Table S1: data used for HI-SCALE

Platform	Instrument	Measurements	Datastream name	DOI or link
Ground	Aerosol chemical speciation monitor (ACSM)	Aerosol composition	sgpaosacsmC1.b1	DOI: 10.5439/1762267
	Scanning mobility particle sizer (SMPS)	Aerosol size distribution (20-700 nm)	sgpaossmpsS01.a1 (IOP1) and shilling-smps (IOP2)	DOI: 10.5439/1476898
	Nano scanning mobility particle sizer (nanoSMPS)	Aerosol size distribution (2-150 nm)	sgpaosnanosmpsS01.a1	DOI: 10.5439/1242975
	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	sgpaosuhsasS01.a1	DOI: 10.5439/1333828
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	sgpaoscpcC1.b1	DOI: 10.5439/1025152
	Condensation particle counter – ultrafine (CPCU)	Aerosol number concentration (> 3 nm)	sgpaoscpcuS01.b1	DOI: 10.5439/1046186
	Cloud condensation nuclei (CCN) counter	CCN number concentration	sgpaosccn1colavgC1.b1	DOI: 10.5439/1342133
	Multifilter rotating shadowband radiometer (MFRSR)	Cloud optical depth and effective radius	sgpmfrsrcldod1minC1.c1	DOI: 10.5439/1027296
	Cloud droplet number concentration retrieval (Ndrop)	Cloud droplet number concentration	sgpndropmfrsrC1.c1	DOI: 10.5439/1131339
	Active remote sensing of clouds (ARSCL)	Cloud base height, cloud top height	sgparsclkazrbnd1kolliasC1.c0	DOI: 10.5439/1393438
	Surface meteorological station (MET)	Temperature, relative humidity, wind, pressure, precipitation	sgparmbeatmC1.c1	DOI: 10.5439/1333748
	Radiosonde	Temperature, relative humidity, wind		
	Cloud radar, lidar, ceilometer	Cloud fraction	sgparmbecldradC1.c1	DOI: 10.5439/1333228
	Total sky image (TSI)	Cloud fraction		
	Surface radiation measurement	Longwave and shortwave radiation		
	Microwave radiometer (MWR)	Liquid water path		
Satellite	Geostationary satellite-based retrievals using Visible Infrared	TOA shortwave and longwave radiation; cloud fraction; height, pressure and temperature at cloud	sgpvisstgridg13v4minnisX1.c1 sgpvisstpx2dg13minnisX1.c1	https://adc.arm.gov/discovery/#/results/s::sgpvisstgridg13v4minnisX1.c1

	Solar-Infrared Split Window Technique (VISST) algorithm	top; liquid water path; cloud optical depth; droplet effective radius		
Aircraft	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	tomlinson-uhsas	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/tomlinson-uhsas
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	mei-cpc	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/mei-cpc
	Condensation particle counter – ultrafine (CPCU)	Aerosol number concentration (> 3 nm)	mei-cpc	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/mei-cpc
	Cloud condensation nuclei (CCN) counter	CCN number concentration	mei-ccn	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/mei-ccn
	Interagency working group for airborne data and telemetry systems (IWG)	navigation information and basic atmospheric state parameters	mei-iwg1	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/mei-iwg1
	Fast integrated mobility spectrometer (FIMS)	Aerosol size distribution (10 – 425 nm)	wang-fims	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/wang-fims
	Passive cavity aerosol spectrometer (PCASP)	Aerosol size distribution (120 – 3000 nm)	tomlinson-pcasp	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/tomlinson-pcasp
	Best estimate aerosol size distribution (BEASD)	Aerosol size distribution combining FIMS, PCASP, CAS, and FCDP (~10nm - 10µm)	pekour-aafbe	DOI: 10.5439/1838448
	Cloud probe merged size distribution (mergedSD)	Cloud size distribution combining FCDP, 2-DS and HVPS (1.5µm - 9075µm)	mei-merged	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/mei-merged
	High-resolution time-of-flight aerosol mass spectrometer (AMS)	Aerosol composition	shilling-ams	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/shilling-ams
	Water content measuring system (WCM)	Cloud liquid and total water content	matthews-wcm	https://iop.archive.arm.gov/arm-iop/2016/sgp/hiscale/matthews-wcm

Table S2: data used for ACE-ENA

Platform	Instrument	Measurements	Datastream name	DOI or link
Ground	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	enaaosuhsasC1.a1	DOI: 10.5439/1409033
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	enaaoscpcfC1.b1	DOI: 10.5439/1046184
	Cloud condensation nuclei (CCN) counter	CCN number concentration	enaaosccn1colavgC1.b1	DOI: 10.5439/1342133
	Aerosol chemical speciation monitor (ACSM)	Aerosol composition	enaaosacsmC1.b2	DOI: 10.5439/1762267
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	mei-cpc	DOI: 10.5439/1440985
	Multifilter rotating shadowband radiometer (MFRSR)	Cloud optical depth and effective radius	enamfrsrcldod1minC1.c1	DOI: 10.5439/1027296
	Cloud droplet number concentration retrieval (Ndrop)	Cloud droplet number concentration	enandropmfrsrC1.c1	DOI: 10.5439/1131339
	Cloud droplet number concentration retrieval by Wu et al (2020)	Cloud droplet number concentration, cloud effective radius	Wu_etal	DOI: 10.1175/jcli-d-20-0272.1
	Active remote sensing of clouds (ARSCL)	Cloud base height, cloud top height	enaarsclkazrbnd1kolliasC1.c0	DOI: 10.5439/1393438
	Surface meteorological station (MET)	Temperature, relative humidity, wind, pressure, precipitation	enaarmbeatmC1.c1	DOI: 10.5439/1333748
	Radiosonde	Temperature, relative humidity, wind		
	Cloud radar, lidar, ceilometer	Cloud fraction	enaarmbecldradC1.c1	DOI: 10.5439/1333228
	Total sky image (TSI)	Cloud fraction		
	Surface radiation measurement	Longwave and shortwave radiation		
	Microwave radiometer (MWR)	Liquid water path		

Satellite	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	TOA shortwave and longwave radiation; cloud fraction; height, pressure and temperature at cloud top; liquid water path; cloud optical depth; droplet effective radius	enavisstgridm10minnisX1.c1 enavisstpx2dm10minnisX1.c1	https://adc.arm.gov/discovery/#/results/s::enavisstgridm10minnisX1.c1 https://adc.arm.gov/discovery/#/results/s::enavisstpx2dm10minnisX1.c1
Aircraft	Condensation particle counter – ultrafine (CPCU)	Aerosol number concentration (> 3 nm)	mei-cpc	DOI: 10.5439/1440985
	Cloud condensation nuclei (CCN) counter	CCN number concentration	enaaafccn2colaF1.b1, enaaafccn2colbF1.b1	DOI: 10.5439/1349242 DOI: 10.5439/1349243
	Interagency working group for airborne data and telemetry systems (IWG)	navigation information and basic atmospheric state parameters	mei-iwg1	https://iop.archive.arm.gov/arm-iop/2017/ena/aceena/mei-iwg1
	Fast integrated mobility spectrometer (FIMS)	Aerosol size distribution (10 – 425 nm)	wang-fims	https://iop.archive.arm.gov/arm-iop/2017/ena/aceena/wang-fims
	Passive cavity aerosol spectrometer (PCASP)	Aerosol size distribution (100 – 3000 nm)	tomlinson-pcas	https://iop.archive.arm.gov/arm-iop/2017/ena/aceena/tomlinson-pcas
	Optical particle counter (OPC)	Aerosol size distribution (390 – 15960 nm)	pekour-opc_iso	https://iop.archive.arm.gov/arm-iop/2017/ena/aceena/pekour-opc_iso
	Best estimate aerosol size distribution (BEASD)	Aerosol size distribution combining FIMS, PCASP, CAS, and FCDP (~10nm - 10µm)	pekour-asdbe	DOI: 10.5439/1867870
	Cloud probe merged size distribution (mergedSD)	Cloud size distribution combining FCDP, 2-DS and HVPS (1.5µm - 9075µm)	mei-2dsfcdphvps	https://iop.archive.arm.gov/arm-iop/2017/ena/aceena/mei-2dsfcdphvps
	High-resolution time-of-flight aerosol mass spectrometer (AMS)	Aerosol composition	shilling-hrfams	Doi: 10.5439/1468474
	Water content measuring system (WCM)	Cloud liquid and total water content	matthews-wcm	Doi: 10.5439/1465759

Table S3: Instruments for MAGIC

Platform	Instrument	Measurements	Datastream name	DOI or link
Ship	Meteorological station (MET)	Temperature, relative humidity, wind, pressure	raynolds-marmet	https://iop.archive.arm.gov/arm-iop/2012/mag/magic/reynolds-marmet/
	Microwave radiometer (MWR)	Liquid water path, precipitable water vapor	magmwrret1liljclouM1.s2	DOI: 10.5439/1027369
	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	magaosuhsasM1.a1	DOI: 10.5439/1333828
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	magaoscpcfM1.a1	DOI: 10.5439/1046184
	Cloud condensation nuclei (CCN) counter	CCN number concentration	magaosccn100M1.a1	DOI: 10.5439/1227964
	Cloud droplet number concentration retrieval by Wu et al (2020)	Cloud droplet number concentration, cloud effective radius	Wu_etal	DOI: 10.1175/jcli-d-20-0272.1
Satellite	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	TOA shortwave and longwave radiation; cloud fraction; height, pressure and temperature at cloud top; liquid water path; cloud optical depth; droplet effective radius	magvisstpxg15minnisX1.c1	https://adc.arm.gov/discovery/#/results/s::magic%20visst

Table S4: Instruments for MARCUS

Platform	Instrument	Measurements	Datastream name	DOI or link
Ship	Meteorological station (MET)	Temperature, relative humidity, wind, pressure	maraadmetX1.b1	DOI: 10.5439/1593144
	Microwave radiometer (MWR)	Liquid water path, precipitable water vapor	marmwrret1liljclouM1.s2	DOI: 10.5439/1027369
	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	maraosuhsasM1.a1	DOI: 10.5439/1333828
	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	maraoscpcf1mM1.b1	DOI: 10.5439/1418260
	Cloud condensation nuclei (CCN) counter	CCN number concentration	maraosccn1colavgM1.b1	DOI: 10.5439/1342133
	Reprocessed CN and CCN	CN and CCN number concentration	MARCUS ARM CN and CCN data reprocessed to remove ship exhaust influence. v1.	DOI: 10.25919/ezp0-em87
Satellite	Geostationary satellite-based retrievals using Visible Infrared Solar-Infrared Split Window Technique (VISST) algorithm	TOA shortwave and longwave radiation; cloud fraction; height, pressure and temperature at cloud top; liquid water path; cloud optical depth; droplet effective radius	marvisstgridh08minnisX1.c1	https://adc.arm.gov/discovery/#/results/s::marvisstgridh08minnisX1.c1

Table S5: Instruments for CSET

Platform	Instrument	Measurements	Datastream name	DOI or link
aircraft	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	Low Rate (LRT - 1 sps) Navigation, State Parameter, and Microphysics Flight-Level Data. Version 1.3	DOI: 10.5065/D65Q4T96
	Condensation nuclei counter (CNC)	Aerosol number concentration (11-3000 nm)	Same as above	DOI: 10.5065/D65Q4T96
	Passive cavity aerosol spectrometer (PCASP)	Aerosol size distribution (120 – 3000 nm)	Same as above	DOI: 10.5065/D65Q4T96
	PMS-King Liquid Water Content (LWC)	Liquid water content	Same as above	DOI: 10.5065/D65Q4T96
	1DC	Cloud droplet number size distribution (12.5 – 1590 µm)	Same as above	DOI: 10.5065/D65Q4T96
	2DC	Cloud droplet number size distribution (12.5 – 1590 µm)	Same as above	DOI: 10.5065/D65Q4T96
	CDP	Cloud droplet number size distribution (2 – 50 µm)	Same as above	DOI: 10.5065/D65Q4T96

Table S6: Instruments for SOCRATES

Platform	Instrument	Measurements	Datastream name	DOI or link
aircraft	Cloud condensation nuclei (CCN) counter	CCN number concentration	SOCRATES CCN measurements. Version 1.1	DOI: 10.5065/D6Z036XB
	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	Low Rate (LRT - 1 sps) Navigation, State Parameter, and Microphysics Flight-Level Data. Version 1.3	DOI: 10.5065/D6M32TM9
	Condensation nuclei counter (CNC)	Aerosol number concentration (11-3000 nm)	Same as above	DOI: 10.5065/D6M32TM9
	PMS-King Liquid Water Content (LWC)	Liquid water content	Same as above	DOI: 10.5065/D6M32TM9
	1DC	Cloud droplet number size distribution (12.5 – 1590 µm)	Same as above	DOI: 10.5065/D6M32TM9
	2DC	Cloud droplet number size distribution (12.5 – 1590 µm)	Same as above	DOI: 10.5065/D6M32TM9
	2DS	Cloud droplet number size distribution (5 – 2565 µm)	Same as above	DOI: 10.5065/D6M32TM9
	CDP	Cloud droplet number size distribution (2 – 50 µm)	Same as above	DOI: 10.5065/D6M32TM9

Table S7: long-term measurements at SGP

Datastream name	Instrument	Measurements	Data period	DOI or link
sgpmetE13.b1	Surface meteorological station (MET)	Temperature, relative humidity, wind, pressure	20110101 – 20201231	DOI: 10.5439/1786358
sgpaoscpcC1.b1 sgpaoscpcfE13.b1	Condensation particle counter (CPC)	Aerosol number concentration (> 10 nm)	20110310 – 20160711 20161114 – 20200402	DOI: 10.5439/1025152 DOI: 10.5439/1046184
sgpaossmpsE13.b1	Scanning mobility particle sizer (SMPS)	Aerosol size distribution (20-700 nm)	20161115 – 20201231	DOI: 10.5439/1476898
sgpaosnanosmpsE13.b1	Nano scanning mobility particle sizer (nanoSMPS)	Aerosol size distribution (2-150 nm)	20161115 – 20201231	DOI: 10.5439/1635016
sgpaosuhsasE13.b1	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 – 1000 nm), number concentration	20190109 – 20200731	DOI: 10.5439/1409033
sgptdmasizeC1.b1	Tandem Differential Mobility Analyzer (TDMA)	Aerosol size distribution (12 nm - 15 μm)	20110101 – 20141120	DOI: 10.5439/1025303
sgpaosccn1colspectraC1.b1 sgpaosccn2colaspectraE13.b1	Cloud condensation nuclei (CCN) counter	CCN number concentration, spectral data	20110810 – 20170816 20170412 – 20201231	DOI: 10.5439/1342134 DOI: 10.5439/1323896
sgpaosacsmC1.b2 sgpaosacsmE13.b2	Aerosol chemical speciation monitor (ACSM)	Aerosol composition	20110101 – 20161003 20161129 – 20201231	DOI: 10.5439/1762267 DOI: 10.5439/1762267
sgpmfrsrcldod1minC1.c1	Cloud optical properties from the multifilter rotating shadowband radiometer (MFRSR)	Cloud optical depth, droplet effective radius, liquid water path, cloud fraction	20110101 – 20201231	DOI: 10.5439/1395157
sgparsclkazrbnd1kolliasC1.c1 sgparsclkazrbnd1kolliasC1.c0	Active remote sensing of clouds (ARSCL) product using Ka-band ARM zenith radars	Cloud base height, cloud top height	20110118 – 20140315 20140316 – 20201231	DOI: 10.5439/1228769 DOI: 10.5439/1393438
sgpinterpolatedsondeC1.c1	Interpolated radiosonde data	Temperature, relative humidity, wind	20110101 – 20201231	DOI: 10.5439/1095316
sgparmbeatmC1.c1 sgparmbecldradC1.c1	ARM best estimate (ARMBE) data for atmospheric variables (armbeatm) or cloud and radiation variables (armbecldrad)	Temperature, relative humidity, wind, pressure, precipitation; cloud fraction, liquid water path, radiative fluxes	20110101 – 20201231 20110101 – 20201231	DOI: 10.5439/1333748 DOI: 10.5439/1333228
sgpvisstgridg13v4minnisX1.c1* sgpvisstgridg16v4minnisX1.c1	Visible Infrared Solar-Infrared Split-Window Technique (VISST)-derived 0.5°×0.5° gridded products from GOES13 or GOES16 satellite	cloud and radiative properties	20110101 – 20171231 20180101 – 20201231	https://adc.arm.gov/discovery/#/results/s::sgpvisstgridg13v4minnisX1.c1
sgpvisstpx2dg13minnisX1.c1 sgpvisstpx2dg16minnisX1.c1	VISST-derived 2D pixel-level (4km) products from GOES13 or GOES16 satellite	cloud and radiative properties	20110101 – 20171231 20180101 – 20201231	https://adc.arm.gov/discovery/#/results/s::sgpvisstpx2dg13minnisX1.c1

*there are missing periods 20120923 – 20121015 and 20130522 – 20130530 which are filled by sgpvisstgridg15v4minnisX1.c1

Table S8: long-term measurements at ENA

Datastream name	Instrument	Measurements	Data period	DOI or link
enametC1.b1	Surface meteorological station (MET)	Temperature, relative humidity, wind, pressure	20160101 – 20181231	DOI: 10.5439/1786358
enaaoscpcfC1.b1	Fine condensation particle counter (CPCF)	Aerosol number concentration (> 10 nm)	20160101 – 20181231	DOI: 10.5439/1046184
enaaosuhsasC1.b1	Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size distribution (60 - 1000 nm), number concentration	20160101 – 20181231	DOI: 10.5439/1409033
enaaosacsmC1.b2	Aerosol chemical speciation monitor (ACSM)	Aerosol composition	20160219 – 20181231	DOI: 10.5439/1762267
enaaosccn1colspectraC1.b1	Cloud condensation nuclei (CCN) counter	CCN number concentration, spectral data	20160623 – 20181231	DOI: 10.5439/1342134
enamfrsrcldod1minC1.c1	Cloud optical properties from the multifilter rotating shadowband radiometer (MFRSR)	Cloud optical depth, droplet effective radius, liquid water path, cloud fraction	20160101 – 20181231	DOI: 10.5439/1027296
enaarsclkazrbnl1kolliasC1.c0	Active remote sensing of clouds (ARSCL) product using Ka-band ARM zenith radars	Cloud base height, cloud top height	20160101 – 20181231	DOI: 10.5439/1393438
enainterpolatedsondeC1.c1	Interpolated radiosonde data	Temperature, relative humidity, wind	20160101 – 20181231	DOI: 10.5439/1095316
enaarmbeatmC1.c1 enaarmbecldradC1.c1	ARM best estimate (ARMBE) data for atmospheric variables (armbeatm) or cloud and radiation variables (armbecldrad)	Temperature, relative humidity, wind, pressure, precipitation; cloud fraction, liquid water path, radiative fluxes	20160101 – 20181231	DOI: 10.5439/1333748 DOI: 10.5439/1333228
enavisstgridm10minnisX1.c1 enavisstgridm11minnisX1.c1	VISST-derived 0.5°×0.5° gridded products from Meteosat-10 or Meteosat-11 satellite	cloud and radiative properties	20160101 – 20180220 20180220 – 20181231	https://adc.arm.gov/discovery/#/results/s::enavisstgridm10minnisX1.c1
enavisstpx2dm10minnisX1.c1 enavisstpx2dm11minnisX1.c1	VISST-derived 2D pixel-level (4km) products from Meteosat-10 or Meteosat-11 satellite	cloud and radiative properties	20160101 – 20180220 20180220 – 20181231	https://adc.arm.gov/discovery/#/results/s::enavisstpx2dm10minnisX1.c1
Wu_etal Retrieval	Stratus cloud retrieval from ground-based measurements using retrieval algorithm from Wu et al. (2020)	Cloud droplet number concentration, effective radius	20160101 – 20181231	DOI: 10.1029/2019JD032205
Aerosol Mask	Masking 1-min CPC data dominated by local aerosol sources using algorithm from Gallo et al. (2020)	Aerosol number concentration and mask flag	20160101 – 20181231	DOI: 10.5194/acp-20-7553-2020
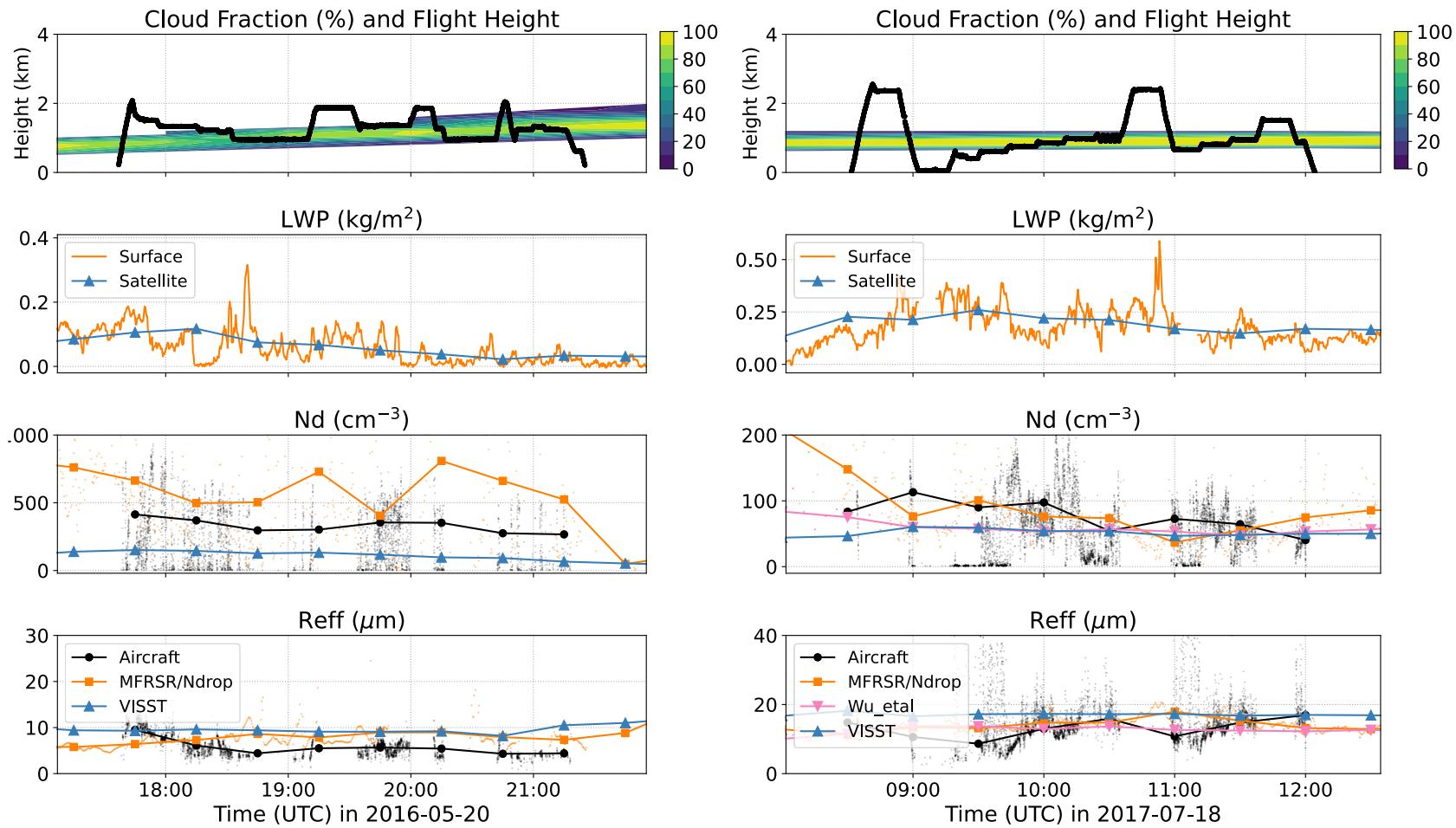
Figure S1: [from top to bottom]: Cloud fraction and aircraft height, LWP, N_d, and R_{eff} from different measurements for two cases: (left) 20 May 2016 in HI-SCALE and (right) 18 July 2018 in ACE-ENA. In N_d and R_{eff} plots, small dots are measurements/retrievals at native resolution, while solid lines with dots are values averaged into 30 minutes resolution. The orange line with square marker represents MFRSR data for R_{eff} and Ndrop data for N_d.
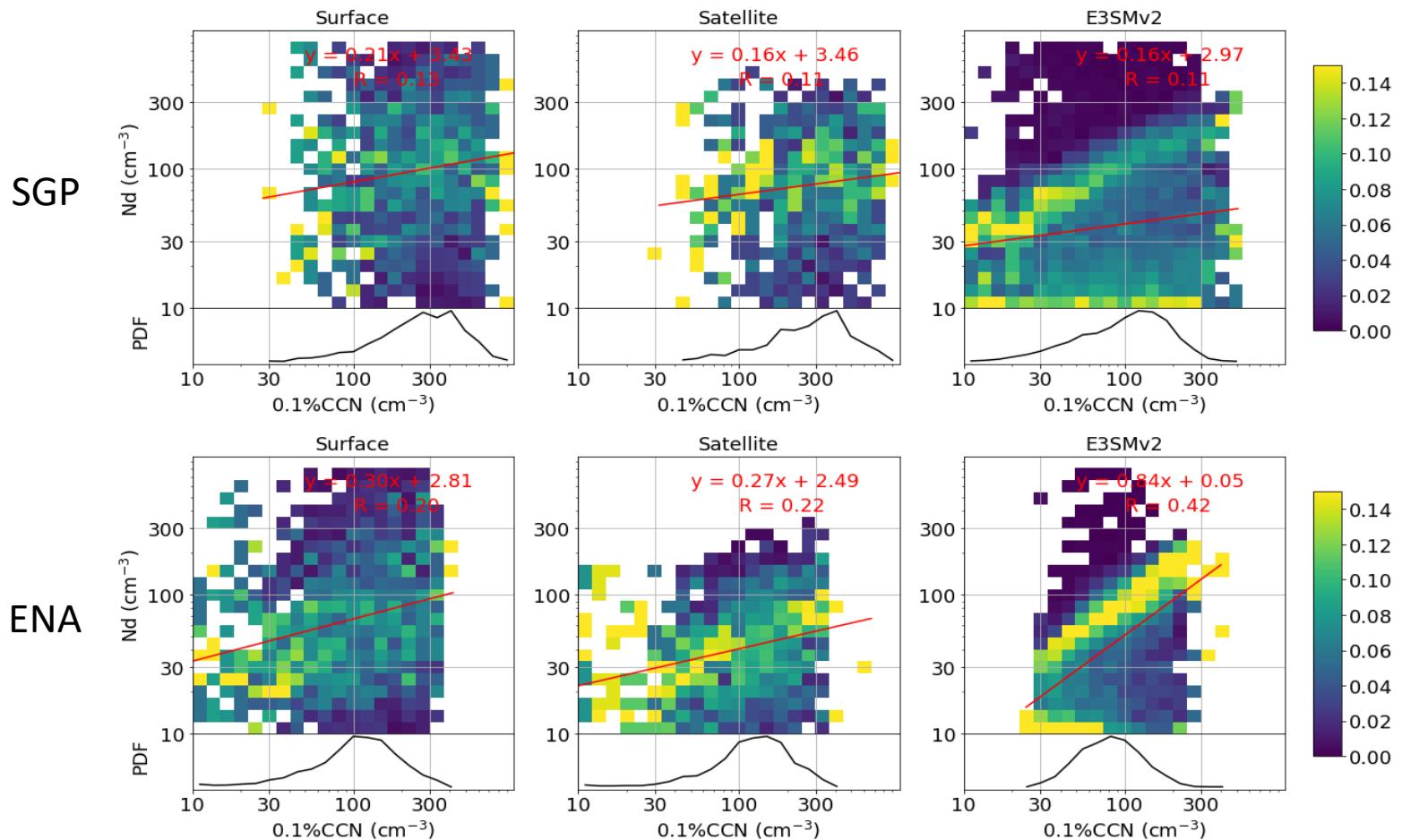
Figure S2: Joint histogram of Nd versus surface CCN concentration at 0.1% supersaturation, normalized by CCN concentration. For all available samples at (top) SGP from 2011-2020 and (bottom) ENA from 2016-2018. Linear fits and R values are shown in red.
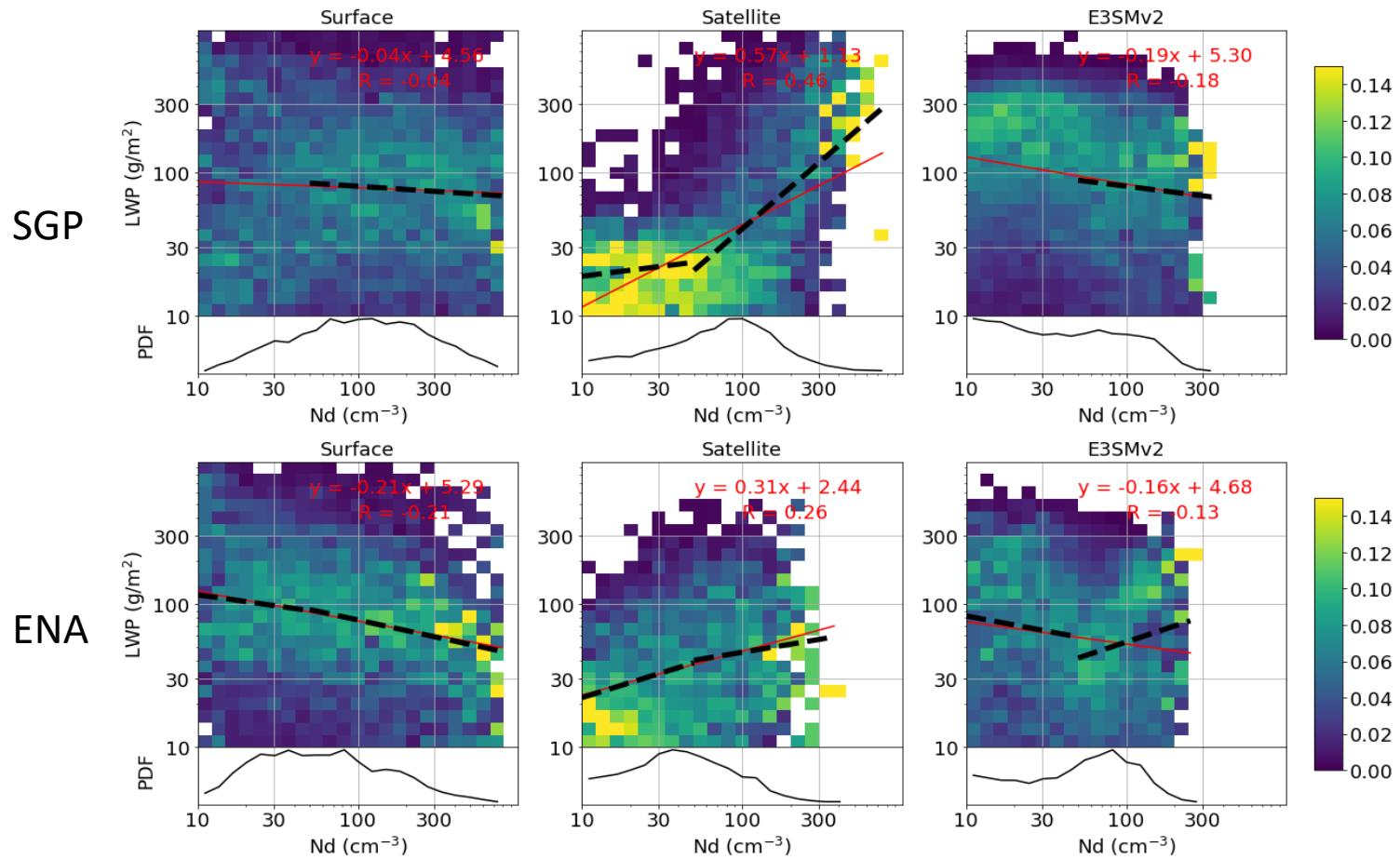
Figure S3: Joint histogram of LWP versus Nd, normalized by Nd concentration. For all available samples at (top) SGP from 2011-2020 and (bottom) ENA from 2016-2018. Red lines and equations are linear fits for all data samples and black dashed lines are linear fits for $N_d < 50\ cm^{-3}$ and $N_d > 50\ cm^{-3}$ when the fits are statistically significant (p < 0.01).
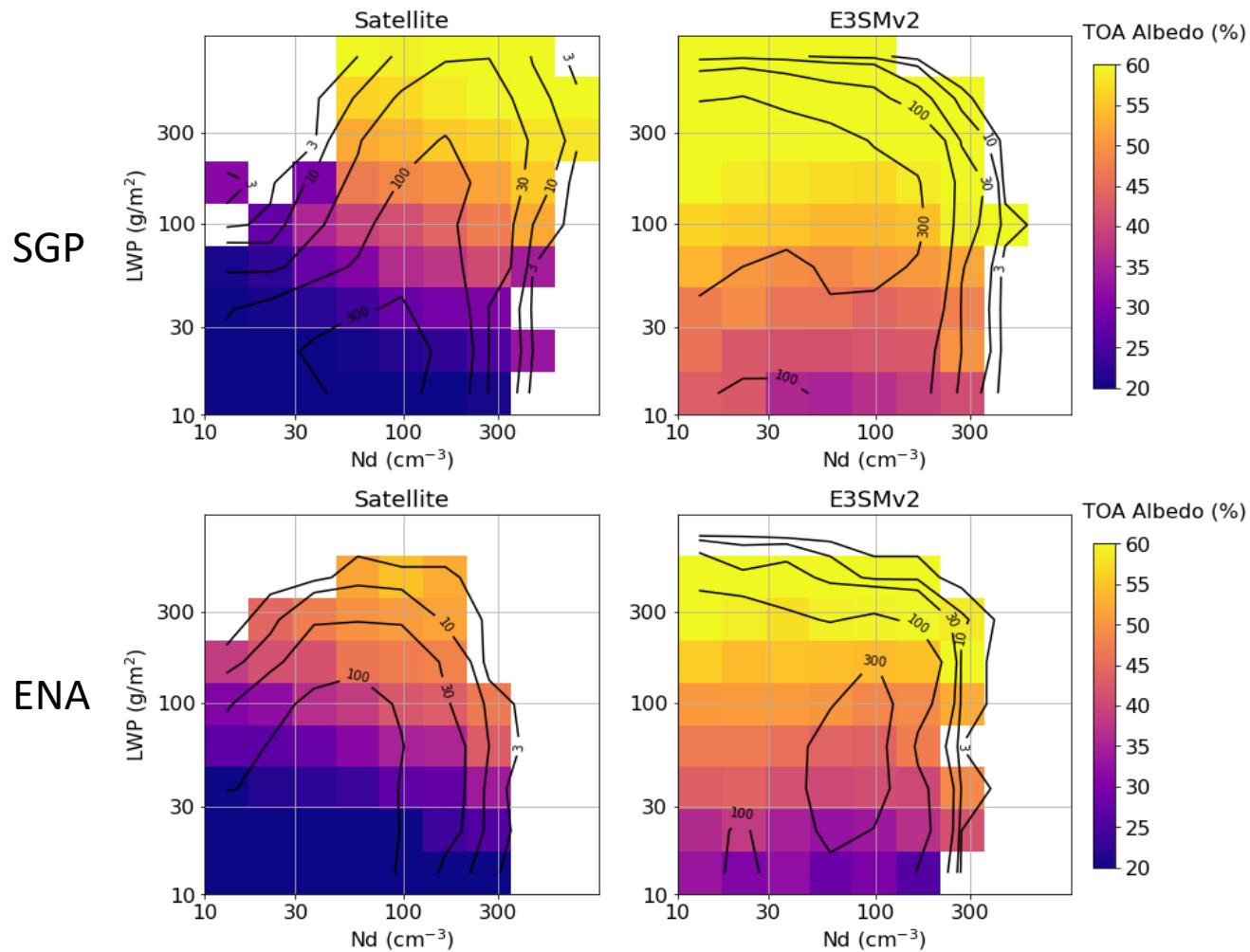
Figure S4: Heatmap of mean TOA albedo in LWP and Nd bins for all available samples at (top) SGP from 2011-2020 and (bottom) ENA from 2016-2018. Valid sample number is shown in black contour lines. Grids with valid sample number < 3 are not filled.
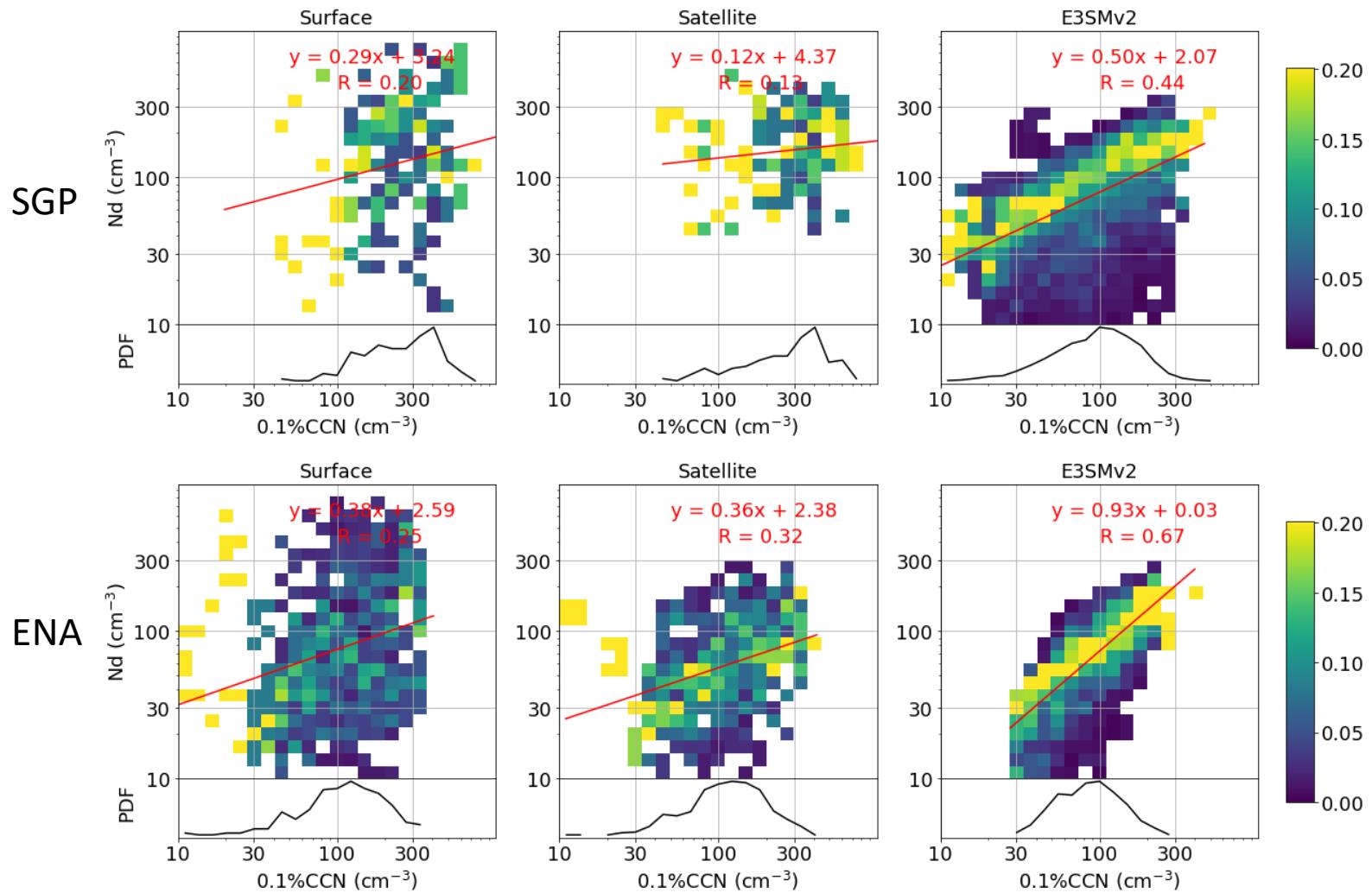
Figure S5: similar as Figure S2 but for samples of surface coupled overcast low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path < 0.01 mm and potential temperature difference between cloud base and surface < 2K).
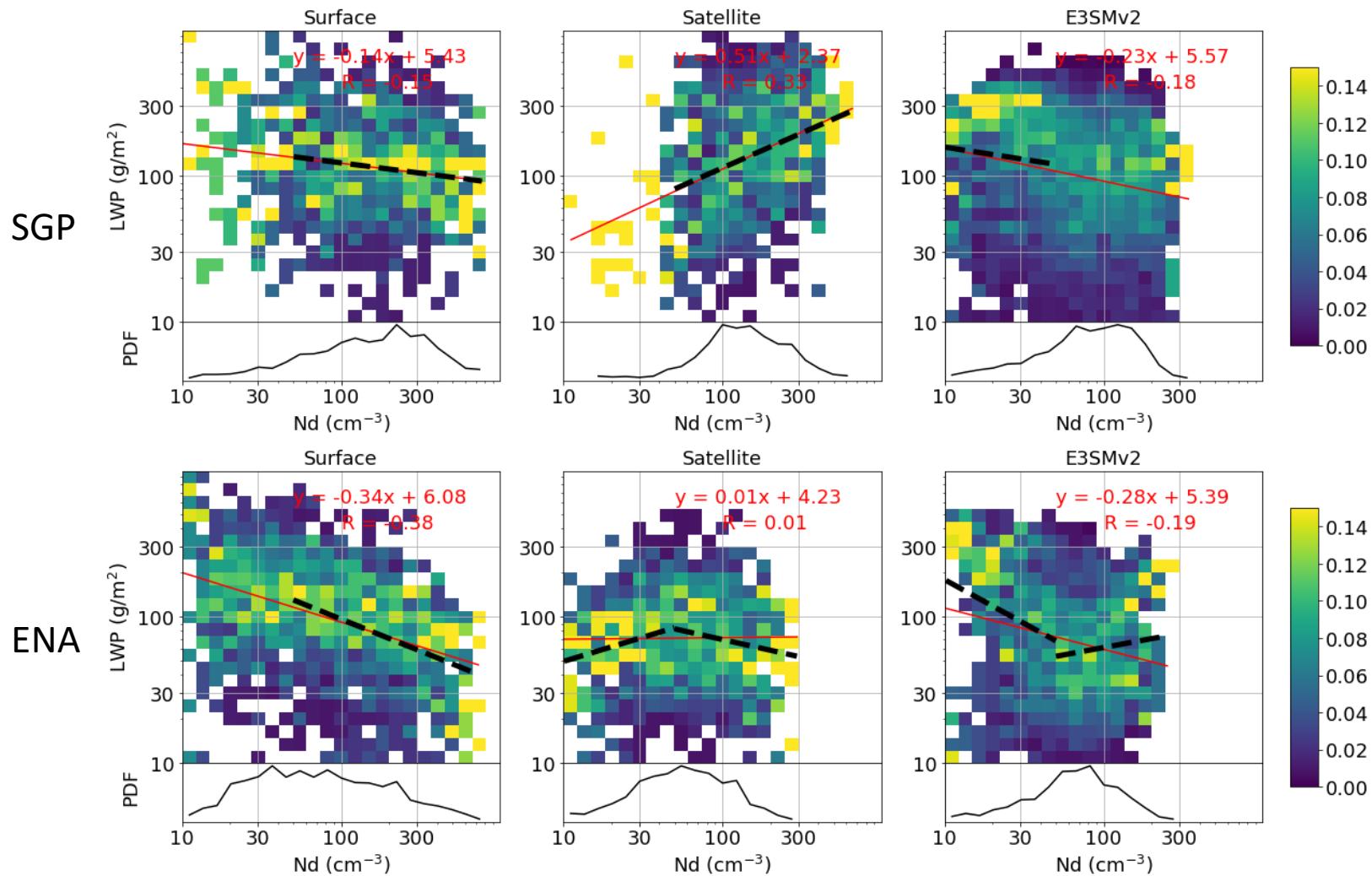
Figure S6: similar as Figure S3 but for samples of surface coupled overcast low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path < 0.01 mm and potential temperature difference between cloud base and surface < 2K).
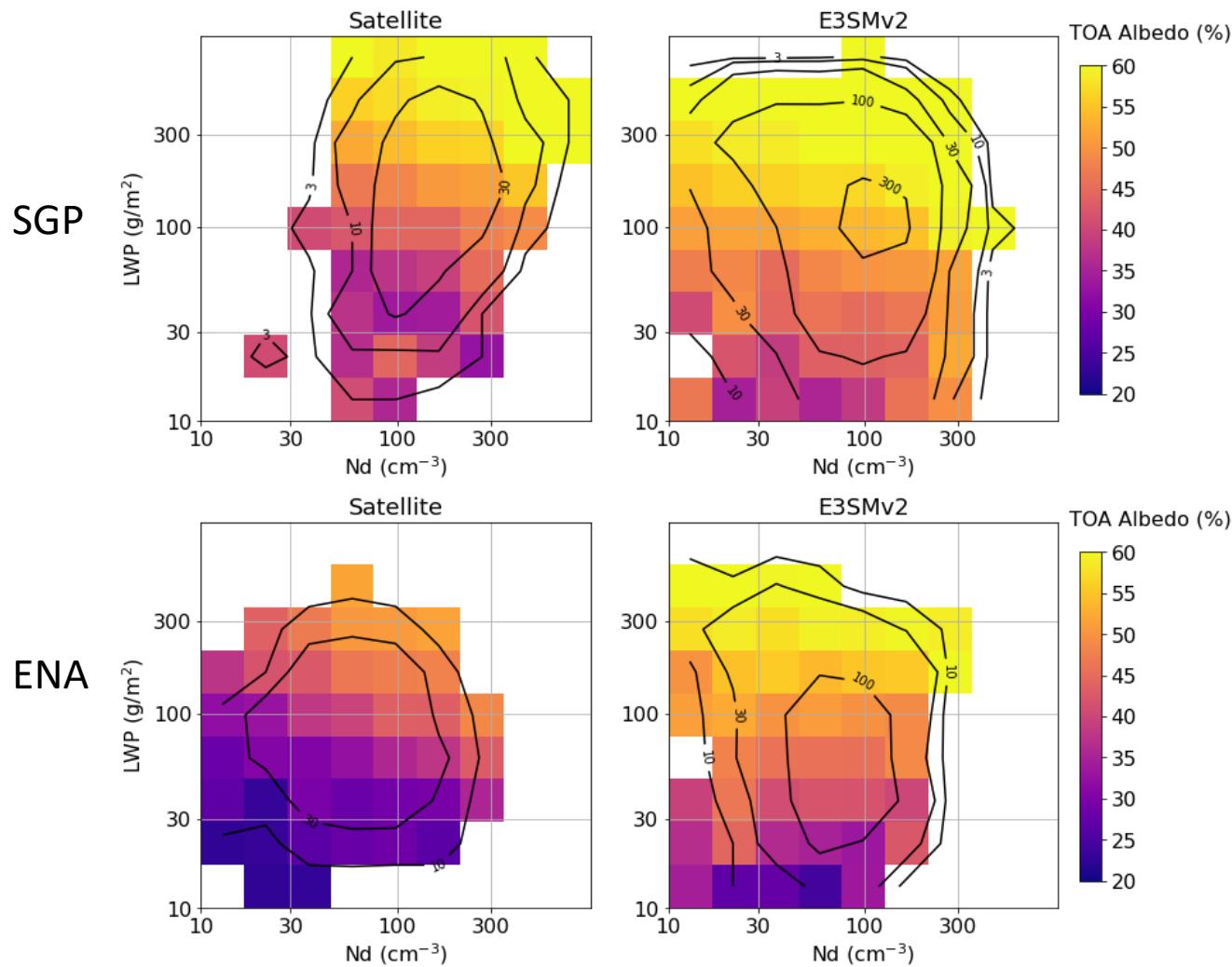
Figure S7: similar as Figure S4 but for samples of surface coupled overcast low-level liquid clouds (cloud top height < 4 km, cloud fraction > 90%, ice water path < 0.01 mm and potential temperature difference between cloud base and surface < 2K).