



- **Daytime aerosol optical depth above low-level**
- ² clouds is similar to that in adjacent clear skies
- 3 at the same heights: airborne observation

above the southeast Atlantic

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23 Abstract

24 To help satellite retrieval of aerosols and studies of their radiative effects, we demonstrate that daytime 532 nm aerosol optical depth over low-level clouds is similar to that in neighboring 25 26 clear skies at the same heights in recent airborne lidar and sunphotometer observations above the 27 southeast Atlantic. The mean AOD difference is between 0 and -0.01, when comparing the cloudy 28 and clear sides, each up to 20 km wide, of cloud edges. The difference is not statistically significant 29 according to a paired t-test. Systematic differences in the wavelength dependence of AOD and in 30 situ single scattering albedo are also minute. These results hold regardless of the vertical distance between cloud top and aerosol layer bottom. AOD aggregated over $\sim 2^{\circ}$ grid boxes for each of 31 32 September 2016, August 2017 and October 2018 also shows little correlation with the presence of 33 low-level clouds. We posit that a satellite retrieval artifact is entirely responsible for a previous 34 finding of generally smaller AOD over clouds (Chung et al., 2016), at least for the region and time of our study. Our results also suggest that the same values can be assumed for the intensive 35 36 properties of free-tropospheric biomass-burning aerosol regardless of whether clouds exist below.

37 **1. Introduction**

A significant amount of atmospheric particles are transported above liquid water clouds on the global scale (Waquet et al., 2013). Aerosols above clouds (AAC) may influence the climate in three ways. Their light absorption is amplified by cloud reflection. The heating of the atmosphere due to the absorption may stabilize the atmosphere. The particles may eventually subside, enter clouds and alter their properties. Estimates of the direct aerosol radiative effect alone see large inter-model spread for areas with large aerosol optical depth (AOD) over widespread clouds (Stier et al., 2013; Zuidema et al., 2016).

Since AAC are difficult to see from the ground or a ship, previous studies have relied on satellite observations (see Table 2 of Kacenelenbogen et al., 2019). Among them is Chung et al. (2016), which used the level 2 products of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) (Winker et al., 2009) to calculate the AOD above the maximum low-cloud-top-height in each grid cell in clear sky as well as the AOD above low clouds on a global $2^{\circ} \times 5^{\circ}$ latitude– longitude grid. Their results indicate that daytime 532 nm AOD above low clouds is generally





51 lower than that in clear sky at the same heights. The difference is up to 0.04 over the southeastern

52 Atlantic Ocean (see their Fig. 2)

53 As Chung et al. (2016) point out, it is conceivable that aerosol amounts over cloud can be 54 different from those in nearby clear sky. There are two sets of potential reasons. The first concerns the effects of meteorology. Large-scale circulation patterns paired with solar reflection from clouds 55 on aerosols could modify the horizontal and vertical extent of aerosols, aerosol concentration and 56 57 chemical composition. For example, the properties of hygroscopic aerosols might vary if the 58 relative humidity in clear skies is somehow higher than above clouds. The second set of reasons 59 pertain to the case of aerosols in close proximity to clouds. The proximity has been variously 60 defined, for example less than 100 m in the vertical direction (Costantino and Bréon, 2013) and 61 less than 20 km in the horizontal direction (Várnai and Marshak, 2018). Chung et al. (2016) note 62 that aerosols were shown to influence underlying cloud by indirect effects and semidirect effects 63 (Costantino and Bréon, 2010, 2013; Johnson et al., 2004; Wilcox, 2010) and that these aerosol-64 cloud interactions and possibly more (e.g., a pronounced if unlikely aerosol entrainment (Diamond et al., 2018)) might somehow affect the aerosol amount over cloud. A bias in the CALIOP standard 65 retrieval was also raised as a possible explanation for the Chung et al. (2016) results. The detection 66 67 threshold in the feature detection algorithm varies depending on the background lighting conditions, the atmospheric features (e.g., aerosols, high altitude cirrus or boundary layer clouds) 68 69 and the horizontal averaging required by CALIOP for detection (see Fig. 4 of Winker et al. (2009)). 70 In the particular case of aerosols above clouds, Kacenelenbogen et al. (2014) show that the 71 CALIOP standard algorithm substantially underestimates the frequency of AAC when the AOD 72 is less than ~ 0.02 . This is due mostly to tenuous aerosols with a backscatter under the detection 73 threshold; however, Kacenelenbogen et al. (2014) saw no clear bias in AOD above clouds between 74 CALIOP and the NASA Langley airborne High Spectral Resolution Lidar (HSRL-1). Liu et al. 75 (2015) show a clear AOD underestimate of the CALIOP level 2 retrieval in comparison to a separate retrieval after Hu et al. (2007) for smoke above opaque water clouds over the southeast 76 77 Atlantic, and explain this by the CALIOP layer detection scheme prematurely assigning layer base 78 altitudes and thus underestimating the geometric thickness of smoke layers. According to Chung 79 et al. (2016), the negative daytime AOD differences between cloudy and cloud-free conditions 80 "might simply be a result of systematic differences between the detection thresholds in clear sky





and above low bright clouds". The authors add that the bias may be enhanced over the ocean dueto the lower albedo compared to that of land.

The subject warrants further investigation, given the importance of AAC on climate. An 83 84 airborne experiment can help by providing direct measurements that are subject to smaller uncertainty, in finer spatial and temporal resolution albeit over limited ranges. The NASA 85 86 ObseRvations of Aerosols above CLouds and their intEractionS (ORACLES) mission was carried 87 out to study key processes that determine the climate impacts of African biomass-burning aerosols 88 above the southeast Atlantic. Of the two deployed aircraft, the NASA P3, equipped with in situ 89 and remote sensing instruments, flew in the lower- to mid-troposphere, mostly in September 2016, 90 August 2017 and October 2018. In September 2016 the NASA ER2 also flew, at about 20 km 91 altitude with downward-viewing sensors. Extensive stratocumulus clouds were observed 92 repeatedly throughout the mission; see a sample satellite image in Redemann et al. (in preparation). 93 Details of the ORACLES mission can be found in Redemann et al. (in preparation), Zuidema et 94 al. (2016) and Shinozuka et al. (2019).

The instrumentation relevant to the present paper is described in Sect. 2 along with sampling and statistical hypothesis testing methods. This is followed by comparisons of AOD and other aerosols properties above the height of cloud top between cloudy and clear skies (Sect. 3). Sect. 4 offers discussion.

992.Methods1002.1.Instrumentation

The remote sensing and in situ instruments used in this study are briefly described below
with references to full descriptions. Note that the measurements each refer to a unique vertical
range, as summarized in Table 1.

The NASA Langley Research Center High Spectral Resolution Lidar (HSRL-2), deployed from the ER2 in 2016 and from the P3 in 2017 and 2018, measures calibrated, unattenuated backscatter and aerosol extinction profiles below the instrument. The data are reported with 10 s intervals. The HSRL-2 signal-to-noise ratio is higher than that of CALIOP, due to the much lower altitude and the inverse square dependence of light intensity. In addition, by the use of a second channel to assess aerosol attenuation, the HSRL technique (Shipley et al., 1983) results in an accurate aerosol extinction product with no assumptions about lidar ratio, and also a more accurate





backscatter product, particularly in the lower atmosphere where attenuation by upper layers can
present difficulties for the spaceborne backscatter lidar. Differences in algorithm are discussed in
Sect. 4. Further details about the instrument, calibration and uncertainty can be found in Hair et al.
(2008), Rogers et al. (2009) and Burton et al. (2018).

Our analysis utilizes the HSRL-2 standard products of cloud top height (CTH), 532 nm particulate backscattering and 532 nm aerosol optical thickness (Burton et al., 2012) in three ways. First, flight segments are isolated using the CTH product (detailed in Sect. 2.2). Second, the bottom and top heights of the smoke plumes are defined with a (somewhat arbitrarily chosen) threshold backscattering coefficient at 0.25 Mm⁻¹sr⁻¹ after Shinozuka et al. (2019).

Third, we evaluate the 532 nm partial-column aerosol optical thickness from below the aircraft down to ~50 m above the CTH, (even for columns without clouds; see Sect. 2.2). The ~50m buffer is designed to reduce the ambiguity associated with the transition at the cloud top. The upper limit of the integral of extinction is 14 km altitude for the 2016 ER2 flights and 1500 m below the P3 altitude for 2017 and 2018. Profiles with possible influences of mid- and high-level clouds are largely excluded from the product, though isolated cases of thin clouds may remain.

We also use partial-column AOD observed upward from the P3 with a sunphotometer. The Spectrometer for Sky-Scanning, Sun-Tracking Atmospheric Research (4STAR) measures hyperspectral direct solar beam. Calculated AOD is reported at 1 Hz. Our analysis excludes data with possible influences of clouds above the instrument. Further details on the instrument as well as data acquisition, screening, calibration and reduction can be found in Dunagan et al. (2013), Shinozuka et al. (2013) and LeBlanc et al. (2019).

132 For 2017 and 2018, we examine a combination of the 4STAR and HSRL-2 AODs, in order 133 to cover the free troposphere both upward and downward from the aircraft that flew in it (Fig. 1). 134 The vertical coverage is compromised by two limitations intrinsic to the lidar measurements. First, 135 the CTH is not sought within 500 m of the instrument (not to be confused with the ~50-m lower buffer for the extinction integral). This means that the flight segments with clouds so close to the 136 137 aircraft enter our analysis only if the clouds extended as deep as to reach 500 m away from it. This 138 is at most a minor fraction of the data, as the fraction with the CTH within 550 m of the P3 altitude 139 is a mere 3%. Second, because of the 1500 m upper buffer for the extinction integral, we only have 140 4STAR above-P3 AOD for the flight segments when the plane was 500-1500 m above the CTH





(Fig. 1b). We add the HSRL-2 AOD to the 4STAR AOD only for the flight segments when the P3
was >1500 m above the CTH (Fig. 1c).

For 2016, we examine the HSRL-2 AOD only, because, with the lidar above the troposphere, two of the missing layers can safely be ignored, leaving the ~50 m lower buffer as the only missing layer (Fig. 1a). We refer to all these AODs from the three campaigns collectively as AOD_{ct} (see Table 1). The wavelength dependence expressed as Angstrom exponent is calculated for 10-s periods with AOD_{ct} at 355 and 532 nm both exceeding 0.1.

148 In situ aerosol instruments operated from the P3 include a nephelometer (TSI model 3563) 149 and a particle soot absorption photometer (PSAP, Radiance Research 3-wavelength version), 150 which measure particulate light scattering and absorption, respectively. After adjustments are 151 made for factors such as angular truncations (Anderson and Ogren, 1998) and filter interference 152 (Virkkula, 2010) for each wavelength, extinction coefficient and single scattering albedo at 550 153 nm are derived for an instrument relative humidity (RH) that is typically below 40%. Pistone et al. 154 (2019) and Shinozuka et al. (2019) have more details. The non-refractory masses of submicron 155 particles were measured by a time-of-flight aerosol mass spectrometer (Aerodyne, Inc HR-ToF 156 AMS, DeCarlo et al. (2006)). A condensation particle counter (TSI model 3010, with ΔT set to 157 22°C) measured the number concentration of particles larger than about 10 nm. These in situ properties refer to the air immediately outside the P3 aircraft, not a vertical column. Only the in 158 159 situ measurements in 2017 and 2018 at 500-1500 m above the CTH are used in this study.

160 **2.2.** Sampling

161 Two methods are employed for selecting subsets of the observations for analysis. In the 162 first (Sect. 2.2.1), we bundle data from areas hundreds of kilometers wide for each of the three 163 campaigns, in a manner as similar to the CALIOP-based study (Chung et al., 2016) as the airborne 164 measurements allow. In the second method (Sect. 2.2.2), we pair cloudy and clear skies with more 165 stringent spatiotemporal criteria to isolate the impact of finer-scale phenomena. Note that both 166 methods ignore time periods for which the 532 nm backscattering product (from which the CTH product is derived) is masked at all altitudes, as well as transit flights into and out of the study area. 167 168 Cases are also excluded where the CTH exceeds 3241 m. This is to be consistent with the study 169 by Chung et al. (2016), which refers to clouds at 680 hPa or higher pressure, although we find 170 similar results with or without this restriction.





171 **2.2.1.** Meso-scale monthly-mean sampling

This method separates profiles measured in the three campaigns into two groups: those concurrent with a presence of low-level clouds as reported by the HSRL-2 and those concurrent with an absence of any cloud detected by HSRL-2 in the column. The groups are each aggregated into grid boxes approximately 2° by 2°, as shown in Fig. 2. This grid is adapted from Shinozuka et al. (2019) but with additional boxes for the São Tomé-based 2017 and 2018 campaigns. In total, 109 hours and 39 hours of flight segments are selected for the cloudy and clear groups, respectively, including minor double-counting where boxes overlap.

The arithmetic mean of the CTH of the cloudy group is calculated for each day for each box and 50 m above it is set as the lowest altitude for computing AOD_{ct} for each 10 s period (Sect. 2.1). Then the arithmetic mean and standard deviation are calculated for the AOD_{ct}, as well as other measurements (Sect. 2.1, Table 1), for each group and each box. After excluding the time periods with mid- and high-level clouds and instrument/aircraft issues, 49 hours and 26 hours of the AOD_{ct} measurements enter the analysis for cloudy and clear-sky groups, respectively.

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2.2.2. Local-scale near-synchronous sampling

This method identifies cloud edges and demarcates the cloudy side and clear side of each edge based on the time series of the CTH detected by HSRL-2, for level flight legs only. Cloud edges are defined by the points in time when a cloud is detected in a profile adjacent to a profile with no cloud detection.

190 A clear sky and a cloud are represented by the time period of a certain length, say 60 s, 191 preceding each edge and the same length following it. To ensure that clear skies and clouds are 192 not interrupted for the length, we exclude edges for which another one is found within the length. 193 The longer the length, the smaller the number of cloudy-clear pairs, because longer continuous 194 clouds and clear skies are rarer. Furthermore, we set another length, 20 s in the example illustrated 195 in Fig. 3a, to exclude immediately before and after the edge, in order to reduce ambiguity 196 associated with a gradual transition from cloud droplets to unactivated particles, the so-called 197 twilight zone (Koren et al., 2007; Schwarz et al., 2017; Várnai and Marshak, 2018). We convert 198 the temporal dimensions into horizontal ones using the mean true horizontal aircraft speed, 200 ms⁻¹ for the ER2 (Fig. 3a) and 140 ms⁻¹ for the P3 (Fig. 3b and Fig. 3c). 199





200 We change both the maximum and minimum limits of separation, in order to assess scale 201 dependence and sampling error as much as our airborne data permit. The way the edges are 202 identified ensures that a measurement cannot be counted more than twice for a given range of 203 separation. A measurement can, however, enter multiple ranges of separation. For example, a 204 measurement 4-6 km away from a cloud edge enters the ranges of 0-6 km, 2-6 km, 2-12 km, 4-12 205 km, 4-24 km, etc. In total, 5.0 hours of horizontal flight are selected, including the double-counting 206 for a given range but excluding the multiple-counting over multiple ranges. Exactly half of them 207 are over clouds. Note that these expressions of separation are only notional; we discuss this in Sect. 208 4.

209 As with the meso-scale monthly-mean sampling, we take the arithmetic mean of the CTH 210 of the cloudy side and add 50 m (red lines in Fig. 3). The height is extended to the adjacent clear 211 sky (orange lines) for the calculation of AOD_{ct} (Sect. 2.1). The in situ measurements (Sect. 2.1, 212 Table 1) are each averaged over the cloudy sides and over the clear sides. Cases where aerosol 213 measurements are unavailable for 33% or more of the time period, for example due to calibration 214 or operation problems, are excluded. This makes the number of cloudy-clear pairs vary from 215 property to property for a given range of separation. In total, 3.8 hours of AOD_{ct} measurements 216 enter the analysis.

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2.3. Statistical hypothesis testing

218 We employ the paired t-test, also called paired-samples t-test or dependent t-test, to 219 determine whether the mean difference in each property (e.g., AOD_{ct}) between the presence and 220 absence of low-level clouds is statistically consistent with the null hypothesis of zero difference. 221 The procedure entails calculating the t statistic, the ratio of the mean cloudy-clear differences to 222 their standard error. Here the standard error is the standard deviation computed for N-1 degrees of 223 freedom divided by the square root of N, where N is the number of sample pairs. Note that the 224 standard deviation is close to the root-mean-square deviation (RMSD) for small absolute mean 225 difference, unless N is smaller than five.

For the calculated t statistic, the two-tailed p value is looked up. Small p values are associated with large t statistics and hence generally large mean differences relative to RMSD. If the p value is smaller than 0.05, we reject the null hypothesis. If it is greater, we do not.





229 The procedure makes several assumptions. One is independence of the differences. 230 Synoptic- and meso-scale phenomena prevalent throughout ORACLES (e.g., subsidence and anticyclones) reduce the independence of the samples. The low day-to-day meteorological 231 232 variability and repeated flight paths might mean that the same aerosol-cloud conditions were 233 sampled day after day. It is unclear whether this would reduce the independence of the cloudy-234 clear differences - a potential, seemingly untestable caveat for the meso-scale monthly-mean 235 sampling (Sect. 2.2.1). In the local scale the exclusion of contiguous cloud edges (Sect. 2.2.2) 236 should attain a high level of independence from one another. The procedure also assumes 237 continuous (not discrete), approximately normally distributed data free of outliers.

238 **3. Results**

The meso-scale monthly-mean method finds little systematic difference in 532 nm AOD_{ct} (Fig. 4). Most markers lie near the 1:1 line. The mean difference, an indicator of systematic differences, is +0.02. This is only +16% of the RMSD, an indicator of the total (random and systematic) variability. The p value from the paired t-test is 0.23. Thus, the AOD above low-level clouds is not significantly different from that at the same heights above nearby clear skies in this scale. The p value is also greater than 0.05 for log₁₀ of AOD_{ct}, the Angstrom exponent and in situ aerosol properties (Table 2, see the rows labeled "box means").

The only exception is the particle number concentration. Four of the 32 horizontal boxes see 3-7 times as large concentration over clouds as that over neighboring clear skies (4600-5700 cm⁻³ vs. 700-2100 cm⁻³). The mean cloudy-clear difference among all box means is about +40% of the RMSD. The t-test yields a p value of 0.01. One of the assumptions underlying the test, the absence of outliers, may be broken in this case.

The local-scale near-synchronous method finds virtually the same results. The AOD_{ct} is compared in Fig. 5a for 2-6 km separation. The time period corresponds to approximately 10-30 s temporal range on the ER2 (13 data points from the 2016 campaign) and 14-43 s at the average P3 speed (53 from 2017 and 2018). All data points lie near the 1:1 line. The mean difference, -0.002, is only -21% of the RMSD for 2-6 km separation. The p value is 0.08.

We run the same calculation for other combinations of minimum and maximum separation. Fig. 6 shows the resulting statistics. The mean difference for 2-6 km separation, for example, is represented in Fig. 6a at maximum separation (x axis) of 6 km by the solid orange line that starts





after the minimum separation of 2 km. This line also shows that the mean difference is -0.01 if the maximum separation is set to 20 km while keeping the minimum at 2 km. The longest blue line represents the calculations for zero minimum separation (i.e., with the twilight zone included). All other solid lines represent the results with greater minimum separation. For example, the green line that is missing data up to 4 km indicates that the mean difference is closer to -0.01 at 12 km, as shown in Fig. 5b.

For the separation up to 20 km, the mean difference is mostly between 0 and -0.01. The p value, shown in Fig. 6b, is below 0.05 for only a handful of the ranges of separation, many with minimum separation of 0-2 km. This is also true for \log_{10} of AOD_{ct}, the Angstrom exponent and in situ aerosol properties including the number concentration (Table 2). Large p values are also found for the ER2- and P3-borne measurements separately and for the 4STAR and the HSRL-2 AOD separately for 2017 and 2018.

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4. Discussion and Conclusions

272 Virtually no systematic differences in aerosol properties are found between the air above 273 low-level clouds and that above clear areas nearby in ORACLES daytime airborne measurements. 274 The finding holds for a range (0-20 km) of distances between, and expanses of, the two air masses. 275 Note that the temporal and horizontal dimensions associated with the local-scale near-synchronous 276 sampling must be collectively overestimated, because the aircraft may have been running parallel 277 to cloud edge. There is no easy way to know how far from the nearest cloud edge the airplane was 278 in reality. Images from cameras on the plane and satellites may give some context. But we stop 279 short of examining them, discouraged by the perceived difficulty in unifying definition of cloud 280 edges between the cameras and the lidar, among other image processing issues. Although we do 281 not know what the real distances and expanses are, that probably does not matter for the region 282 and season of our study, judging by the consistently large p values across the notional distances 283 and expanses. The meso-scale monthly-average sampling, resting on larger data, provides 284 consistent results. Note that this conclusion may or may not apply to environment elsewhere with 285 less uniform clouds.

Our analysis does not support aerosol-cloud interactions, circulation patterns or anything else as a cause for a significant systematic difference, simply because such a difference is not evident. The lack of obvious sensitivity to the smoke-cloud gap height, indicated by marker color





in Fig. 5, is consistent with this conclusion. The smoke bottom height minus the mean CTH gives an estimate of whether aerosols may be physically in contact with clouds and therefore there is a chance of wet removal. Our analysis does not detect any sign of local aerosol removal by the underlying clouds.

293 An important difference between the present analysis and the CALIOP-based one (Chung 294 et al., 2016), apart from the spatiotemporal range and resolution, is that the HSRL algorithm (Hair 295 et al., 2008) does not use any explicit layer detection. The return signal in the molecular signal 296 provides a measure of the aerosol attenuation and extinction. A very tenuous aerosol layer still 297 produces a reported extinction with a reported error bar. If the aerosol extinction is very small, the 298 error bar may exceed the retrieved value, but there is no cutoff at small values that produces the 299 kind of bias one gets from a detection threshold. Furthermore, the signal-to-noise is higher than 300 that of CALIOP, as explained in Sect. 2.1.

We posit that the systematic differences shown in Chung et al. (2016) are solely a CALIOP retrieval artifact, at least for the ORACLES region and season. As the authors discuss, the CALIOP standard algorithm has a detection bias. The algorithm confines itself to distinct aerosol layers whose signals are high enough compared to detector noise and, during the day, solar background light. If the signal-to-noise ratio of a layer is not high enough, no extinction is reported for the portion of the aerosol profile; summing up the extinction produces a low-biased AOD.

The depolarization/multiple scattering method by Hu et al. (2007) retrieves above-cloud AOD from CALIOP without a layer detection algorithm. This method may lead to a different result from Chung et al. (2016). A future study based on the Hu et al. method and extended to the globe as in Kacenelenbogen et al. (2019) will also address environment under a wider variety of synopticand meso-scale conditions that produce specific opaque water clouds.

312 The absence of systematic differences is good news, because satellite retrievals and studies 313 of radiative effects do not need to treat these two conditions as different. Our results on AOD_{ct} 314 justify, for example, temporal and horizontal extrapolation of above-cloud AOD to adjacent clear 315 skies and attribution of the difference from full-column AOD to the planetary boundary layer. Our 316 results on the aerosol intensive properties suggest that a single set of aerosol models can be used 317 for the aerosols in the free troposphere regardless of whether clouds exist below, which may allow 318 better characterization of the underlying clouds and the radiative effects (Matus et al., 2015; Meyer 319 et al., 2015). It seems reasonable to use aerosol properties retrieved in clear skies for estimating





320 the direct radiative effects of aerosols above nearby clouds. But challenges remain. Random variability in AOD and other aerosol properties is significant, as indicated by RMSD in the present 321 322 study and quantified for smoke elsewhere (Shinozuka and Redemann, 2011). It may be 323 problematic to assume the same values for intensive properties for reasons not investigated here, 324 for example: form of combustion, degree of aerosol ageing and influence of the boundary layer. These may be tackled more effectively by combining sensors of various capabilities with improved 325 326 spatiotemporal resolution and retrieval algorithms (National Academies of Sciences, Engineering, 327 and Medicine et al., 2019). Improved spatiotemporal satellite observations of aerosol properties in 328 clear skies and above clouds are urgently needed to reduce the uncertainty in total aerosol radiative 329 forcing (National Academies of Sciences, Engineering, and Medicine et al., 2019). For this, we 330 are looking forward to the next generation of space-borne lidars, radars, microwave radiometers, 331 polarimeters and spectrometers such as the ones that will address joint Aerosols, Clouds, 332 Convection and Precipitation (ACCP) science goals and objectives (https://science.nasa.gov/earth-333 science/decadal-accp)

Data availability

The P3 and ER2 observational data (ORACLES Science Team, 2017, 2019) are available
 through www.espo.nasa.gov/oracles.

337 Author contribution

All authors participated in the investigation during the ORACLES intensive observation periods. In addition, MSK led conceptualization, funding acquisition, methodology, project administration and supervision. YS led data curation, formal analysis, software and validation and wrote the original draft. YS and MSK contributed visualization. All but CJF reviewed and edited the manuscript.

343 **Competing interests**

344 The authors declare that they have no conflict of interest.





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Table 1. Properties and instruments used in this study and the altitudes they refer to.

| Property | September 2016 on the ER2 aircraft | | August 2017 and October 2018* on the P3 aircraft | |
|--|---------------------------------------|--|--|---|
| | Instrument | Altitude | Instrument | Altitude |
| cloud top height (CTH) | HSRL-2 | limited to \leq 3241 m in this study | HSRL-2 | no higher than 500 m below the P3 and \leq 3241 m |
| aerosol optical depth above cloud top height (AOD _{ct}) | HSRL-2 | from ~50 m above the CTH to 14 km | 4STAR HSRL-2 and 4STAR | from the P3 to top of atmosphere (TOA), when the P3 is 500-1500 m above CTH from ~50 m above the CTH to TOA, except 0-1500 m below the P3, when the P3 is >1500 m above CTH |
| extinction coefficient, single scattering albedo, submicron non- refractory organic mass, number concentration | - | - | nephelomete r, PSAP, HR-ToF AMS and condensatio n particle counter | at the P3 when the P3 is 500-1500 m above CTH |

471 * One day in September 2017 and two days in September 2018 are also included.

472 - Not presented in this study. Observations were made from the P3, away from the ER2 for most

473 cases.

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| Sampling* | Mean Difference | RMSD | р | Number of Pairs |
|-----------|-------------------|--|---------------------------------|-----------------|
| | | 532 nm AOD _{ct} | | |
| 2-6 km | -0.00 | 0.01 | 0.08 | 60 |
| 4-12 km | -0.01 | 0.02 | 0.23 | 18 |
| box means | +0.02 | 0.12 | 0.23 | 54 |
| |] | log ₁₀ 532 nm AOD _{ct} | t | |
| 2-6 km | -0.00 | 0.01 | 0.15 | 60 |
| 4-12 km | -0.00 | 0.02 | 0.27 | 18 |
| box means | +0.05 | 0.19 | 0.07 | 54 |
| | Angs | trom Exponent of A | OD _{ct} | |
| 2-6 km | -0.04 | 0.11 | 0.00 | 5 |
| 4-12 km | -0.02 | 0.05 | 0.08 | 1 |
| box means | -0.02 | 0.10 | 0.14 | 5. |
| | In Situ 550 n | m Extinction Coeffi | cient (Mm ⁻¹) | |
| 2-6 km | -0.2 | 3.0 | 0.87 | |
| 4-12 km | -3.6 | 5.1 | 0.31 | |
| box means | +19.0 | 67.9 | 0.18 | 2- |
| | In Situ 550 |) nm Single Scatteri | ng Albedo | |
| 2-6 km | -0.00 | 0.01 | 0.14 | |
| 4-12 km | -0.01 | 0.01 | 0.35 | |
| box means | -0.00 | 0.05 | 0.99 | 2 |
| | Submicron Non-ref | ractory Aerosol Org | ganic Mass (ugm ⁻³) | |

Table 2. Statistics on the cloudy-clear differences





| 2-6 km | +0.1 | 0.5 | 0.75 | 9 | | |
|--|------|------|------|----|--|--|
| 4-12 km | -0.4 | 0.6 | 0.38 | 3 | | |
| box means | +1.2 | 4.4 | 0.14 | 28 | | |
| Number Concentration of Particles >10 nm (cm ⁻³) | | | | | | |
| 2-6 km | 5 | 59 | 0.82 | 10 | | |
| 4-12 km | -110 | 121 | 0.09 | 3 | | |
| box means | 614 | 1411 | 0.01 | 31 | | |

478 * Either the temporal separation from cloud edges or box means.





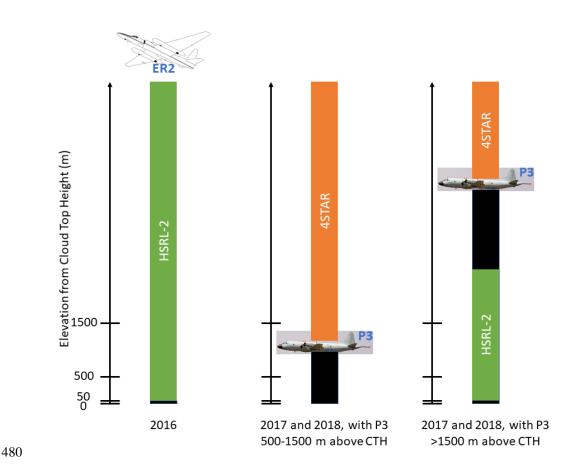


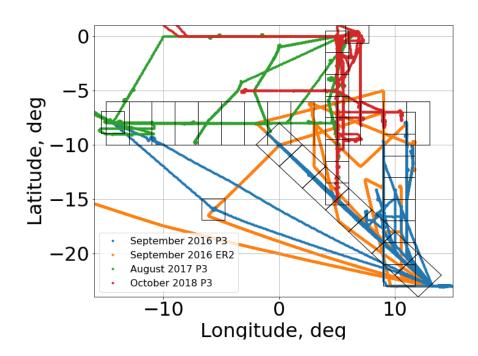


Figure 1. AOD above cloud top height (AODct). See text and Table 1 for details.





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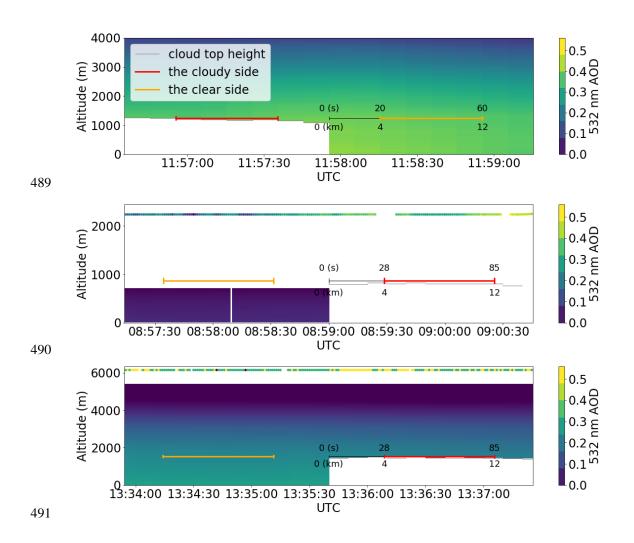
484Figure 2. The flight paths of ORACLES. The boxes for meso-scale monthly-mean
sampling are superimposed.

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492 Figure 3. (a) Local-scale near-synchronous sampling based on the HSRL-2 cloud top height 493 (CTH) product. In this subset of the ER2 flight on September 12, 2016, a cloud edge is found 494 at 11:57:56. The cloudy and clear side, each with horizontal separation of 4-12 km measured 495 from cloud edge, are marked by red and orange lines, respectively. The HSRL-2 AOD 496 profiles are given for altitudes from ~50 m above the CTH. (b) An example of local-scale 497 near-synchronous sampling from the P3 aircraft with both HSRL-2 and 4STAR onboard. 498 With the P3 500-1500 m above the CTH, as is the case with this example from October 5, 499 2018, we use 4STAR AOD only. The 4STAR AOD is indicated at the P3 altitudes just above 500 2000 m but refers to all altitudes above them. (c) Another example of local-scale near-501 synchronous sampling from the P3 aircraft with both HSRL-2 and 4STAR onboard. For the 502 time periods when it flew >1500 m above the CTH, as is the case for this example from



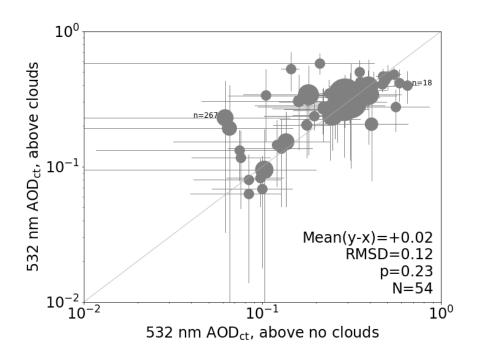


503 October 15, 2018, the 4STAR AOD, indicated at the P3 altitudes just above 6000 m, is added 504 to the HSRL-2 AOD at ~50 m above the CTH.

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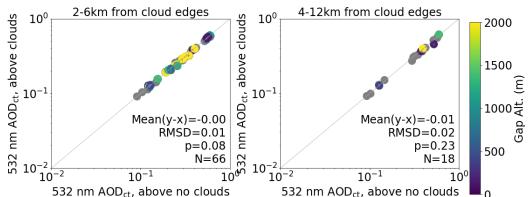


508Figure 4. The meso-scale monthly-mean samples of the AOD above cloud top height.509Each marker represents the mean over a box shown in Fig. 2. The bar represents the510±1 standard deviation range. The marker size is proportional to the number (n) of 10511s measurements, the fewer of the cloudy and clear groups.

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516 532 nm AOD_{ct}, above no clouds 532 nm AOD_{ct}, above no clouds 532 nm AOD_{ct}, above no clouds ■₀
517 Figure 5. (a) The local-scale near-synchronous samples of the AOD above cloud top height.
518 Each marker represents the mean over the cloudy and clear sides of a cloud edge, each 2-6
519 km from the edge. The bars indicate the standard deviation of the measurements in each
520 side, almost all of them too short to be discernible. (b) Same as (a) except for the horizontal
521 separation of 4-12 km.

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- 523
- 524





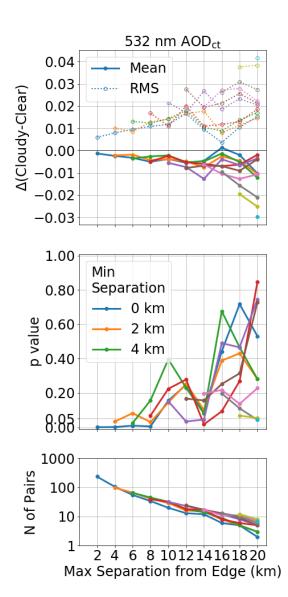


Figure 6. (a) The mean and root-mean-square deviations of the AOD above cloud top between the cloudy and clear sides of cloud edges. Each side is defined by the horizontal separation from cloud edge. The maximum separation (e.g., 12 km in Fig. 3) is indicated on the x axis. Each line represents the minimum temporal separation (e.g., 4 km in Fig. 3) of 0, 2, 4, ..., 18 km in descending order of line length. (b) The p values determined through the paired t-test. (c) the number of cloudy/clear pairs.