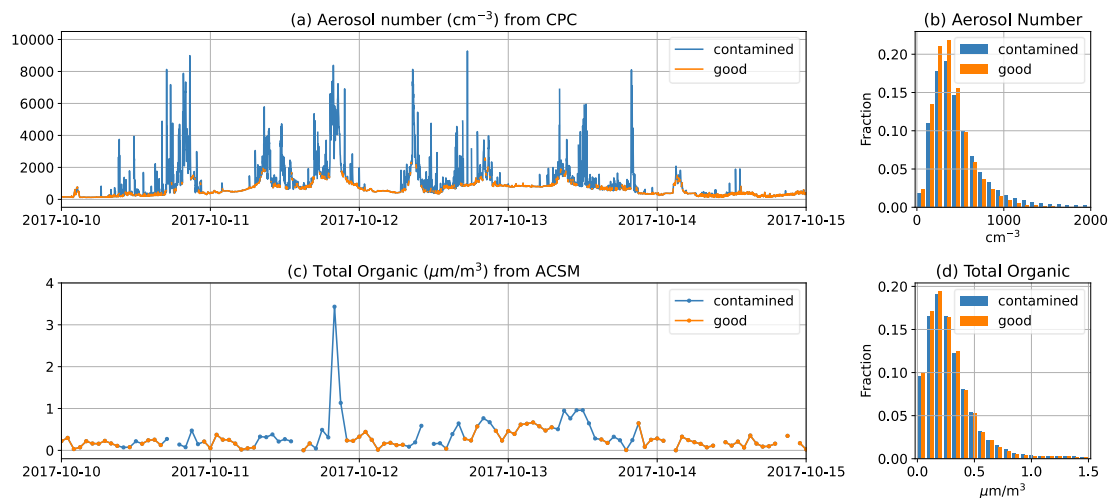
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218 Figure 2: (a) CPC-measured CN from 10 to 15 October 2017 (1-minute resolution) with
219 local contamination flagged by Gallo et al. (2020). (b) histogram of CPC-measured CN for
220 all data from 2016-2018. (c) ACSM measured total organic matter from 10 to 15 October
221 2017 (1-hour resolution). Hours with more than half or the hour flagged in 1-minute CPC

3. Verification of cloud retrievals with in-situ measurements

225 Cloud microphysical properties such as droplet number concentration (N_d) and effective radius (R_{eff}) are
226 important variables that connect clouds to other aspects in the climate system such as aerosols and
227 radiation. Except in field campaigns where in-situ aircraft measurements are available, remote sensing
228 retrieval algorithms are usually needed to derive these quantities. Several cloud retrieval products from
229 ground and satellite measurements with different algorithms are used in ESMAC Diags v2. This section
230 compares these cloud retrievals with in-situ aircraft measurements to assess retrieval limitation,
231 uncertainty, and utility. N_d and R_{eff} from aircraft measurements taken during HI-SCALE and ACE-ENA
232 field campaigns are calculated from merged cloud droplet number size distributions (mergedSD) from
233 three different cloud probes with different size ranges. The mergedSD covers the size range from 1.5 µm
234 to 9075 µm, covering the entire E3SM cloud droplet size distribution range and extending to rain droplet
235 size range (> 100 µm). For field campaigns used in this study, the aircraft only flied through non-
236 precipitating or drizzling clouds, in which the airborne measurements usually measure rain droplet
237 number 3 to 5 orders of magnitude smaller than cloud droplet number. Therefore, the inclusion of rain
238 droplet size range has ignorable impact on the aircraft-estimated N_d and R_{eff}.

239 Table 3 lists R_{eff} and N_d retrieval products used in ESMAC Diags v2. We retrieved Nd_sat with input
240 data from VISST products using the algorithms described in Bennartz (2007), but assuming a ratio of the
241 drop volume mean radius to R_{eff} (commonly referred to as *k*) of 0.74 and a cloud adiabaticity of 80%
242 (Varble et al., 2023). Other datasets are all available as released products. All retrievals assume a
243 horizontally homogeneous single-layer liquid phase cloud with constant N_d throughout the cloud layer.
244 However, retrieval algorithms are usually run for all conditions whenever they return valid values. When
245 assumptions are not satisfied, retrieved properties may contain large errors and likely alter statistics such
246 as increasing the occurrence frequency of small N_d as will be shown next.

247 Table 3: Cloud droplet effective radius R_{eff} and number concentration N_d retrievals

Variable	Dataset	Platform	Campaign/site	Retrieved from	Reference
R_{eff}	MFRSRCLDOD	Ground	HI-SCALE, ACE-ENA, SGP, ENA	SW diffuse flux, LWP	(Min and Harrison, 1996; Turner et al., 2021)
	VISST	Satellite	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA	Brightness temperature	(Minnis et al., 2011)
	Wu_etal	Ground	ACE-ENA, MAGIC, ENA	Radar reflectivity, LWP	(Wu et al., 2020)
N_d	Ndrop	Ground	HI-SCALE, ACE-ENA, SGP, ENA	LWP, COD, cloud height	(Riihimaki et al., 2021; Lim et al., 2016)
	Nd_sat (calculated from VISST)	Satellite	HI-SCALE, ACE-ENA, MAGIC, MARCUS, SGP, ENA	LWP, COD, CTT	(Bennartz, 2007)

	Wu_etal	Ground	ACE-ENA, MAGIC, ENA	Radar reflectivity, LWP	(Wu et al., 2020)

248 MFRSRCLDOD: Cloud Optical Properties from the MultiFilter Shadowband Radiometer (MFRSR)

249 SW: shortwave

250 COD: cloud optical depth

251 CTT: cloud top temperature

252

253 Figures 3 shows the occurrence fraction histograms of N_d retrievals with aircraft measurements for HI-

254 SCALE and ACE-ENA field campaigns, with the comparison of original temporal resolution versus 30-

255 minute mean, and the use of all available samples and samples that are filtered as overcast (cloud

256 fraction > 90%) low-level (cloud top height < 4 km) clouds. Figure 4 shows similar plots but for R_{eff}.

257 We also selected two cases with single-layer boundary layer stratus or stratocumulus clouds and plotted

258 their timeseries of original-resolution and 30-min averaged R_{eff} and N_d in Figure S1. The high-frequency

259 aircraft measurements and MFRSR/Ndrop retrievals exhibit much larger variability than coarse-frequency

260 retrievals of Wu_etal and VISST. They frequently sample cloud edges or cloud top/base (for aircraft),

261 where N_d is typically less than further into the cloud. This causes large occurrence fractions in the lowest

262 few bins in the N_d histograms (Figure 3a and 3d). The 30-min VISST products also show large

263 occurrence fraction in the lowest N_d bin for HI-SCALE (Figure 3a), likely due to high frequency of

264 partial cloudy condition over continental U.S. Filtering conditions to only include overcast low-level

265 clouds (Figure 3b, e) and averaging into a coarser resolution (Figure 3c, f) both contribute to the reduction

266 of occurrence fraction in small-N_d bins, and make the measurements from different instruments more

267 comparable.

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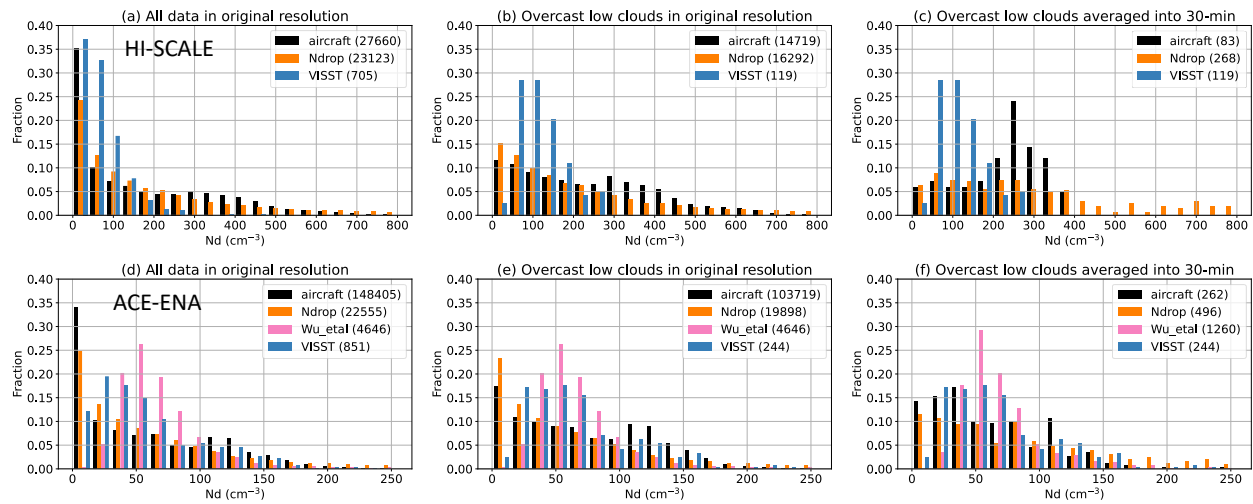
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270 Figure 3: Histogram of N_d from different measurements/retrievals in (top) HI-SCALE and

271 (bottom) ACE-ENA field campaigns, with total sample numbers in the parentheses. (a) and

272 (d) use data samples in their original resolution (1 s for aircraft measurements, 20 s for

273 Ndrop data, 5 min for Wu_etal data, and 30 min for VISST data). (b) and (e) include only

274 overcast low-cloud situations. For aircraft data, this means N_d is > 1 cm^{-3} for 5 s before

275 and after the sampling time; for Ndrop and VISST data, it means cloud fraction > 90% and

276 cloud top height < 4km. (c) and (f) include only overcast low-cloud situations, and

10

average into 30-min resolution. For all the plots, VISST data with solar zenith angle > 65°
278 are removed to avoid artifact from sunlight.

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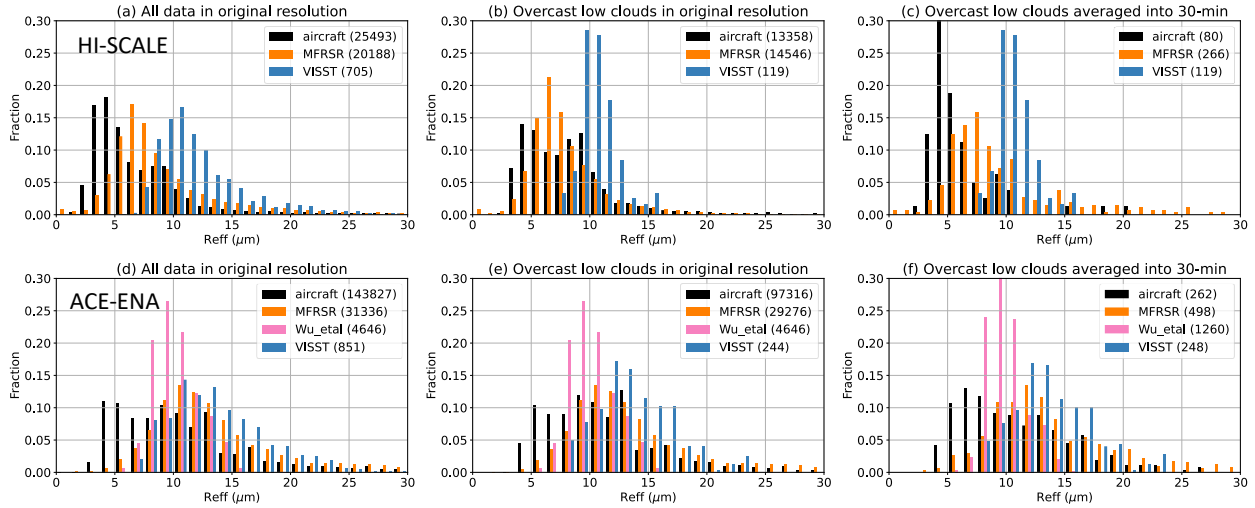
Figure 4: similar as in Figure 3 but for R_{eff}.

Overall, the remote sensing retrievals and aircraft measurements produce reasonable ranges of N_d and R_{eff}. Marine clouds (ACE-ENA) have smaller N_d (Figure 3) and larger R_{eff} (Figure 4) than continental clouds (HI-SCALE). Different retrievals are more consistent with each other for marine clouds than continental clouds. Even after rescaling to the same temporal resolution, aircraft and Ndrop data exhibit broader N_d distributions than satellite retrieval, likely due to their high sampling frequency that may capture more extreme conditions with very high or low N_d. Moreover, the assumption of a fixed adiabaticity (0.8) in satellite retrieval will also narrow N_d distribution. For R_{eff}, we do not expect different datasets to be perfectly agree with each other, as cloud droplet size grows with height in the cloud. All remote sensing retrievals have larger R_{eff} values than aircraft measurements, potentially because remote sensors weight more towards the upper cloud where droplet size and liquid water content (LWC) are larger. Wu_etal retrieves vertical profiles of R_{eff}, and a median value of the R_{eff} profile is used to represent the entire cloud. This makes Wu_etal retrieval weight less toward large droplets thus its R_{eff} is less than MFRSR and VISST. VISST data have the largest R_{eff} values, likely because satellite retrievals reflect conditions at the cloud top. Given the spread in retrieved cloud properties, the limitations and uncertainties of cloud microphysics retrievals clearly need to be considered when they are used to evaluate model performances.

4.   Structure of diagnostics package

Figure 5 shows the directory structure of ESMAC Diags v2. It is substantially changed from ESMAC Diags v1 (Tang et al., 2022a). First, we save all data separately as *raw_data,* which stores all input datasets collected from field campaigns, and *prep_data*, which stores preprocessed data with standardized time resolution and quality controls as described in Section 2. The structure is still designed to be flexible for future extension with additional measurements and/or functionality. Second, the diagnostics functions now give users more freedom to modify analyses, such as selecting different time periods, performing

305 additional data filtering or treatments, and examining ACI relationships in specified variable
306 combinations (for scatter plots, joint histograms or heatmaps). We provide a set of example scripts to
307 assist users design their own diagnostics based on their needs. We also provide the source code of data
308 preparation for observations and model output, and a detailed instruction on how to run the code. Users
309 can revise the code to process their own observational data or model output. All the information is
310 available in the ESMAC Diags github repository.
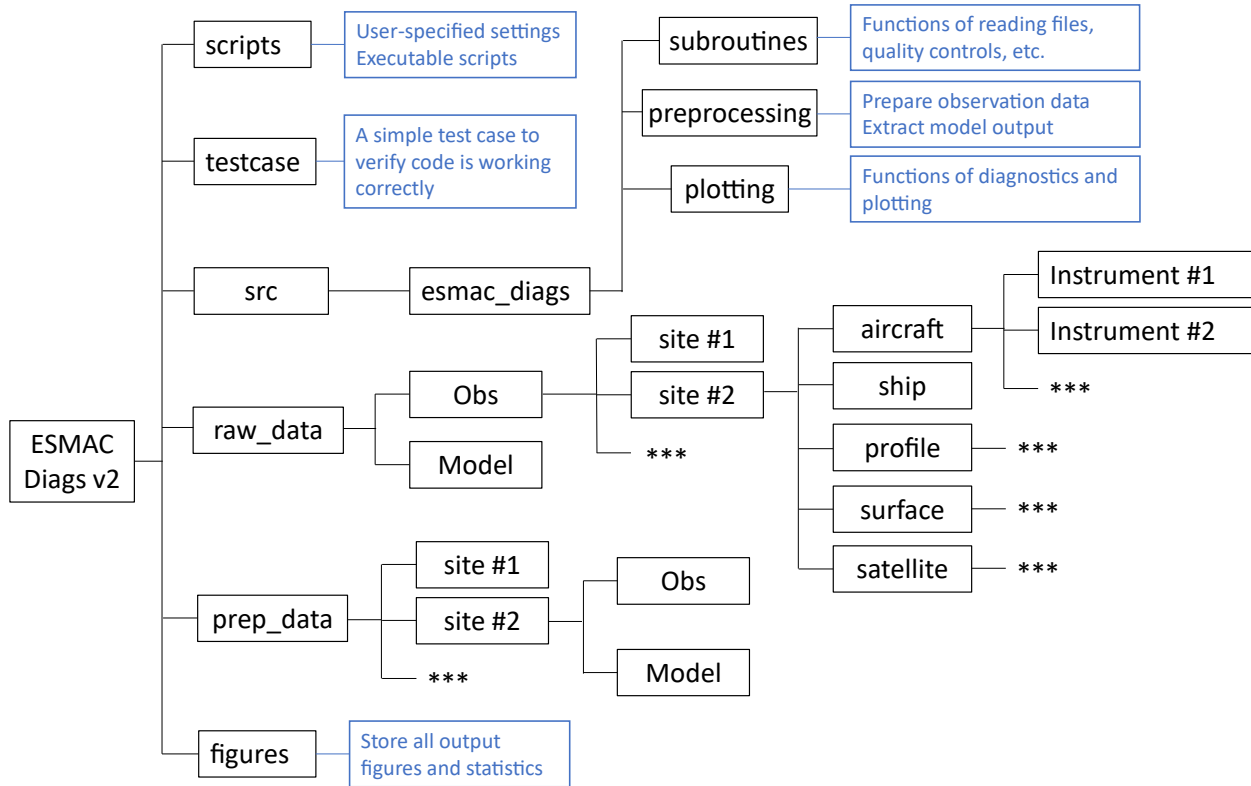
311

312 Figure 5: Directory structure of ESMAC Diags v2. Blue boxes describe the functions of
313 the directory. Asterisks represent boxes that follow the same format as those shown in
314 parallel.

315 ESMAC Diags v1 included diagnostics of aerosol mean statistics (mean, bias, RMSE, correlation),
316 timeseries, diurnal cycle, vertical profiles, mean particle number size distribution, percentiles by
317 height/latitude, and pie/bar charts (Tang et al., 2022a). ESMAC Diags v2 now includes the following new
318 diagnostics that include cloud variables:

319   - 5th, 25th, 50th, 75th and 95th percentiles,
320   - Seasonal cycle at SGP and ENA,
321   - Histograms for individual variables,
322   - Scatter plots,
323   - Joint histograms of two variables, and
324   - Heatmaps of three variables (mean of one variable binned by two other variables).

325   The inclusion of two-variable scatter plots, joint histograms, and three-variable heatmaps provides the
326   functionality to study ACI-related relationships. We present a few examples in the next section to
327   demonstrate these new diagnostics.

328
329   5.   Diagnostics Examples

330   In this section, we show some examples of diagnostics applied to E3SM version 2 (E3SMv2) (Golaz et
331   al., 2022). Compared to the aerosol and cloud parameterizations in E3SMv1 (Rasch et al., 2019; Golaz et
332   al., 2019), E3SMv2 updated the treatments on dust particles, incorporated recalibration of parameters (Ma
333   et al., 2022), changed the call order and refactored the code of the Cloud Layers Unified By Binormals
334   (CLUBB) parameterization, and retuned some parameters (Golaz et al., 2022). We constrain the model
335   simulations by nudging the horizontal winds towards the 3-hourly Modern-Era Retrospective analysis for
336   Research and Applications, Version 2 (MERRA-2, Gelaro et al., 2017) with a nudging time scale of 6
337   hour. Previous studies have shown that with nudging, E3SM can well simulate the large-scale circulations
338   in reanalyses (Sun et al., 2019; Zhang et al., 2022). The model was run for individual field campaigns
339   (Table 1) and from 2010 to 2020 for long-term diagnostics at SGP and ENA sites, with hourly model
340   output saved over the field campaign regions for detail evaluation. As described in Section 2, all
341   diagnostics for ground and ship campaigns are in 1-hour resolution while diagnostics for aircraft
342   campaigns are in 1-minute resolution. For aerosol and cloud variables, model raw output variables (not
343   from instrument simulators) are used in this paper to reveal the intrinsic ACI relationships in E3SM.
344   However, as can be seen later in this section, instrument simulators can be better used in some diagnostics
345   to ensure more consistent comparison. Users may choose whether or not to use simulators in their
346   diagnostics depending on their purpose.

### 347   5.1. Single-variable diagnostics

348   Figures 6 and 7 show mean and percentile values of aerosol and cloud properties measured from field
349   campaigns in the four geographical regions: CUS, ENA, NEP and SO. Figure 6 is for aircraft platforms
350   and Figure 7 is for ground or ship platforms with satellite data included when available. Note that the
351   aircraft and ground/ship campaigns may cover different time periods (Table 1), thus some differences
352   seen between aircraft and ship measurements may be caused by seasonal variation. As cloud
353   microphysical properties are usually retrieved with assumptions (Section 3), for ground/ship/satellite data,
354   we only focus on overcast low-level liquid cloud condition here (cloud fraction > 90%, cloud top height <
355   4 km and ice water path < 0.01 mm). E3SM does not output cloud top height, which is derived using a
356   weighting integration method as described in Varble et al. (2023).

357   From both aircraft and ground/ship data, HI-SCALE has much larger aerosol and cloud droplet number
358   concentrations with smaller droplet sizes compared to other campaigns, which is expected for a
359   continental environment compared to a marine environment. The cloud optical depth is also greater for
360   HI-SCALE than other campaigns, which is driven by smaller droplet sizes rather than LWP differences.
361   Satellite retrievals generally produce smaller N_d, LWP, and cloud optical depth with greater R_{eff} than
362   surface retrievals. As discussed in Section 3, retrieval uncertainties need to be kept in mind when these
363   retrieved microphysical properties are used to evaluate models.

364 E3SMv2 overestimates CN (> 10 nm) over CUS, ENA and NEP. Larger particle concentration (CN > 100
365 nm) is generally underestimated over CUS and overestimated over ENA and NEP. Over SO, E3SMv2
366 produces fewer small aerosol particles (CN > 10 nm) and about the same number of large aerosol
367 particles (CN > 100 nm) compared to the observations. These results are confirmed by both aircraft and
368 ground/ship campaigns, except for the HI-SCALE aircraft campaign where small particles from local
369 emissions were occasionally observed but unable to be simulated. These results are consistent with our
370 previous diagnostics for E3SMv1 (Tang et al., 2022a). E3SMv2 also underestimates N_d over CUS and
371 SO, which corresponds with the underestimation of accumulation mode (> 100 nm) CN over CUS but
372 underestimation of Aitken mode (> 10 nm) CN over SO. It is possible that over very clean regions such as
373 SO, small particles are more important in cloud formation than over continental regions such as CUS.
374 Simulated LWP (LWC) is generally consistent with satellite (aircraft) measurements, but smaller than
375 ground/ship measurements, which may be partly caused by rain contamination of ground/ship retrievals.
376 R_{eff} evaluation is less certain given large discrepancies between satellite and ground retrievals.
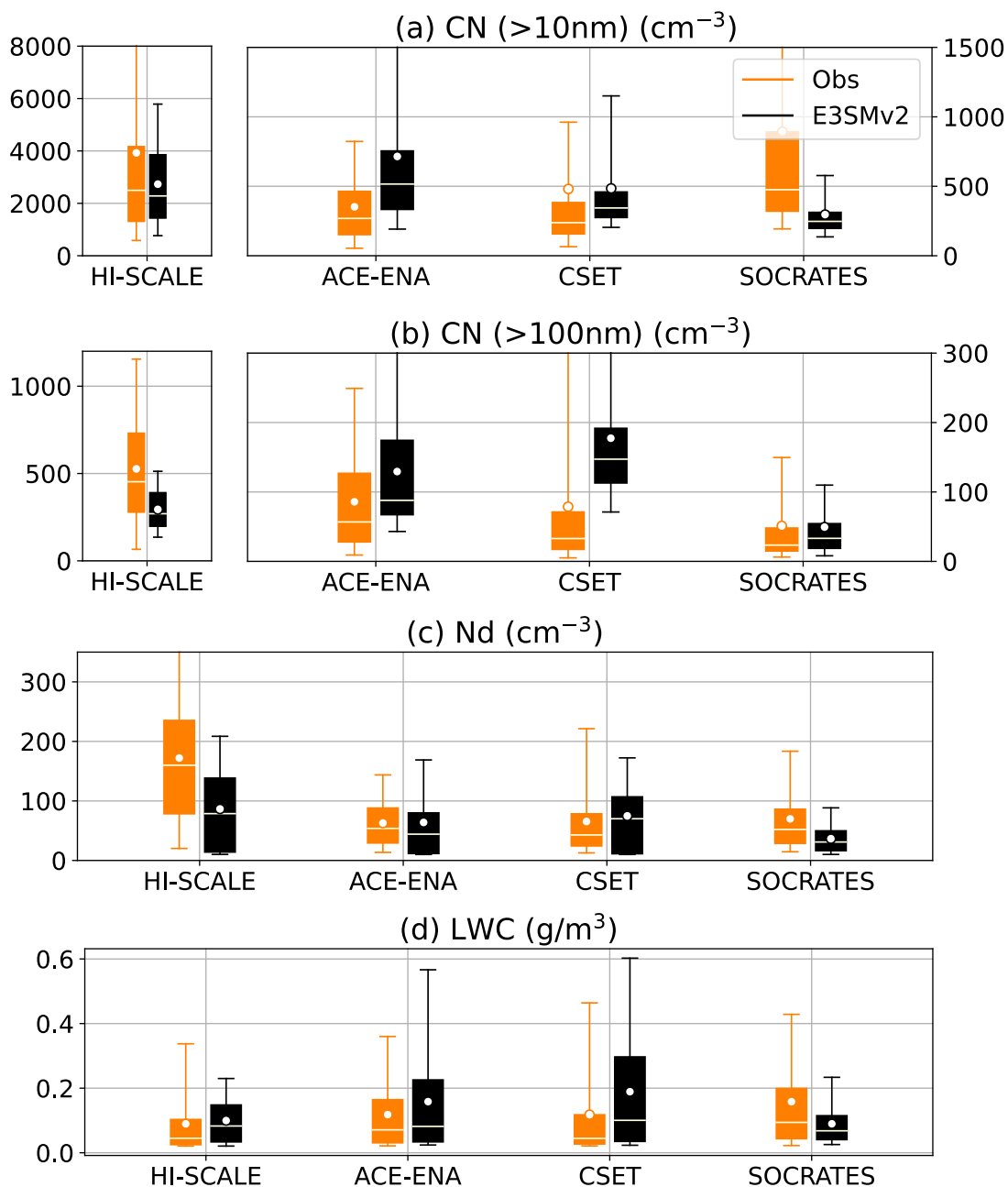
Figure 6: Box-whisker plots of (a) CN for size > 10 nm, (b) CN for size > 100 nm, (c) in-cloud N_d, (d) LWC for all data from aircraft field campaigns at CUS, ENA, NEP and SO regions from left to right. Boxes denote 25th and 75th percentiles, whiskers denote 5th and 95th percentiles, the white horizontal line represents median values, and the white dot represents mean values. For aerosol number concentrations, the y axes for HI-SCALE are separated from other field campaigns for better visualization. The top whiskers that are out of the y-axis range are: (a) HI-SCALE obs: 13681. ACE-ENA E3SMv2: 2061. SOCRATES obs: 2745. (b): ACE-ENA E3SMv2: 304. CSET obs: 305. CSET E3SMv2: 400. (c): HI-SCALE obs: 397.
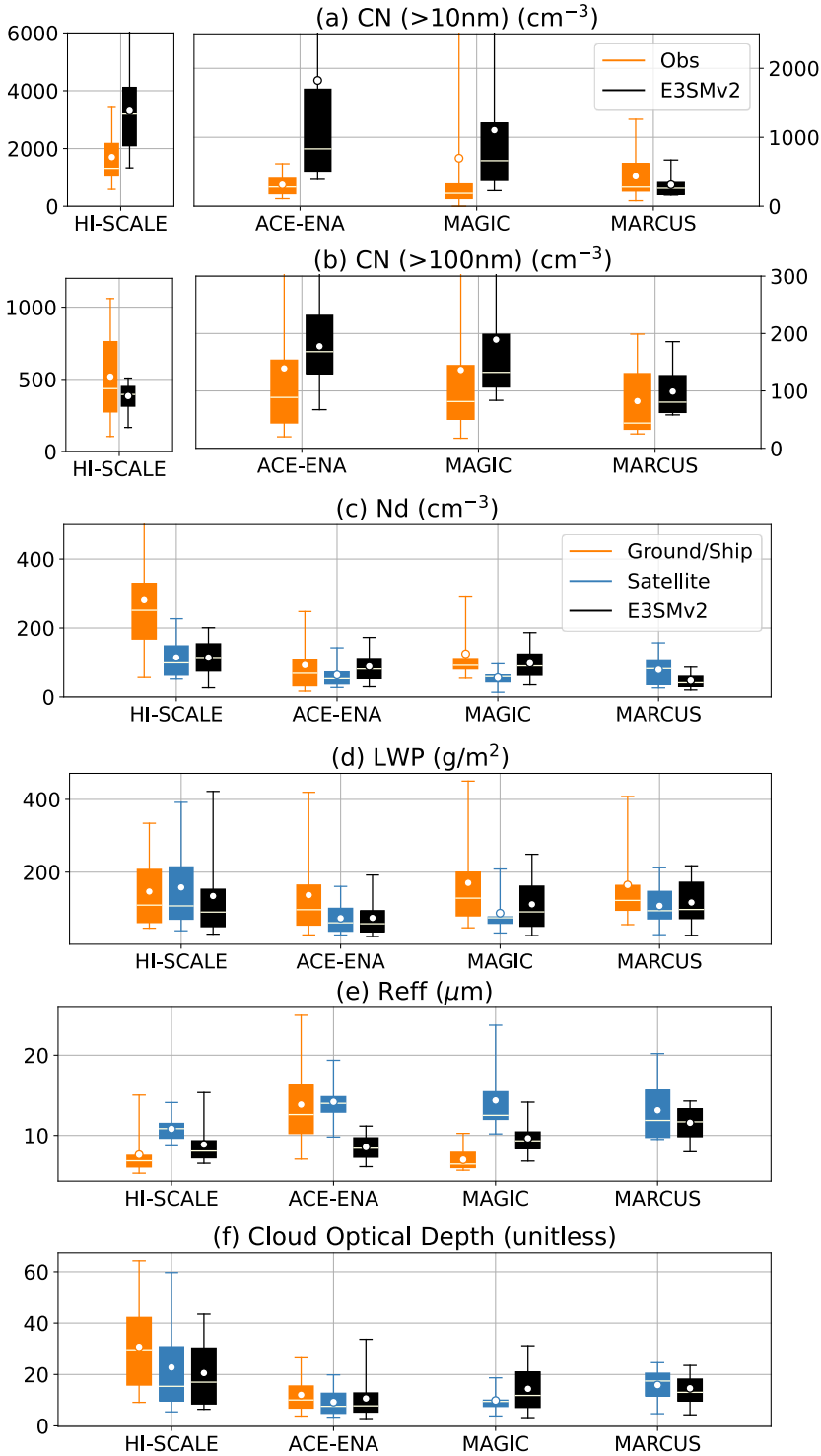
Figure 7: Box-whisker plots of (a) CN for size > 10 nm, (b) CN for size > 100 nm, (c) layer-mean N_d, (d) LWP, (e) R_{eff}, (f) cloud optical depth for overcast low-level liquid cloud conditions (cloud top height < 4 km, cloud fraction > 90% and ice water path < 0.01 mm) in ground and ship field campaigns at CUS, ENA, NEP and SO regions from left to right. Boxes denote 25th and 75th percentiles, whiskers denote 5th and 95th percentiles, the

393   white horizontal line represents median values, and the white dot represents mean values.

394   For aerosol number concentrations, the y axes for HI-SCALE are separated from other

395   field campaigns for better visualization. The top whiskers that are out of the y-axis range

396   are: (a) HI-SCALE E3SMv2: 6102. ACE-ENA E3SMv2: 7575. MAGIC obs: 3330. MAGIC

397   E3SMv2: 3771. (b): ACE-ENA obs: 304.7. ACE-ENA E3SMv2: 328.3. MAGIC obs: 377.7.

398   MAGIC E3SMv2: 577.8. (c): HI-SCALE obs: 670.9.

399
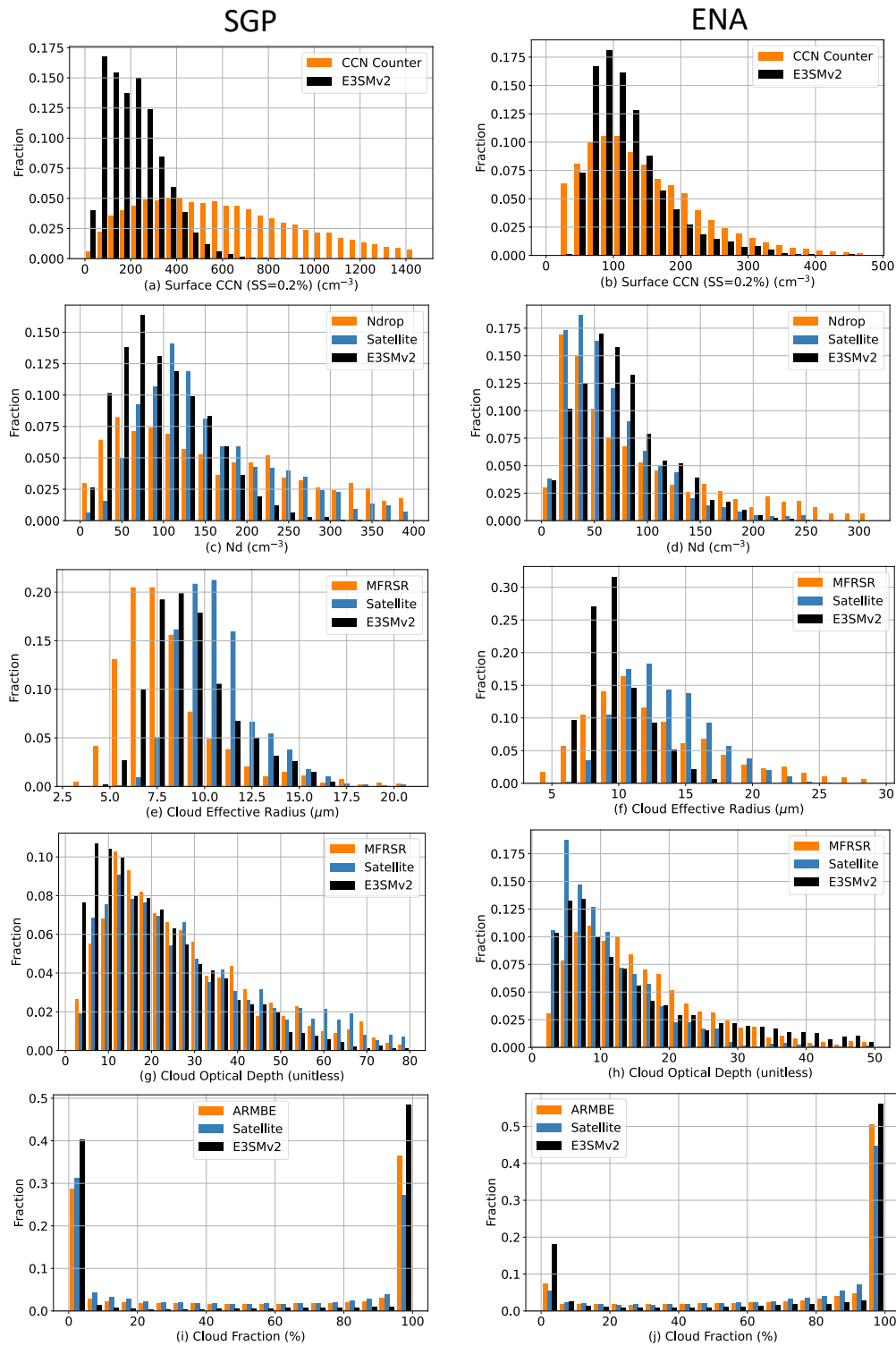
Figure 8: histogram of (from top to bottom) surface CCN number concentration, layer-mean N_d, R_{eff}, cloud optical depth and total cloud fraction at (left) SGP from 2011 to 2020 and (right) ENA from 2016 to 2018. Surface CCN and total cloud fraction are using all-condition samples while N_d, R_{eff}, cloud optical depth data are filtered for overcast

404   low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path <

405   0.01 mm).

406   Figure 8 shows histograms of surface CCN number concentration in 0.2% supersaturation, cloud layer

407   mean N_d, R_{eff}, cloud optical depth and total cloud fraction for long-term diagnostics at SGP (year 2011-

408   2020) and ENA (year 2016-2018) sites. E3SMv2 fails to reproduce the long tail of large values in CCN

409   and N_d, especially over SGP. This is consistent with the underestimation of CN (> 100 nm) during the

410   HI-SCALE field campaign shown in Figures 6 and 7. Compared with ground retrievals, E3SMv2 R_{eff} is

411   larger at SGP but smaller at ENA. However, satellite-retrieved R_{eff} has larger values than E3SMv2 at

412   SGP. As discussed before, discrepancies between satellite and ground retrievals can be substantial for

413   some locations and variables, and considering both in evaluating model performance gives a sense for

414   how uncertain comparisons are. E3SMv2 generally captures the histograms of cloud optical depth and

415   total cloud fraction, although it underestimates the frequency of partial-cloudy conditions and

416   overestimates the frequency of clear-sky and overcast conditions.

417
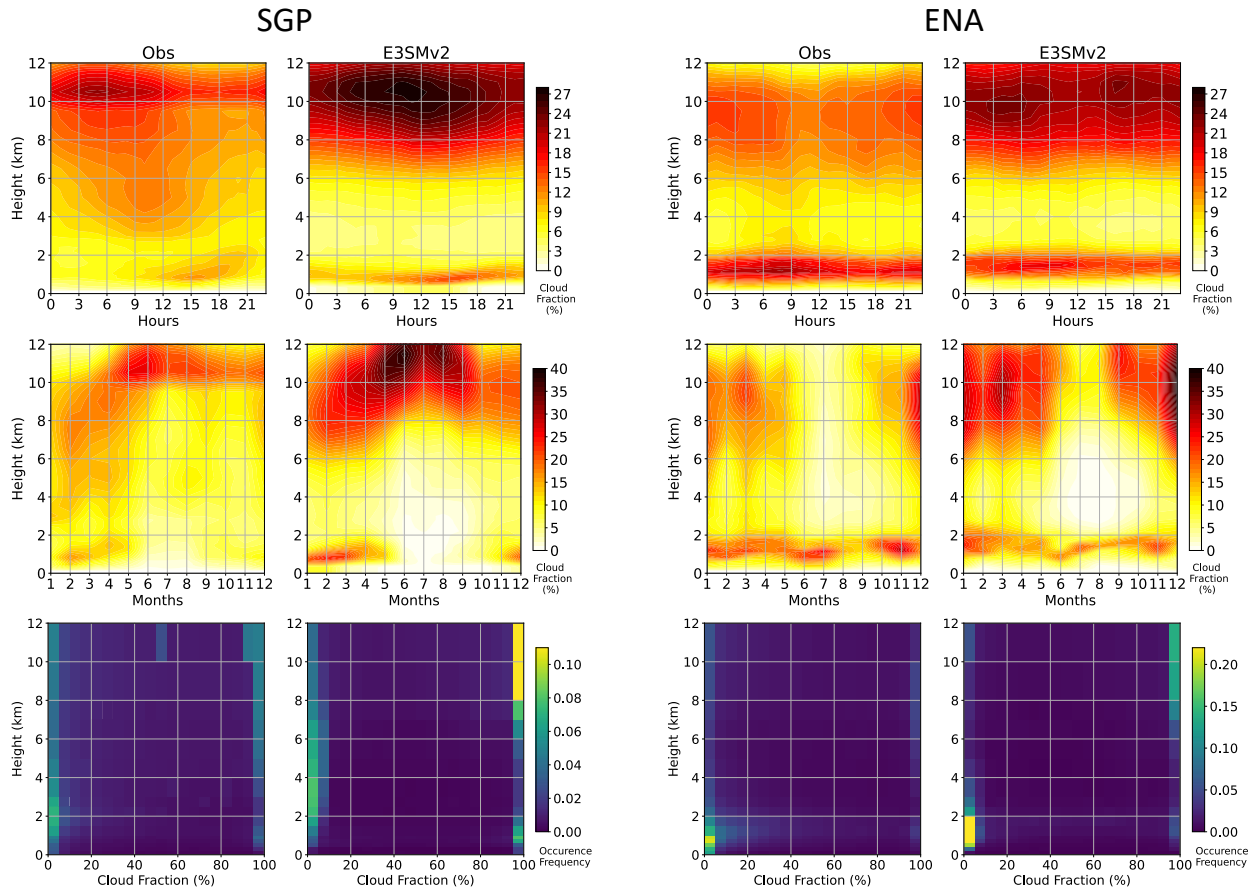
419   Figure 9: (top) Diurnal cycle, (middle) seasonal cycle, and (bottom) occurrence frequency

420   of vertical cloud fraction at (left) SGP from 2011 to 2020 and (right) ENA from 2016 to

421   2018.

19

422 Figure 9 shows the long-term diagnostics of mean diurnal cycles, seasonal cycles and histograms of cloud
423 fraction by height at SGP and ENA sites. Overall, the mean fraction of high clouds looks overestimated in
424 E3SMv2. Similar results has been reported in many previous studies in the Community Earth System
425 Model (CESM)-E3SM model family (e.g., Song et al., 2012; Cheng and Xu, 2013; Xu and Cheng, 2013b,
426 a; Tang et al., 2016; Zhang et al., 2020). However, this is not an apple-to-apple comparison, as cloud
427 fraction in ESMs includes clouds that are optically very thin that cannot be detected by satellite passive
428 sensors or cloud radars. The comparison of high cloud fraction from simulators with the corresponding
429 satellite observations showed that E3SM slightly underestimates high clouds over most tropical deep
430 convection regions (Zhang et al., 2019; Xie et al., 2018; Rasch et al., 2019). Unfortunately, ground-based
431 radar simulator of cloud vertical profiles is not available in the current model, which prevents a direct
432 apple-to-apple comparison. Thus, caution should be taken when comparing magnitude of cloud fraction
433 from direct model output and radar measurements. Here we focus on the temporal variabilities (diurnal
434 and seasonal cycles) and the occurrence frequency distribution of cloud fraction, which are less relevant
435 to the detection threshold of cloud radars.

436 At SGP, observations show formation of low clouds in the afternoon and in late winter through
437 springtime. High clouds peak overnight into the early morning and in the spring to summer,
438 corresponding to nocturnal deep convective systems common over SGP (Tang et al., 2022b; Tang et al.,
439 2021; Jiang et al., 2006). These features are reasonably well represented in E3SMv2, although low-level
440 cloud deepening in the afternoon is not well predicted, and high-level clouds peak in the late rather than
441 early morning. At ENA, marine stratus or stratocumulus clouds occur in any month and at any time of the
442 day, but with less frequency in late summer and in afternoon. High clouds are more frequent in winter
443 months than in summer months and occur throughout the diurnal cycle with a slight mid-day minimum.
444 These features are well captured by E3SMv2. At both sites, high clouds usually occur with high fraction
445 ($> 95\%$) while low clouds are more likely associated with small fraction ($< 5\%$) (bottom row). At SGP,
446 high occurrence of low cloud fraction extends vertically up to the tropopause, representing frequently
447 occurring deep convection. At ENA, low clouds have less vertical extension but are more likely to expand
448 to greater fraction. E3SMv2 reproduces these cloud features in occurrence frequency.

449   **5.2. Multi-variable relationships related to ACI**

450 The effective radiative forcing due to ACI processes are complex, nonlinear, and highly uncertain despite
451 their significant impact on climate. ACI studies are usually conducted by examining relationships
452 between aerosols, clouds, and radiation variables that are known to interact with one another. Given so
453 many variable combinations related to ACI, ESMAC Diags v2 provides a framework for users to examine
454 relationships between the variables they choose with joint histograms, scatter plots and heatmaps. Here
455 we show a few examples to assess relationships between CCN, N_d, LWP, and top of atmosphere (TOA)
456 albedo. ESMAC Diags v2 calculate layer-mean N_d from three sources: integrated vertically from native
457 model output, retrieved using Ndrop algorithm and using Nd_sat algorithm, as shown in Table 3. In this
458 study we only show the ACI diagnostics using native model output, as it reveals the "true" ACI relations
459 in the model. Users can choose to use the retrieved N_d in their studies for their purposes.

460 The dependence of TOA albedo on CCN number concentration for stratiform warm clouds can be
461 decomposed (e.g., following Quaas et al. (2008)) as:

462
$$\frac{dA}{dlnCCN} = \left(\frac{\partial A}{\partial lnN_d} + \frac{\partial A}{\partial lnLWP}\frac{dlnLWP}{dlnN_d}\right)\frac{dlnN_d}{dlnCCN}$$
(1)

463 which allows isolation of "Twomey effect" ($\frac{\partial A}{\partial lnN_d}$) ($\frac{dlnN_d}{dlnCCN}$) and "LWP adjustment" ($\frac{dlnLWP}{dlnN_d}$) associated

464 with specific ACI processes. Here we use joint histograms and heatmaps to evaluate each component,

465 $\frac{dlnN_d}{dlnCCN}$, $\frac{dlnLWP}{dlnN_d}$, $\frac{\partial A}{\partial lnN_d}$ and $\frac{\partial A}{\partial lnLWP}$ based on long-term ground and satellite measurements at SGP (2011-

466 2020) and ENA (2016-2018) sites. The analysis in this section (except Figure 11) is limited to overcast

467 (cloud fraction > 90%), low-level (cloud top height < 4 km) liquid (ice water path < 0.01 mm) clouds.

468 Since there is no direct measurement of cloud base CCN concentration from remote sensors, surface CCN

469 concentration is used in this study and only clouds that are most likely to be affected by surface

470 conditions are examined. These clouds are identified as having cloud base potential temperature minus

471 surface potential temperature smaller than 2 K. For satellite measurements, samples with solar zenith

472 angle greater than 65° are removed to avoid N_d retrieval biases (Grosvenor et al., 2018). The sample

473 number of (ground, satellite, E3SM) for overcast low-level liquid clouds are (1766, 1217, 6369) at SGP

474 and (3450, 1345, 2884) at ENA, respectively. To increase sample size for more robust statistics, satellite

475 retrievals and E3SM outputs over a 5°×5° domain centered on SGP and ENA sites are included. This

476 increases the sample number to (1766, 71942, 15231) at SGP and (3450, 104260, 28184) at ENA.

477 Analyses of all-sky conditions and overcast low-level liquid clouds for a single grid point over each site

478 are shown in Figures S2-S7 in the supplementary material. Increasing sample domain for satellite and

479 E3SM data does not change the overall statistics shown here.

480 The change of N_d in response to a change of surface CCN number concentration ($\frac{dlnN_d}{dlnCCN}$) is heavily

481 influenced by processes such as aerosol activation. Figure 10 shows the joint probability density function

482 (PDF)of N_d and surface CCN number concentration at 0.1% supersaturation normalized within each CCN

483 bin. Ground and satellite observations show similar linear fit of $lnN_d - lnCCN$ relation, although ground-

484 based plots have much smaller sample number. E3SMv2 shows more sensitive N_d – CCN relationships

485 than observations at both SGP and ENA sites, with the relationship tighter at ENA and more scattered at

486 SGP. As a cross validation, Figure 11 shows the N_d – CCN relationships from short-term aircraft

487 campaign during HI-SCALE and ACE-ENA. The comparison with in-situ aircraft measurements

488 confirms that E3SMv2 has more sensitive N_d to CCN relationship than observations. These results

489 indicate that aerosol activation in E3SMv2 may be too weak in low CCN conditions and too strong in

490 high CCN conditions, which may be related to the differences in simulated and observed updraft velocity

491 and supersaturation (Varble et al., 2023). Note that E3SMv2 produces a significant number of small N_d (<

492 20 cm^{-3}) samples (Figure 11). This feature is reported in Golaz et al. (2022) and is partially removed by

493 setting a minimum threshold of $N_d = 10$ cm^{-3}. However, as seen in Figure 11, there are still a large

494 number of N_d between 10 and 20 cm^{-3}. Further investigation is underway to diagnose the causes of the

495 abundant low-N_d values. The diagnostics shown here indicate that a more physical method should be

496 applied to improve the simulated N_d.
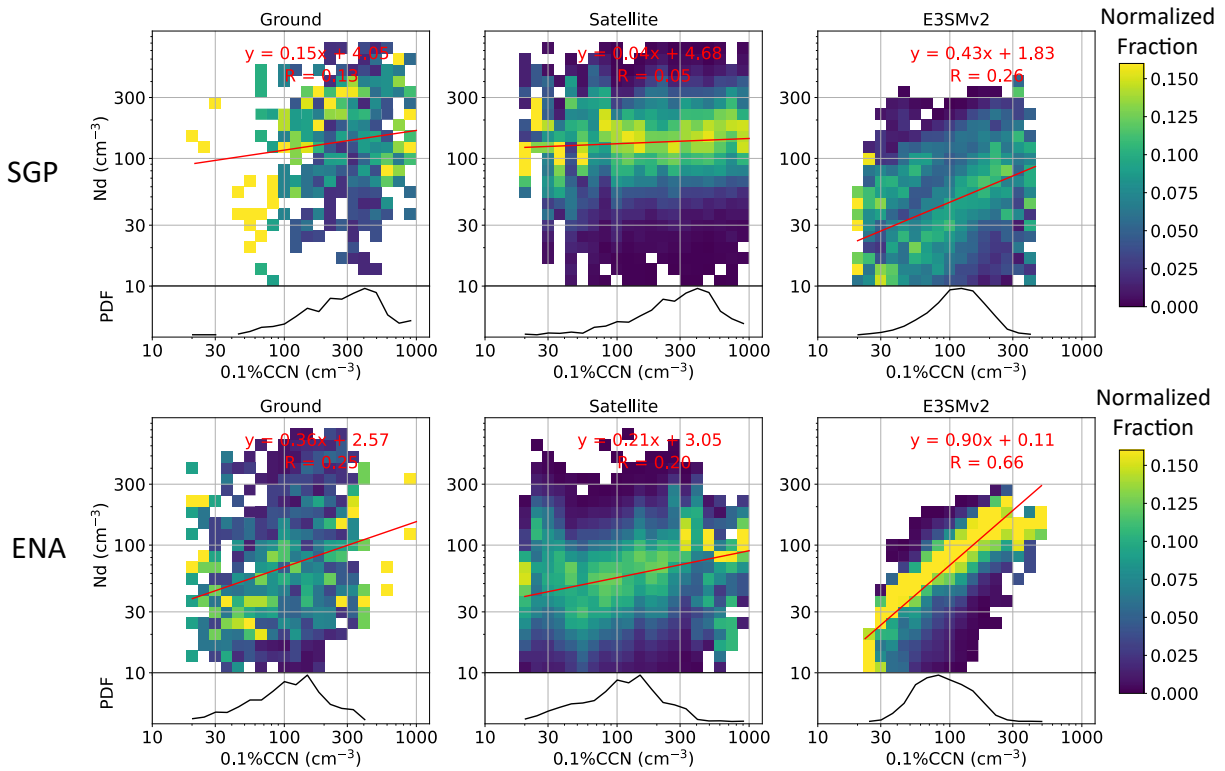
Figure 10: Joint histogram of layer-mean N_d versus surface CCN number concentration at 0.1% supersaturation, normalized within each CCN number concentration bin (PDF of CCN shown in the bottom of each panel). Samples are constrained to likely surface-coupled, overcast low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path < 0.01 mm and potential temperature difference between cloud base and surface < 2 K). Available samples within a 5°×5° region centered on SGP (top) and ENA (bottom) for satellite and E3SMv2 datasets are included. Linear fits and R values are shown in red.
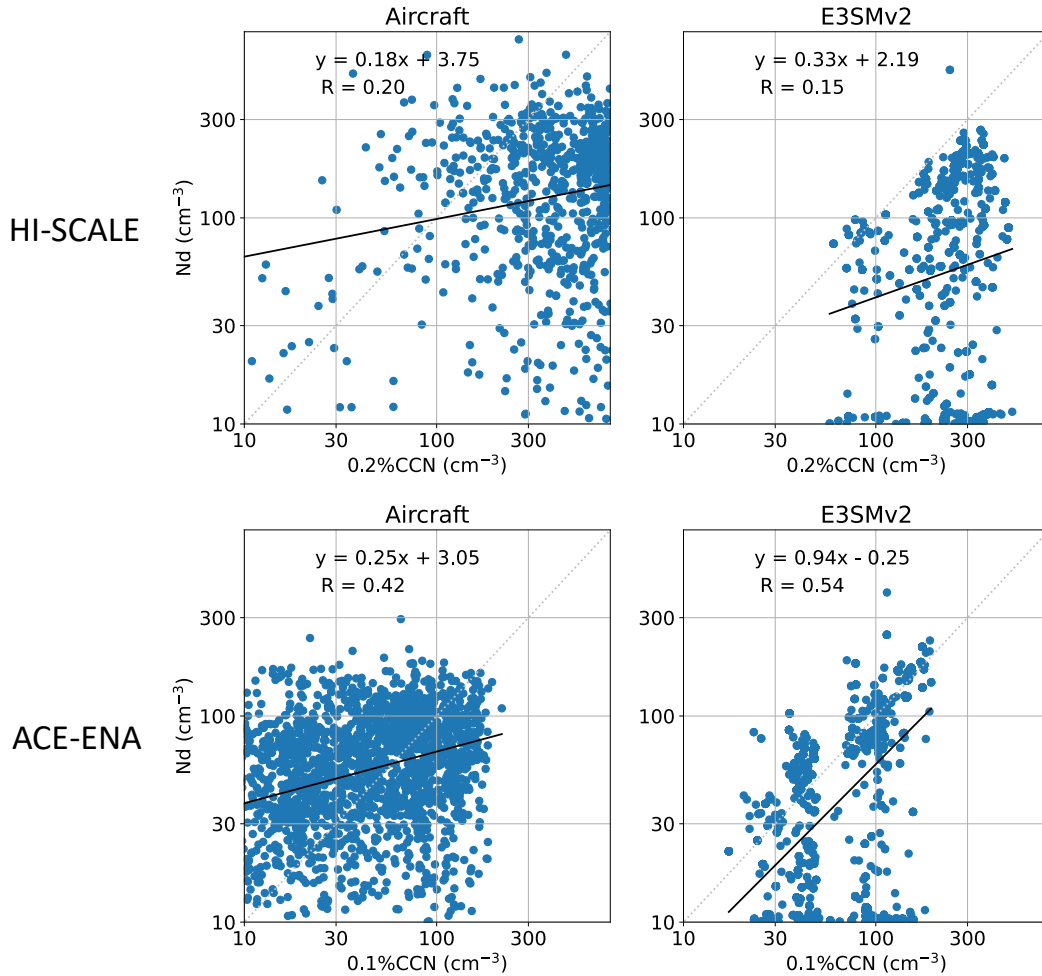
Figure 11: Scatter plots for N_d versus CCN along the flight tracks from (top) HI-SCALE and (bottom) ACE-ENA campaigns. Note that CCN number concentration measurements are taken under ~0.2% supersaturation for HI-SCALE and under ~0.1% supersaturation for ACE-ENA. Linear fits and R values are shown in each panel. R = 0.34 (SGP) and 0.74 (ENA) for E3SMv2 if a minimum $N_d = 20$ cm^{-3} is applied.

The term $\frac{dlnLWP}{dlnN_d}$ is commonly interpreted as the response of LWP to a perturbation in N_d tied to suppression of precipitation (increase LWP) or enhancement of evaporation (decrease LWP) (e.g., Glassmeier et al., 2019). Gryspeerdt et al. (2019) show that the satellite retrieved LWP over ocean increases with N_d when $N_d < \sim30\ cm^{-3}$ and decreases when $N_d > \sim30\ cm^{-3}$. This relation is also seen in satellite retrievals at ENA (Figure 12) when using a higher threshold $N_d = 50\ cm^{-3}$ to perform linear fits (black dashed lines). The linear fit is insignificant for $N_d < 50\ cm^{-3}$ in surface retrievals at both sites, partly due to small sample number, and also potentially related to drizzle contamination of LWP. The slope of the LWP $- N_d$ relation in satellite retrievals at SGP is positive for both N_d ranges. This is opposed to slope shown in the ground retrievals and indicates that retrieval biases may cause opposite

23

523 results in ACI studies. The reason why satellite retrievals show positive LWP – N_d relation at SGP is
524 subject to further investigation.

525 The E3SMv2 simulated LWP – N_d relation is quite different from satellite retrievals at both sites. At
526 SGP, it generates a positive slope for $N_d < 50\ cm^{-3}$, and a negative slope for $N_d > 50\ cm^{-3}$. At ENA, it
527 shows an opposite relation, with LWP decreases for small N_d and increases for large N_d. We examined a
528 few other oceanic regions with frequent stratus or stratocumulus clouds in E3SMv2 and saw similar
529 behavior (not shown). However, LWP – N_d relation in E3SMv1 performs quite differently, as shown in
530 Varble et al. (2023). The causes of the different LWP – N_d relation behaviors in E3SM are under further
531 investigation. Varble et al. (2023) discussed potential physical mechanisms that may affect the different
532 LWP responses to N_d in observation and simulation, such as different atmospheric states in E3SM and
533 observations. Our user-friendly diagnostics package allows these analyses to be routinely performed for
534 the purpose of better understanding critical model behaviors at process- and mechanistic-levels, providing
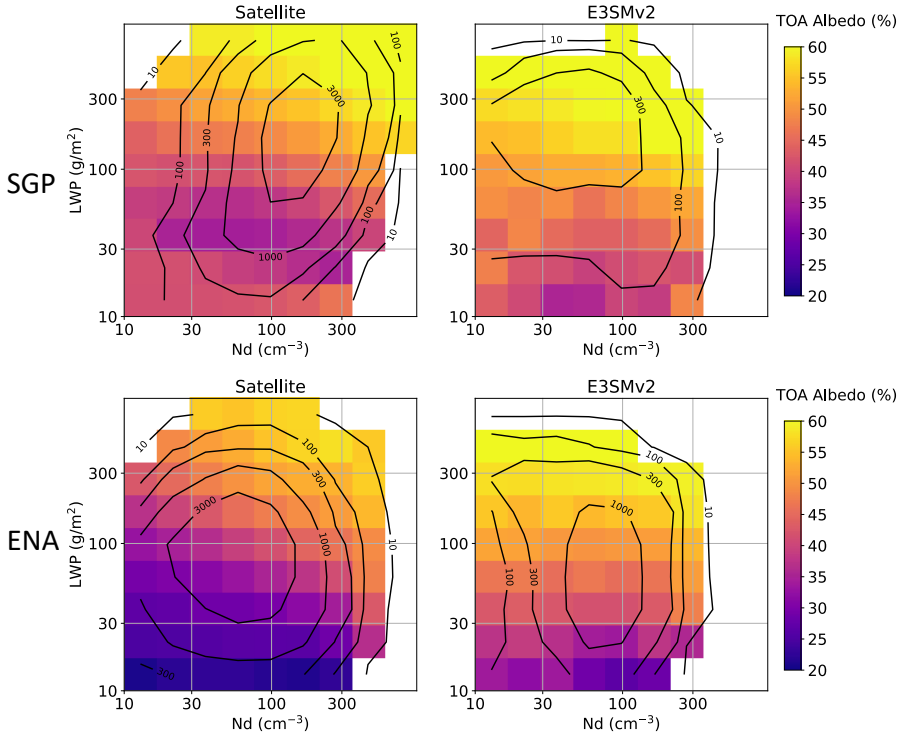535 observational constraints to facilitate model development efforts.

536
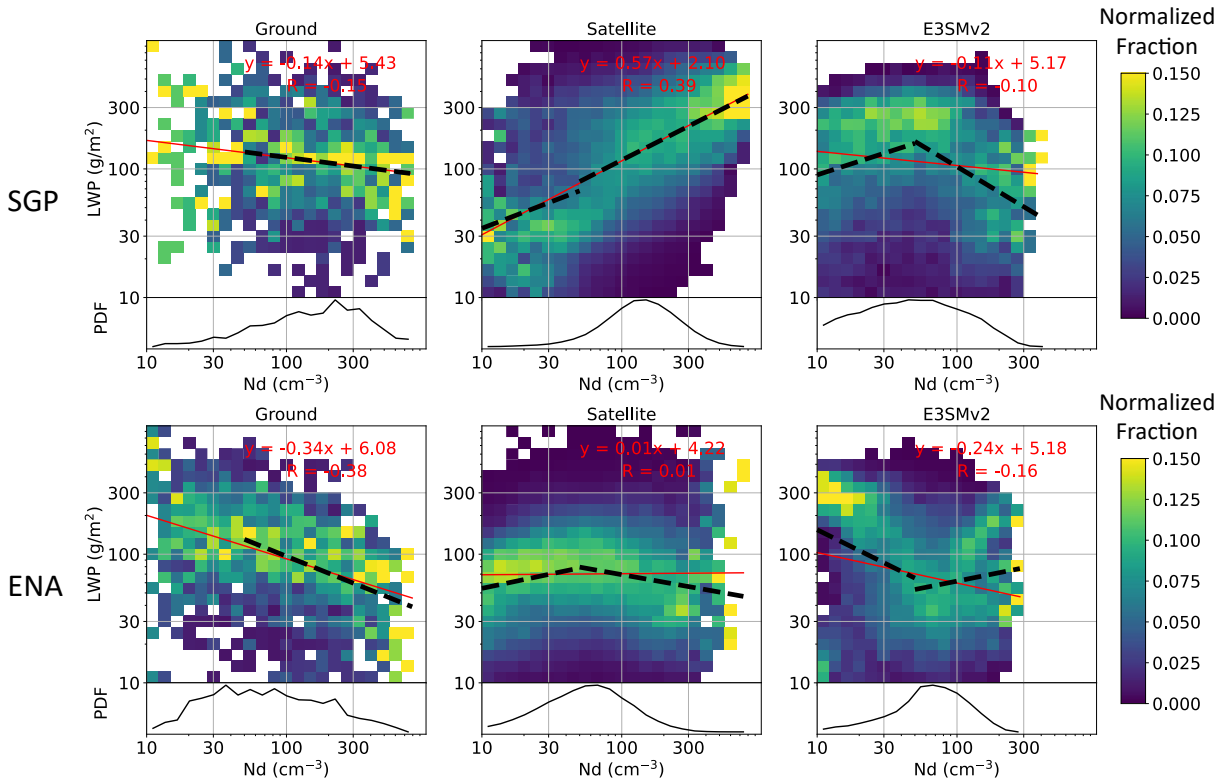
538 Figure 12: Following Figure 10, but for the N_d bin-normalized joint histogram of LWP
539 versus N_d. Red lines and equations are linear fits for all data samples and black dashed
540 lines are linear fits for $N_d < 50\ cm^{-3}$ and $N_d > 50\ cm^{-3}$ when the fits are statistically
541 significant (p < 0.01).

542

Figure 13: Heatmaps of mean TOA albedo versus LWP and N_d for likely surface-coupled, overcast low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path < 0.01 mm and potential temperature difference between cloud base and surface < 2 K). Data include samples within a 5°×5° region centered on SGP (top) and ENA (bottom). Valid sample number is shown in black contour lines. Grids with valid sample number < 10 are not filled. Ground data is not included, since the TOA albedo is not available.

Figure 13 shows heatmaps of mean TOA albedo with respect to LWP and N_d from which $\frac{\partial A}{\partial lnN_d}$ and $\frac{\partial A}{\partial lnLWP}$ can be derived. At both ENA and SGP, TOA albedo generally increases with increases of LWP and N_d, except at SGP when LWP is small. The increasing albedo in small LWP may be due to retrieval artifact as uncertainty becomes large when LWP is small (e.g., < 20 g/m²), solar zenith angle is large (e.g., > 55°), or cloud optical depth is small (e.g., <5) (Grosvenor et al., 2018). In most LWP-N_d bins, TOA albedo at SGP is generally higher than at ENA, which is expected for clouds with smaller droplet sizes. Increasing TOA albedo with increases of LWP is also seen in E3SMv2, but the dependence with N_d is weak. This can be impacted by correlation between solar zenith angle and N_d in E3SM simulation, as discussed in Varble et al. (2023). For a given LWP and N_d, TOA albedo is generally higher in E3SMv2 than in satellite observations, indicating that shallow clouds may be too reflective in the model, possibly due to smaller cloud R_{eff} (Figure 8).

The above illustration of single-variable and multi-variable diagnostics present examples to demonstrate the capability of ESMAC Diags v2. More analyses, such as selecting other variables, performing additional data filtering or treatments, and examining ACI relationships with other variable combinations, can be conducted through user-specified settings. A detailed user guide and a collection of example

25

564   scripts are included in the diagnostics package to assist users design customized diagnostics suited to their
565   specific needs.

5. Summary

567   We developed the Earth System Model aerosol-cloud diagnostics package (ESMAC Diags) to facilitate
568   routine evaluation of aerosols, clouds and ACI in the U.S. DOE's E3SM model using multiple platforms
569   of observations. As an updated version of ESMAC Diags v1 (Tang et al., 2022a) which mainly focuses on
570   aerosol properties, this paper described ESMAC Diags v2 that focuses on both aerosols, clouds, as well as
571   their interactions. In addition to the short-term field campaigns included in ESMAC Diags v1, long-term
572   diagnostics from two permanent ARM sites (SGP and ENA, each represents continental and maritime
573   conditions, respectively) are now conducted to provide more robust evaluation. The newly added multi-
574   variable joint histograms, scatter plots and heatmaps allow users to examine correlations between
575   variables that are relevant to the study of ACI.

576   Ground- and ship-based aerosol measurements are frequently impacted by local-scale emissions sources
577   such as those from airport or ship exhaust. These local sources are not resolved by coarse-resolution
578   ESMs, which usually represent an environment averaged within a region of tens to hundreds of kilometers
579   in size. In ESMAC Diags, we used available contamination-removed aerosol data, such as those from
580   Gallo et al. (2020) for ENA, and Humphries (2020) for MARCUS, and applied data filtering for other
581   field campaigns. The observations are harmonized into a uniform data format and temporal resolution that
582   are comparable with ESMs. Aircraft measurements retain higher resolution (currently 1-min) to preserve
583   high spatiotemporal variability, although ESMs have to be downscaled for evaluation with aircraft
584   measurements. This limitation of scale mismatch must be accepted to perform evaluation in current
585   coarse-resolution ESMs. Nevertheless, as ESM grid spacing approaches a few kilometers via regional
586   refinement (Tang et al., 2019) or global convection-permitting configuration (Caldwell et al., 2021), the
587   scale inconsistency between models and observations is reduced. ESMAC Diags can easily adjust the
588   preprocessing output resolution to facilitate the evaluation of high-resolution model output.

589   Cloud microphysical properties heavily rely on remote sensing measurements to achieve more robust
590   sampling, with imperfect retrieval algorithms needed to estimate these variables. Microphysical retrievals
591   are more uncertain than typical atmospheric state measurements due to the need for many assumptions
592   related to cloud dynamical and physical processes. We have shown (in Section 3) that ground- and
593   satellite-based retrievals of N_d and R_{eff} are overall consistent with each other and with in-situ aircraft
594   measurements, with some systematic differences such as smaller N_d and larger R_{eff} in satellite retrievals.
595   The discrepancies between different retrievals can be larger for individual days (e.g., Figure S1) but can
596   be mitigated to some degrees when considering broader statistics (Figures 3 and 4). The usage of multiple
597   retrieval datasets is critical to understand the robustness of evaluation results, as the spread between
598   different datasets indicates how robust model-observation differences are and guides interpretations of
599   model biases to support model development.

600   Finally, this paper presents a few examples of how well E3SMv2 simulates aerosols, clouds and ACI. We
601   showed that ESMAC Diags can be used to target further investigation into specific parameterization
602   components. For example, the analysis of N_d – CCN correlation indicates that E3SMv2 may exhibit too
603   weak aerosol activation in low CCN conditions and too strong in high CCN conditions; the analysis of
604   LWP – N_d correlation indicates that either the precipitation suppression and cloud evaporation

605 mechanisms are not well represented, or there are other mechanisms dominating LWP $- N_d$ correlation in
606 E3SMv2. These diagnostic analyses provide insights into areas in aerosols, clouds and ACI that warrant
607 special attention in future model development efforts. As ESMs continuously improve its physical
608 parameterizations, resolution, and numerical schemes, ESMAC Diags offers a valuable tool for
609 systematically evaluating the performance of the newer versions of a model in simulating aerosol, clouds
610 and ACI.

Code availability:

The current version of ESMAC Diags is publicly available through GitHub (https://github.com/eagles-project/ESMAC_diags) under the new BSD license. The exact version (2.1.2) of the code used to produce the results used in this paper is archived on Zenodo (https://doi.org/10.5281/zenodo.7696871). The model simulation used in this paper is version 2.0 (https://doi.org/10.11578/E3SM/dc.20210927.1) of E3SM.

Data availability:

Measurements from the HI-SCALE, ACE-ENA, MAGIC, and MARCUS campaigns as well as the SGP and ENA sites are supported by the DOE Atmospheric Radiation Measurement (ARM) user facility and available at https://adc.arm.gov/discovery/. Measurements from the CSET and SOCRATES campaigns are supported by National Science Foundation (NSF) and obtained from NCAR Earth Observing Laboratory at https://data.eol.ucar.edu/master_lists/generated/cset/ and https://data.eol.ucar.edu/master_lists/generated/socrates/, respectively. DOI numbers or references of individual datasets are given in Tables S1-S8. All the preprocessed observational and model data used to produce the results used in this paper is archived on Zenodo (https://doi.org/10.5281/zenodo.7478657).

Author contribution:

ST, JDF and PM designed the diagnostics package; ST and ACV wrote the code and performed the analysis; PW, XD, FM and MP processed the field campaign datasets and provided discussions on the data quality issues; KZ contributed to the model simulation; JCH contributed to the package design and setup; ST wrote the original manuscript; all authors reviewed and edited the manuscript.

Competing interests:

Po-Lun Ma is a Topical Editor of Geoscientific Model Development. Other authors declare that they have no conflict of interest.

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