

Interactive comment on “Top-down and Bottom-up aerosol-cloud-closure: towards understanding sources of uncertainty in deriving cloud radiative flux” by Kevin J. Sanchez et al.
Anonymous Referee #1

5 Received and published: 12 May 2017

Reviewer overview:

10 The authors provide an analysis of cloud droplet closure using data collected at Mace Head, Ireland during summer 2015. The dataset includes surface based aerosol and remote sensing data from the Mace Head station. In addition, in situ vertical profile data was collected from a new UAV platform, which was deployed with a rotating payload comprising of meteorological probes, an aerosol optical sizing spectrometer and a cloud extinction monitor. Finally, the authors also make use of satellite cloud remote sensing products.

15 The authors conduct an aerosol-cloud microphysical closure analysis from the surface based data input into a parcel model (bottom-up) and from the satellite and in situ cloud extinction (top-down) to assess the uncertainty in deriving shortwave cloud radiative effects associated with microphysics. The authors find that when they account for reductions in cloud drop number concentration associated with entrainment, the difference between modelled and observed shortwave fluxes are reduced. The authors also find that decoupled clouds result in larger differences between modelled and observed shortwave fluxes, compared to well mixed cases. Overall the paper is interesting and suitable for publication in ACP. I have a number of minor points listed below, which I urge the authors to consider before the paper is finalized.

25 **We thank the reviewer for their comments that significantly contributed to improving the original manuscript. Please see below responses to each of the authors comments and suggestions.**

30 **The authors want to note that values in Table 3 have slightly changed. These changes were brought about by reviewer #2’s comment to present cloud optical thickness. It was noticed that the ‘observed’ optical thickness was not consistent between calculations that including and excluding cloud top entrainment. The observed optical thickness is calculated from the observed cloud droplet extinction. The observed droplet extinction is calculated by subtracting the simulated cloud droplet extinction and fitted difference in droplet extinction ($\delta\sigma_{\text{ext}}$) (Figure 8b,d,f). This was necessary to take into account the fact that the UAV’s often missed portions of the cloud. The linear fit made it possible to fill the gaps. Since the observations should be consistent, the observations from the fit that excluded entrainment was compared to simulations with entrainment.**

General point: It might be useful to clarify in the abstract (and in sections before you define RF) that you are discussing shortwave radiative flux

40 **We have changed “radiative flux” to “shortwave radiative flux” in both the abstract and throughout the paper.**

45 L80 “surface latent heat flux, (i.e. evaporative cooling at the surface)” – this is misleading: surface latent heat flux does not induce cooling. It is independent of the heat budget at the surface. The mechanism, described in BW97, results in decoupling because under high LHF, there is a larger jump in buoyancy flux at cloud base, the cloud layer drives the turbulent motions and a zone of negative buoyancy flux develops in the sub-cloud layer. When this zone becomes

too large it becomes dynamically favorable for the cloud layer to decouple from the sub-cloud layer.

The text has been altered and is present with the response to the reviewer's comment on line 87.

L81 BW97 claim that drizzle is not necessary for their “deepening-warming decoupling” mechanism, however they do show that it can have a substantial impact on the promotion of negative sub-cloud buoyancy fluxes and induce decoupling.

The text has been altered and is present with the response to the reviewer's comment on line 87.

L87 also related, moving air over a higher SST does not induce cooling. Suggest reviewing Stevens, 2002, Bretherton and Wyant, 1997 and Schubert et al., 1979 (not exhaustive list) for information about the mechanism of decoupling driven by increased surface latent heat fluxes and negative sub-cloud buoyancy fluxes.

Based on the previous 3 comments and a review of the literature, this section has been re-written to more accurately describe the processes taking place that cause boundary layer decoupling:

“Marine boundary layer decoupling is often seen in the tropics and has been attributed to processes that involve cloud heating from cloud-top entrainment, leading to decoupling of the boundary layer (Bretherton et al., 1997; Bates et al., 1998; Albrecht et al., 1995; Zhou et al., 2015; Stevens, 2002). In addition, Bretherton and Wyant (1997) have shown that the decoupling structure is mainly driven by a high latent heat flux that results in a large buoyancy jump across the cloud base. This high latent heat flux is attributed to easterlies bringing air over increasing SST, where the boundary layer becomes deeper and more likely to decouple (Albrecht et al., 1995). The cloud layer drives the turbulent motion and a zone of negative buoyancy flux develops below cloud. The turbulent motion is driven by radiative cooling at cloud top, causing air to sink (Lilly, 1968). The zone of negative buoyancy occurs because the deepening of the boundary layer causes the lifting condensation level of the updraft and downdraft to separate. This is important because latent heating in the cloud contributes significantly to the buoyancy in the cloud (Schubert et al., 1979). If this zone of negative buoyancy flux becomes deep enough, it is dynamically favorable for the cloud layer to become decoupled from the cloud layer (Bretherton et al., 1997). Bretherton and Wyant (1997) also show that drizzle can have a substantial impact on enhancing the negative buoyancy flux below cloud, but drizzle is not necessary for decoupling mechanism they proposed. Other factors, such as the vertical distribution of radiative cooling in the cloud, and sensible heat fluxes, play less important roles. Turton and Nicholls (1987) used a two-layer model to show that decoupling can also result from solar heating of the cloud layer; however, only during the day. Furthermore, Nicholls and Leighton (1986) showed observations of decoupled clouds with cloud-top radiative cooling and the resulting in-cloud eddies do not mix down to the surface (further suggesting radiative cooling plays a less important role). Russell et al. (1998) and Sollazzo et al. (2000) showed that, in a decoupled atmosphere the two distinct layers have similar characteristics (e.g., aerosol and trace gases composition), with different aerosol concentrations that gradually mix with each other, mixing air from the surface-mixed layer into the decoupled layer and vice versa. These previous studies also show that aerosol concentrations in the decoupled layer are lower than those in the surface-mixed

layer implying an overestimation in cloud **shortwave** radiative flux when using ground-based aerosol measurements.

L123 (sp) Nafion.

The spelling has been corrected.

L145-147 how is the scaling done? In Figure 7 and Figure 11, RH values are shown to be < 100% in the cloud layer.

Referenced text: “As RH sensors are not accurate at high RH ($> 90\%$), the measured values have been scaled such that RH measurements are 100% in a cloud. At altitudes where the UAV is known to be in-cloud (based on *in-situ* cloud extinction measurements) the air mass is considered saturated (RH $\sim 100\%$).”

The calibration for RH values of 70-100% were adjusted (the slope of the calibration linear fit was modified) so that the maximum RH was 100%. The maximum RH before this correction which was typically between 90 and 95%. For the calculation of in-cloud water vapor content (for figure 10) RH values in cloud were recalibrated using cloud as 100% RH. The simulation calculated RH values $>100\%$ in-cloud and therefore was not affected.

L155 typo – Aerosols

The typo is fixed.

L204-206 Mixing state: can you clarify what you mean by “externally mixed types of particles”.

You then state that aerosols are internally mixed: is it fair to say that aerosols are internally mixed when this paper is discussing evidence of a significant fraction of air entrained into the boundary layer from above? Would aerosols from the free troposphere not have different chemical characteristics from the boundary layer? The phrase “lack of aerosol sources” is also ambiguous.

The ACPM has the capability of including both internally and externally mixed particles.

As indicated by the reviewer, the aerosols were internally mixed. We have removed

“externally mixed types of particles” to avoid confusion.

Only parts of the cloud layer are suggested to have free tropospheric air entrained. Though the fraction of free tropospheric air in parts of the cloud are high, homogeneous entrainment would not result in the activation of new particles and therefore would not alter the cloud shortwave radiative forcing. Also, typically aerosols in the free troposphere are too small to be CCN active.

We have changed “owing to lack of aerosol sources” to “because there were no immediate strong sources of pollution”.

L215-226 Does the model include the effects of coalescence scavenging, which may be quite significant for a marine cloud over the 2-hour period given here.

The model does not include the effects of coalescence scavenging. However, after looking further into our results, the simulation time is less than 20 minutes at the average updraft velocity, with the exception of the C21Cu case. Based on results from Feingold et al. [2013], coalescence scavenging rates are negligible for the CDNC and LWC ($<0.4 \text{ g/m}^3$) for the case studies except for the C21Cu case. The C21Cu case does have significantly high Liquid water content ($>1.0 \text{ g/m}^3$), and therefore is susceptible to coalescence of droplets.

The following text has been added to clarify this point:

“Feingold et al. (2013) showed that autoconversion and accretion rates are negligible for the modeled values of LWC and CDNC except for the C21Cu case, which had LWC $> 1 \text{ g m}^{-3}$. Thus, droplet number loss by collision coalescence can be neglected for all cases except for the C21Cu case.”

A footnote has been added to the table to indicate the C21Cu is susceptible to coalescence of droplets.

L222 should there be a negative sign in your equation for the adiabatic cooling term (i.e. – gdz/cp)?

Yes, we have added the negative sign to correct the equation.

L340-342 I think you could be a bit clearer about how you come to this conclusion from the data shown in Table 2.

Previous text: “For example, in the C11Sc case, in-situ observations do indeed show cloud-top inhomogeneous entrainment; consequently, the usual 30% reduction in CDNC does not need to be applied (Table 2).”

The text has been changed to the following to clearly indicate the reason for not applying the correction.

“For the C11Sc case, before the correction, proposed by Freud et al. (2011), is applied the satellite derived CDNC (83 cm^{-3}) is within 30% of the ACPM CDNC (88 cm^{-3}) similar to the other cases (Figure 6), but if the correction is applied, the satellite derived CDNC (58 cm^{-3}) is not within 30% of the ACPM CDNC. This indicates cloud top entrainment for the C11Sc case is already inhomogeneous and the usual 30% reduction in CDNC to correct for the inhomogeneous assumption does not need to be applied. “

L374-390 in both well-mixed and decoupled boundary layers, there are diabatic processes affecting the cloud layer namely, long-wave cooling of the cloud top, short wave absorption, drying due to drop sedimentation. To what extent do these processes interfere with the assumption of a cloud parcel being a mixture of cloud base air and entrained air?

While these processes were not taken into account, they are expected to be small. The vertical extent of these clouds is small, consequently droplet diameters are relatively small ($R_{eff} < 15$ microns) which limits the impact of droplet sedimentation. Typically, shortwave absorption is small and only slightly offsets long-wave cooling (Harrington et al. 1999). If long-wave cooling were the dominate process, the in-cloud lapse rate would be super-adiabatic. However, the in-cloud measured lapse rate was sub-adiabatic, so we conclude that entrainment warming was dominant mechanism in changing the in-cloud temperature. Also, long-wave cooling is greatest near the cloud top, meaning it is only important if a parcel remains near the cloud-top for a significant amount of time (Harrington et al. 1999; Hartman et al. 2004). For the entrainment cases considered in this study, the air masses have short residence times in the clouds (less than 20 minutes) and only spend a small fraction of this time at cloud-top.

Fig 10: suggest putting the flight details in the caption (like Fig 11) for clarity

We have added the case names to the description of figure 10 (like figure 11).

L388-400 I think this section could be reworded to improve its clarity. I also have a few concerns: 1) It's not clear what you are referring to with the linear proportional relationship (L392). As you clarified, the $q_v = q_t$ is only true outside the cloud, but if this mixing diagram is only used to illustrate processes in the cloud, what new information do you get for cloudy air with the addition of the second dimension (q_v) over the 1D θ -E mixing calculation done with Eq.4? 2) The dashed line is linear by design, on a q_t axis. Since $q_t = q_v$ at the two end points these would indeed be the end points of the dashed line but on this q_v axis the line would be curved 3) It is not clear what the adiabatic line is supposed to represent. Why does θ -E change during an adiabatic process?

Original referenced text:

“Figure 10a and b present the relationships between two conservative variables measured by the UAV (water vapor content, q_v , and θ_e) for C11Sc and D05Sc. The q_v is derived from relative humidity measurements and is equivalent to the q_t for sub-saturated, cloud-free air (i.e., < 100% RH).

Figure 11 shows the relative humidity and θ_e profiles used in Figure 10. For both C11Sc and D05Sc, $\theta_{e,c}(z)$ is directly measured in-cloud, and q_v and θ_e exhibit an approximately linear, proportional relationship (Figure 10; Eq. 4). The linear relationship is assumed to be a result of the cloud reaching a steady-state, with air coming from cloud-base and cloud-top (e.g. cloud lifetime >> mixing time). The observed in-cloud q_v in Figure 10a and b is less than the conservative variable q_t , however, the figure also includes q_t based on simulated adiabatic and cloud-top entrainment conditions. Eq. (4) is used to derive the simulated cloud-top entrainment conditions (Figure 10a and b), where the fraction entrained is used to calculate q_t and shows a linear relationship between q_t and θ_e . Measurements above cloud-top (RH < 95%) with $q_v > 5.1 \text{ g kg}^{-1}$ and $q_v > 6.5 \text{ g kg}^{-1}$ are used to represent the properties of the entrained air for C11Sc and D05Sc, respectively (Figure 10).”

Modified text:

“Figure 10a and b present the relationships between two conservative variables measured by the UAV (water vapor content, q_v , and θ_e) for C11Sc and D05Sc. The q_v is derived from relative humidity measurements and is equivalent to the q_t for sub-saturated, cloud-free air (i.e., < 100% RH). The cloud-free air is shown in blue in Figure 10, where the below cloud measurements have lower θ_e than in-cloud and the above cloud measurements have higher θ_e than in-cloud.

Figure 11 shows the relative humidity and θ_e profiles used in Figure 10. For both C11Sc and D05Sc, $\theta_{e,c}(z)$ is directly measured in-cloud, and q_t and θ_e exhibit an approximately linear relationship (Figure 10; Eq. 4). The linear relationship of q_t and θ_e (between the non-mixed sources of air indicated by orange circles in Figure 10) is assumed to be a result of the cloud reaching a steady-state, with air coming from cloud-base and cloud-top (e.g. cloud lifetime >> mixing time). The observed in-cloud q_v in Figure 10a and b is less than the conservative variable q_t , however, the figure also includes q_t based on simulated adiabatic (marked with an ‘X’) and cloud-top entrainment (dashed black line) conditions. Under adiabatic conditions q_t and θ_e do not change in the cloud, which is why the adiabatic simulations only consists of one point in Figure 10. Eq. (4) is used to derive the simulated cloud-top entrainment conditions (Figure 10a and b), where the fraction entrained is used to calculate q_t and shows a linear relationship between q_t and θ_e . Measurements above cloud-top (RH < 95%), labeled entrained air, with $q_v > 5.1 \text{ g kg}^{-1}$ and $q_v > 6.5 \text{ g kg}^{-1}$ are used to represent the properties of the entrained air for C11Sc and D05Sc, respectively (Figure 10). These conditions were chosen because these values are on the mixing line, between the non-mixed sources identified by the orange circles.”

Responses to each part of the comment:

1. The text now refers to the linear relationship in Figure 10: “The linear relationship of q_t and θ_e (between the non-mixed sources of air indicated by orange circles in Figure 10) is assumed to be a result of the cloud reaching a steady-state, with air coming from cloud-base and cloud-top (e.g. cloud lifetime >> mixing time).”

The q_v is not necessary for equation 4, but the linear relationship between these 2 conservative variables in the cloud enables the visualization of a mixing line and enables us to show the change in total water content between adiabatic (without entrainment) and entrainment scenarios. Also, the linear relationship helps define which observations best represent entrained air (red points in figure 10).

2. The graph has now been modified so that the left axis represents observed q_v and the right axis represents simulated q_t .
3. θ_e should not change in an adiabatic process. Figure 10 has been modified so that the simulated θ_e in an adiabatic process does not change. The following text was added to the discussion of Figure 10: “Under adiabatic conditions q_t and θ_e do not change in the cloud, which is why the adiabatic simulations only consists of one point in Figure 10.”

L417 what is the sensitivity of cloud extinction if mixing is homogeneous v.s. inhomogeneous compared to, say, the magnitude of the entrainment? Are there any other clues from your data set that could help confirm that the inhomogeneous process is a suitable assumption?

We cannot calculate the degree to which entrainment was homogeneous with traditional methods because they involve cloud droplet size distributions observations, which were not possible with the class of UAVs used here. Nonetheless, previous observations (Burnet and Brenguier, 2007; Beals et al. 2015) have used cloud droplet size distribution observations to show cloud top entrainment is mostly inhomogeneous entrainment. The evaporation rate for homogeneous mixing strongly depends on mixing scales, so there is not a unique answer for homogeneous mixing (Lehmann et al. 2009).

Based on our results, the inhomogeneous correction used for the satellite measurements greatly increases the error in CDNC (when comparing to the ACPM CDNC) for the coupled entrainment case (C11Sc) suggesting the entrainment is inhomogeneous. Furthermore, inhomogeneous entrainment would result in greater CDNC and therefore, greater error in radiative flux.

L470 What was happening on the other cases? Was the cloud layer more vigorously mixed, such that entrainment warming and drying was homogenized through the layer more rapidly?

Referenced text: “...and decoupling of the boundary layer occurs on 4 of the 13 flight days.”

The remaining 2 cases with a decoupled layer have insufficient in-cloud measurements for analysis and the clouds were too thin for satellite analysis. Figure 6 consist of the OPC concentration profile from one of these 2 cases and has a cloud thickness of less than 50 m. A parenthetical statement was added to the referenced text:

“and decoupling of the boundary layer occurs on four of the 13 flight days (two decoupled cloud cases were not discussed due to the lack of in-cloud measurements).”

L474 “presence (of) marine biogenic. . .”

We have added the word “of”.

L474 local anthropogenic. . .what?

The sentence was removed since the focus of the paper is marine boundary layer observations and not anthropogenic sources.

L475 "observations and simulat(ed)"?

We have changed the word “modeled” to “simulated” as suggested by the reviewer.

Interactive comment on “Top-down and Bottom-up aerosol-cloud-closure: towards understanding sources of uncertainty in deriving cloud radiative flux” by Kevin J. Sanchez et al.
Anonymous Referee #2

Received and published: 23 May 2017

Reviewer overview:

Summary: This manuscript presents an observational analysis to understand sources of uncertainty in deriving cloud radiative flux. The observations are from a number of platforms, including ground based, UAV, and satellite measurements. They used a 1-D microphysical model in conjunction with observations to derive microphysical and optical properties of observed clouds. The differences were found in radiative fluxes between the simulated and the observed. They concluded that the cloud-top entrainment is an important source of uncertainty for the cloud radiative flux calculation; it is particularly true for decoupled cloud boundary layers because ground-based measurements are no longer enough to obtain reliable data in the decoupled cloud layer. Authors’ overall analysis technique is good and their conclusion is important and interesting. My main criticism is that some discussions and figures are not clear and confusing. I recommend publication after following comments are addressed.

We thank the reviewer for their comments that significantly contributed to improving the original manuscript. Please see below responses to each of the authors comments and suggestions.

The authors want to note that values in Table 3 have slightly changed. These changes were brought about by reviewer #2’s comment to present cloud optical thickness. It was noticed that the ‘observed’ optical thickness was not consistent between calculations that including and excluding cloud top entrainment. The observed optical thickness is calculated from the observed cloud droplet extinction. The observed droplet extinction is calculated by subtracting the simulated cloud droplet extinction and fitted difference in droplet extinction ($\delta\sigma_{\text{ext}}$) (Figure 8b,d,f). This was necessary to take into account the fact that the UAV’s often missed portions of the cloud. The linear fit made it possible to fill the gaps. Since the observations should be consistent, the observations from the fit that excluded entrainment was compared to simulations with entrainment.

I am wondering about the significance of showing the cloud-top extinction in Table 2 and 3. Even though the cloud-top radiative flux differences (ΔFR) in the two decoupled cases are larger than those in the coupled cases, $\delta\sigma_{\text{ext}}$ values are similar for all the cases as shown in Table 3. The cloud-top value $\delta\sigma_{\text{ext}}$ doesn’t seem to mean a lot in terms of cloud optical property. Because the cloud-top radiative flux (RF) depends on the optical depth as shown in (2), it is probably more appropriate to show cloud optical depth (τ).

Cloud optical depth has been added to Tables 2 and 3.

Page 2, line 71: “Such decoupled layers often contain two distinct cloud layers, . . . a lower layer within the well-mixed surface layer and a higher decoupled residual layer between the free atmosphere and surface layer”. I don’t think the surface layer can be well mixed because turbulent eddies there are too small near the surface to produce strong mixing. You probably meant surface based mixed layer. That is, a mixed layer that is connected to, but deeper than the surface layer. Why do you call a decoupled layer “residual layer”? Is there turbulence source in the decoupled layer? Does it have clouds?

We have modified the text to say “surface mixed layer”. We have also changed “residual layer” to “decoupled layer”. The decoupled layer can have clouds and therefore a source of turbulence which is described by the following text that has been added:

“The cloud layer drives the turbulent motion and a zone of negative buoyancy flux develops below cloud. The turbulent motion is driven by radiative cooling at cloud top, causing air to sink [Lilly et al., 1968].”

Page 3, line 75: “the surface mixed layer”. Surface based mixed layer?

We have chosen to use “surface mixed layer” to define the lower layer in a decoupled boundary layer. This is consistent with a previous Mace Head paper on decoupling boundary layers (Milroy et al. 2011)

Page 3, line 77 and line 80: “. . . involve cloud heating and surface cooling” and “ i.e., evaporative cooling at the surface” I am not sure what is meant by the “surface cooling” or “evaporative cooling”. Note that the surface evaporative cooling by surface moisture flux only cools the ocean surface, not the sub-cloud layer. I do not think the “surface evaporative cooling” directly contributes to the decoupling. Could you give a bit more explanation on this? An increase in the moisture flux with increasing SST enhances the cloud layer buoyancy flux, which intensifies the cloud-top entrainment to mix warmer and drier air into clouds, leading to negative buoyancy flux below cloud base.

The text in this section has been largely modified to more accurately explain the processes. The text has been restated in the response to reviewer 1 and is also shown below:

“Marine boundary layer decoupling is often seen in the tropics and has been attributed to processes that involve cloud heating from cloud-top entrainment, leading to decoupling of the boundary layer (Bretherton et al., 1997; Bates et al., 1998; Albrecht et al., 1995; Zhou et al., 2015; Stevens, 2002). In addition, Bretherton and Wyant (1997) have shown that the decoupling structure is mainly driven by a high latent heat flux that results in a large buoyancy jump across the cloud base. This high latent heat flux is attributed to easterlies bringing air over increasing SST, where the boundary layer becomes deeper and more likely to decouple (Albrecht et al., 1995). The cloud layer drives the turbulent motion and a zone of negative buoyancy flux develops below cloud. The turbulent motion is driven by radiative cooling at cloud top, causing air to sink (Lilly, 1968). The zone of negative buoyancy occurs because the deepening of the boundary layer causes the lifting condensation level of the updraft and downdraft to separate. This is important because latent heating in the cloud contributes significantly to the buoyancy in the cloud (Schubert et al., 1979). If this zone of negative buoyancy flux becomes deep enough, it is dynamically favorable for the cloud layer to become decoupled from the cloud layer (Bretherton et al., 1997). Bretherton and Wyant (1997) also show that drizzle can have a substantial impact on enhancing the negative buoyancy flux below cloud, but drizzle is not necessary for decoupling mechanism they proposed. Other factors, such as the vertical distribution of radiative cooling in the cloud, and sensible heat fluxes, play less important roles. Turton and Nicholls (1987) used a two-layer model to show that decoupling can also result from solar heating of the cloud layer; however, only during the day. Furthermore, Nicholls and Leighton (1986) showed observations of decoupled clouds with cloud-top radiative cooling and the resulting in-cloud eddies do not mix down to the surface (further suggesting radiative cooling plays a less important role). Russell et al. (1998) and Sollazzo et al. (2000) showed that, in a decoupled atmosphere the two distinct layers have similar characteristics

(e.g., aerosol and trace gases composition), with different aerosol concentrations that gradually mix with each other, **mixing** air from the **surface-mixed layer** into the decoupled layer and vice versa. These previous studies also show that aerosol concentrations in the **decoupled layer** are lower than those in the **surface-mixed layer** implying an overestimation in cloud **shortwave** radiative flux when using ground-based aerosol measurements. “

Page 8, line 281-282 about Figure 8. Could you put the flight code (D05Sc, C11Sc and C21Cu) inside the plot boxes? That would be easy to see. The caption of Figure 8 mentions the difference between UAV-observed (green measurements) and ACPMsimulated (black line) to calculate $\Delta \sigma_{\text{ext}}$. But it looks like you also calculate the cloud free values too. Although the (a)-(f) are labeled in each plot, they are not used in the caption.

The flight code has been put inside the plot boxes. We have removed “(green measurements)” since we do calculate $\Delta \sigma_{\text{ext}}$ for cloud free values as the reviewer has pointed out. We have also included the letters in the caption to refer to each plot in the figure.

Page 10, line 354-357: “The UAV observations show both C11Sc have sub-adiabatic lapse rate measurements, compared to simulated moist-adiabatic lapse rates within the cloud (Table 2). . . . The sub-adiabatic lapse rate is attributed to cloud-top entrainment . . . at cloud-top (e.g., Figure 7a)” Where is the comparison between the observed and simulated lapse rate? I only see the simulated values in Table 2. Could you draw a line in Figure 7a to show the adiabatic lapse rate? It is hard to see the lapse rate is sub-adiabatic

The sub-adiabtic lapse-rate results are now expressed in the text rather than the table because there were only sub-adiabatic lapse rates for two of the cases. Table 2 is cited to show the measured and simulated lapse rate.

The following text, at the end of section 3.2, compares δRF when using the adiabatic lapse rate and the observed lapse rate (now referred to as the lapse rate adjustment entrainment method):

“Finally, the **lapse rate adjustment entrainment method [Sanchez et al., 2016] does improve ACPM accuracy between in-situ and satellite-retrieved cloud optical properties relative to the adiabatic simulations, but has greater $\delta \sigma_{\text{ext}}$ throughout the cloud than the **inhomogeneous mixing entrainment** method. For the **lapse rate adjustment entrainment** method δRF decreased from 88 Wm^{-2} to 61 Wm^{-2} and 48 Wm^{-2} to 32 Wm^{-2} for D05Sc and D11Sc respectively.”**

We have not added a line to show the adiabatic lapse rate to in figure 7a because the line, with a 1 K km^{-1} greater lapse rate, would not be noticeably different than the measured lapse rate due to the large x-axis range. The reference to Figure 7a has been removed.

Page 11-12, 391-399: “For both C11Sc and D05Sc, exhibit an approximately linear, proportional relationship (Figure10; Eq. 4.)”. This paragraph is a bit confusing. What flights do those curves come from in Fig. 10? Could you state clearly which part you were referring to that is linear? In Fig. 10, the cloudy part (green curve) is not linear because q_v is not conserved variable for condensation/evaporation process.

The following text has been modified to indicate q_t and θ_e have a linear relationship, and that it is shown between the two orange circles:

“For both C11Sc and D05Sc, $\theta_{e,c}(z)$ is directly measured in-cloud, and q_t and θ_e exhibit an approximately linear relationship (Figure 10; Eq. 4). The linear relationship **of q_t and θ_e**

(between the non-mixed sources of air indicated by orange circles in Figure 10) is assumed to be a result of the cloud reaching a steady-state, with air coming from cloud-base and cloud-top (e.g. cloud lifetime \gg mixing time)."

The flight codes are added to figure 10.

What is meant by "entrained air"? Does it consist of both free air and turbulent air or only free atmosphere and non-mixed air? Does it contain any cloud droplets? If not, why is it (red curve) not linear, particularly for the top panel plot?

The entrained air is the air that is mixed into cloud top which is the air directly above the cloud (within 100 m) and do not contain cloud droplets. The air directly above the cloud may or may not be the free troposphere. For example, in the bottom panel of figure 10, the points in between the 2 circles represent the mixed air layer that you have referred to.

Though this air is not necessarily from the free troposphere, it is what will mix with the cloud top. A point in the orange circle (Figure 10) could have been used to represent pure free tropospheric air that would entrain into the cloud, however using the red points in the mixed air yields the same result because it is on the mixing line and they are more physical representation to use since these are directly above the cloud. The entrained fraction (X in equation 5) will change, but approximately the same amount of liquid water will evaporate no matter which point is used on this mixing line for the entrained air properties. We have changed "entrained air sources" to "entrained air properties used in simulations" in the figure caption.

The red curve appears not to be linear (in the top panel of figure 10) mainly because the mixed air (between the two orange circles in Figure 10) has a smaller layer with no cloud so essentially the line is shorter. It is also possible that the UAV partially re-entered the very top of the cloud momentarily, causing an increase in RH even though σ_{ext} does not increase because the change is below the detection limit. Also, as mentioned in the manuscript the RH sensor is not particularly accurate when RH is greater than 90%, and the water vapor content (y axis of figure 10) is calculated from the RH. The variability in the entrained water vapor is included in the errors in Table 3.

What is the flight code (or number) for these two plots in Fig. 10? Please identify the blue dashed line in the text when discussing the entrainment conditions. There is no (a) and (b) in Fig 10. "Measurements above cloud-top ($\text{RH} < 95\%$) with $q_v > 5.1 \text{ g kg}^{-1}$ and $q_v > 6.5 \text{ g kg}^{-1}$ are used to represent the properties of the entrained air". How do you choose this criterion for the entrained air? You should specify clearly the properties of the non-mixed sources of air: what are the values of θ_e and q_v of the air source? The orange circles include too many possibilities of these values.

The flight code has been added to the figure.

We have now indicated the simulated adiabatic and entrainment conditions in the text:

"The observed in-cloud q_v in Figure 10a and b is less than the conservative variable q_t , however, the figure also includes q_t based on simulated adiabatic (marked with an 'X') and cloud-top entrainment (dashed black line) conditions."

Blue cloud-free air (blue points) are now mentioned with the addition of the following sentence:

"The cloud-free air is shown in blue in Figure 10, where the below cloud measurements have lower θ_e than in-cloud and the above cloud measurements have higher θ_e than in-cloud."

(a) and (b) have been added to figure 10.

The quoted text has been supplemented to include the criteria for choosing entrained air: "Measurements above cloud-top ($\text{RH} < 95\%$), labeled entrained air, with $q_v > 5.1 \text{ g kg}^{-1}$ and

470 $q_v > 6.5 \text{ g kg}^{-1}$ are used to represent the properties of the entrained air for C11Sc and D05Sc, respectively (Figure 10). **These conditions were chosen because these values are on the mixing line, between the non-mixed sources identified by the orange circles.**”

The properties of the entrained air (θ_e and q_v) are given by the red “entrained air” points in Figure 10. The orange circles are not meant to define values, but simple point out approximate end points to the mixing line. As stated in the response to the previous comment, using the properties of the “entrained air”, shown in red, is equivalent to using the an observation from the top of this mixed layer.

475
Line 391: “Figure 11 shows the relative humidity and θ_e profiles used in Figure 10. . . .”
The discussion following this sentence seems to be related to Figure 10. There is no discussion on Figure 11. Fig. 11 caption says “. . . used in Figure 9”. It should be Figure 10?

480 **The main point of figure 11 was to show the measurements used to make figure 10 as a vertical profile.**

The figure 11 caption reference to figure 9 has been changed to figure 10.

Page 12, line 401-405. “Figure 12 shows . . . approaches zero”. There is not much discussion on Fig. 12. What does Figure 12 suggest? What is the definition of Delta $\theta_{e,ent}$? Which curve best represents observation? Does the figure mean that σ_{ext} is sensitive or not sensitive to the entrained air properties?

485
The figure caption has been changed to the following to define delta $\theta_{e,ent}$ and delta q_t :

490 **“Figure 12. Sensitivity of simulated cloud extinction based on variability of entrained air potential temperature (θ_{ent} , K) and entrained air total water mixing ratio ($q_{t,ent}$, g kg⁻¹) for the C11Sc case. **The $\Delta\theta_{ent}$ and $\Delta q_{t,ent}$ terms define the change in the entrained θ and q_t values where no change ($\Delta\theta_{ent} = 0$ and $\Delta q_{t,ent} = 0$) is equivalent to the adiabatic simulation with entrainment from Figure 8c.”****

495
The intent with Figure 12 was not to fit the data, but instead show how the sensitive the simulated droplet extinction is to changes in properties of the entrained air. The σ_{ext} is not very sensitive to the entrainment properties that were measured, but under different circumstances (lower θ and q_t) σ_{ext} can be very sensitive.

500 The last sentence has been added to the quoted text to clarify the connection with Figure 12 and equation 5:

505 **“Figure 12 shows the sensitivity of the simulated cloud extinction profile, for the 11 August case, based on measurement uncertainties related to the entrained q_t and θ . The key variable for identifying the entrained fraction (Eq. 5), $\theta_{e,ent}$, is a function of q_t and θ , so a decrease in either parameter results in a proportional decrease in $\theta_{e,ent}$. Eq. (5) shows that entrainment fraction becomes more sensitive to the uncertainty related to the measurement of θ_e as the difference between $\theta_{e,ent}$ and $\theta_{e,CB}$ approaches zero. **This is also shown in Figure 12 where σ_{ext} is more sensitive to lower entrained q_t and θ values.”****

510 Page 12, line 407-419. Does Table 3 include the entrainment sensitivity results from Figure 12?

Yes, the errors given in Table 3 account for the range of $\theta_{e,ent}$ and $q_{t,ent}$ measured (red points in figure 10).

515 Interactive comment on “Top-down and Bottom-up aerosol-cloud-closure: towards
understanding sources of uncertainty in deriving cloud radiative flux” by Kevin J. Sanchez et al.
Anonymous Referee #3

Received and published: 25 May 2017

520 The manuscript presents an interesting study of aerosol-cloud-closure in terms of cloud CDNC
and shortwave radiative flux using ground-based and UAV platform measurements, satellite
retrievals at Mace Head, Ireland during summer 2015, as well as a 1-D aerosol-cloud parcel
model simulations. The authors look at CDNC closure between Hoppel CDNC, satellite
525 retrievals, and ACPM simulations, and cloud-top extinction and shortwave radiative flux closure
between UAV measurements and ACPM simulations. The authors find that clouds in decoupled
boundary layer have larger shortwave radiative flux differences between observations and
simulations. More interestingly, the authors find that accounting for cloud-top entrainment in
simulations greatly reduces the radiative flux differences. The manuscript is well written and
530 organized. Overall, the article is suitable for publication in the ACP with some revisions. Below
are some specific comments.

**We thank the reviewer for their comments. Please see below responses to each of the
authors comments and suggestions.**

535 **The authors want to note that values in Table 3 have slightly changed. These changes were
brought about by reviewer #2’s comment to present cloud optical thickness. It was noticed
that the ‘observed’ optical thickness was not consistent between calculations that including
and excluding cloud top entrainment. The observed optical thickness is calculated from the
observed cloud droplet extinction. The observed droplet extinction is calculated by
540 subtracting the simulated cloud droplet extinction and fitted difference in droplet
extinction ($\delta\sigma_{\text{ext}}$) (Figure 8b,d,f). This was necessary to take into account the fact that the
UAV’s often missed portions of the cloud. The linear fit made it possible to fill the gaps.
Since the observations should be consistent, the observations from the fit that excluded
entrainment was compared to simulations with entrainment.**

545 Specific comments:
L77 and 86: the sentences are repeating.

Referenced text:

550 **“Marine boundary layer decoupling is often seen in the tropics and has been
attributed to processes that involve cloud heating and surface cooling as cloud
warming can result from cloud-top entrainment, leading to decoupling of the
boundary layer [Albrecht et al., 1995; Bates et al., 1998; Bretherton et al., 1997]. In
addition, Bretherton and Wyant [1997] have suggested that the decoupling structure
is mainly driven by an increasing ratio of the surface latent heat flux, (i.e.,
555 evaporative cooling at the surface) to the net radiative cooling within the cloud,
while other factors, such as drizzle, the vertical distribution of radiative cooling in
the cloud, and sensible heat fluxes, play less important roles. Turton and Nicholls
[1987] used a two-layer model to show that decoupling can also result from solar
heating of the cloud layer. Nicholls and Leighton [1986] suggested decoupling
results from cloud-top radiative cooling and the resulting eddies do not mix down to
560 the surface. Zhou et al. [2015] showed that the entrainment of the dry warm air
above the inversion could also be the cause. Marine boundary layer decoupling is**

often seen in the tropics and has been attributed to easterlies bringing air over increasing SST, which increases latent cooling and adds negative buoyancy below the cloud layer [Albrecht et al., 1995].”

The text has been modified to the following based on responses from reviewers 1-3 and previously restated in responses to reviewer 1 and 2:

“Marine boundary layer decoupling is often seen in the tropics and has been attributed to processes that involve cloud heating from cloud-top entrainment, leading to decoupling of the boundary layer (Bretherton et al., 1997; Bates et al., 1998; Albrecht et al., 1995; Zhou et al., 2015; Stevens, 2002). In addition, Bretherton and Wyant (1997) have shown that the decoupling structure is mainly driven by a high latent heat flux that results in a large buoyancy jump across the cloud base. This high latent heat flux is attributed to easterlies bringing air over increasing SST, where the boundary layer becomes deeper and more likely to decouple (Albrecht et al., 1995). The cloud layer drives the turbulent motion and a zone of negative buoyancy flux develops below cloud. The turbulent motion is driven by radiative cooling at cloud top, causing air to sink (Lilly, 1968). The zone of negative buoyancy occurs because the deepening of the boundary layer causes the lifting condensation level of the updraft and downdraft to separate. This is important because latent heating in the cloud contributes significantly to the buoyancy in the cloud (Schubert et al., 1979). If this zone of negative buoyancy flux becomes deep enough, it is dynamically favorable for the cloud layer to become decoupled from the cloud layer (Bretherton et al., 1997). Bretherton and Wyant (1997) also show that drizzle can have a substantial impact on enhancing the negative buoyancy flux below cloud, but drizzle is not necessary for decoupling mechanism they proposed. Other factors, such as the vertical distribution of radiative cooling in the cloud, and sensible heat fluxes, play less important roles. Turton and Nicholls (1987) used a two-layer model to show that decoupling can also result from solar heating of the cloud layer; however, only during the day. Furthermore, Nicholls and Leighton (1986) showed observations of decoupled clouds with cloud-top radiative cooling and the resulting in-cloud eddies do not mix down to the surface (further suggesting radiative cooling plays a less important role).”

Section “UAV vertical profiles”: How cloud-top radiative fluxes are measured? It is not illustrated in the manuscript.

There were no airborne direct measurements of cloud-top radiative flux. Cloud-top radiative flux is calculated using extinction measurements from the cloud droplet sensor measurements and from ACPM simulations. The cloud albedo is calculated from extinction (equations 1-3) and the albedo is used to calculate the cloud-top radiative flux. The following text in section 2.4 explains how the cloud-top shortwave radiative flux is calculated: “the shortwave radiative flux (RF) is calculated as $RF = \alpha Q$, where Q is the daily-average insolation at Mace Head and α is the cloud albedo.”

In the “UAV vertical profiles” section the last sentence of the following text was added for clarity: “In-cloud extinction was measured in-situ using a miniature optical cloud droplet sensor developed at the University of Reading [Harrison and Nicoll, 2014]. The sensor operates by a backscatter principle using modulated LED light which is backscattered into a central photodiode. Comparison of the sensor with a Cloud Droplet Probe (DMT)

demonstrate good agreement for cloud droplet diameters $>5\mu\text{m}$ [Nicoll et al., 2016]. **The extinction measurements were used to calculate cloud-top shortwave radiative flux and is further discussed in section 2.4.**

L205: need a reference here.

A reference is included at the end of the sentence: “The model employs a dual moment (number and mass) algorithm to calculate particle growth from one size section to the next for non-evaporating compounds (namely, all components other than water) using an accommodation coefficient of 1.0 [Raatikainen et al., 2013].”

L260: Reference to Hoppel 1979 is not listed. I would suggest giving more details of using D_{min} to estimate CDNC. How accurate is the estimation?

The Hoppel reference has been added.

The last sentence in the following text has been added to explicitly explain how to calculate the Hoppel CDNC: “The dry aerosol particles with diameters greater than the Hoppel D_{min} have undergone cloud processing and are used here to estimate the CDNC. For each of the case study days, Figure 5 demonstrates the aerosol size distribution measurements, from the SMPS and APS, that are used to find the Hoppel D_{min} , Hoppel CDNC and used to initialize the ACPM. The Hoppel CDNC is calculated by integrating the SMPS and APS combined size distributions for aerosol sizes greater than Hoppel D_{min} .”

The Hoppel CDNC is within 30% of both the simulated CDNC and the satellite estimated CDNC.

Figure 6: It is better to add variations of measured and satellite-retrieved CDNC. For comparisons between D_{min} -estimated CDNC and simulated CDNC, they both use ground-based aerosol distribution measurements as input, therefore, these two are not independent.

We do not have measured CDNC, but instead are using the CDNC calculated by the aerosol-cloud parcel model (ACPM). Even though the D_{min} -estimated CDNC and simulated CDNC both use ground-based measurements of the aerosol distributions, the ACPM simulates the supersaturation to determine the critical diameter based on the size and chemical composition of the particles. The critical diameter is not necessarily the same as the D_{min} diameter. The ACPM is the main link between observations and the satellite measurement, which is why both the satellite CDNC and D_{min} -estimated CDNC are compared to the ACPM CDNC. The main purpose of the figure was to show that the satellite CDNC are within 30% of the ACPM CDNC because the error associated with the satellite retrieval method is 30% (Rosenfeld et al., 2016).

L308: 0.3 or 0.5?

The minimum diameter of the OPC is 0.3 microns. This has been corrected in the manuscript.

L326: Even for simulations with 50% decreased cloud-base aerosol, decoupled cases still have greater radiative differences than the coupled cases. Does that mean there are other factors other than aerosol between decoupled and coupled cases that contribute to the radiative differences?

The main reason the radiative flux difference is large is simply because the cloud (D05Sc) is the thinnest cloud, and therefore error's in extinction (from measurement error or error in simulated) have a larger influence on the radiative differences. From equation 2, a small change in a cloud with low optical thickness (thin cloud) has a greater effect on the albedo than a small change in a high optical thickness (thick cloud). Notice the error in extinction for the D05Sc case in table 2 is similar to the C11Sc case even though the error in RF is lower for C11Sc.

Interactive comment on “Top-down and Bottom-up aerosol-cloud-closure: towards understanding sources of uncertainty in deriving cloud radiative flux” by Kevin J. Sanchez et al.
Anonymous Referee #4

Received and published: 1 June 2017

This paper presents results from a variety of measurements during an intensive field campaign at Mace Head in Ireland. It is perhaps unique in comparing estimates of cloud drop number concentration and radiative fluxes at cloud top based on several significantly different methods for a handful of cases during the campaign. Given the disparity among the cases (i.e. cumulus/stratocumulus; coupled/decoupled; adiabatic/sub-adiabatic), as well as the presentation of the results, it is a little unclear how to generalize the results of the study. The most substantive result seems to be the successful application of method for adjusting a parcel model calculation of the cloud top radiative flux to account for dilution of the cloud by entrainment that results in a flux estimate that agrees better with in-situ measurements of cloud extinction. The paper is appropriate for publication in ACP after addressing some minor revision.

We thank the reviewer for their comments. Please see below responses to each of the authors comments and suggestions.

The authors want to note that values in Table 3 have slightly changed. These changes were brought about by reviewer #2’s comment to present cloud optical thickness. It was noticed that the ‘observed’ optical thickness was not consistent between calculations that including and excluding cloud top entrainment. The observed optical thickness is calculated from the observed cloud droplet extinction. The observed droplet extinction is calculated by subtracting the simulated cloud droplet extinction and fitted difference in droplet extinction ($\delta\sigma_{\text{ext}}$) (Figure 8b,d,f). This was necessary to take into account the fact that the UAV’s often missed portions of the cloud. The linear fit made it possible to fill the gaps. Since the observations should be consistent, the observations from the fit that excluded entrainment was compared to simulations with entrainment.

In a couple of places some fairly arbitrary adjustments were made with inconclusive results. For example, in lines 319-322 the authors describe a test where the aerosol concentration imposed on the parcel model is arbitrarily reduced by 50% based on the notion that the aerosol concentration in the cloud layer of a decoupled boundary layer is likely to be less than what was measured at the surface. Yet the the change resulted in little change in the cloud-top radiative flux. How do the authors reconcile the small change in radiative flux for such a larger perturbation of the imposed aerosol concentration with their ultimate conclusion that the main source of error in their bottom-up radiative closure for the decoupled boundary layer cases is the lack of measurements to constrain the CCN concentration in the decoupled cloud layer?

Figure 9 shows the OPC concentration reduced by almost 50% in decoupled layer (compared to the surface based mixed layer), though this is not the same case. The choice of 50% was loosely based on this given there were no other measurements to base this choice on. We have now referred to Figure 9 in the text:

“ACPM simulations were conducted using aerosol concentrations based on the approximate average decoupled to coupled aerosol concentration ratio (50%, Figure 9) to estimate the difference in shortwave radiative flux. “

Previous literature has shown there are cases where CDNC is sensitive to aerosol concentration (aerosol limited) while others are sensitive to updraft velocity (updraft

705 limited). The manuscript discusses the results of decreasing the aerosol concentrations in simulations of both the D05Sc and D06Cu cases. The D06Cu case which has a large range of updraft velocities (0-1.6 m/s) had significantly fewer (42%) CDNC after reducing the aerosol concentration. The D05Sc has significantly lower updraft velocities, ranging from 0-0.3 m s⁻¹, and therefore, is updraft limited. The CDNC is very sensitive at these low updraft velocities, so it is likely that the combined modeled updraft resolution of 0.1 m s⁻¹ and error in updraft velocity measurements is the cause for the large error in shortwave radiative forcing (δ RF) of 33 W m⁻² (Table 2) for the D05Sc case, after accounting for cloud top entrainment.

The following text has been changed to incorporate this information:

715 “For the D05Sc case, simulations with 50% decreased cloud-base aerosol concentrations show only slight differences in δ RF of 2 Wm⁻² and decreases in CDNC of 10%. The decrease in aerosol concentration resulted in increased supersaturation due to the low water uptake from fewer activating droplets. The increased supersaturation caused smaller aerosols to activate (Raatikainen et al., 2013) and therefore, little change in CDNC. The D05Sc case has very low updraft velocities (0-0.3 m s⁻¹). At low updraft velocities, the CDNC is often updraft limited (Reutters et al., 2009). This means the CDNC is very sensitive to the updraft velocities and less sensitive to aerosol concentration. Small errors in updraft velocity and low modeled updraft resolution (0.1 m s⁻¹) likely contributes significantly to the error in this case. The D06Cu was not influenced as much by low water uptake because the CDNC was much higher at 171 cm⁻³ compared to 86 cm⁻³ for D05Sc. 720 The D06Cu the CDNC decreased by 42% and δ RF decreased by 18 Wm⁻². The updraft velocity range for the D06Cu case is significantly higher than the D05Cu case (0-1.6 m s⁻¹). The higher velocities for the D05Sc and greater sensitivity to aerosol concentration suggest this case is aerosol limited (Reutters et al., 2009). Both decoupled cases still have a δ RF greater than the coupled cases.”

730 For the D06Cu case, the 42% decrease in CDNC, significantly reduced δ RF from 74 to 56 w m-2. A δ RF of 56 w m-2 is still high compared to the decoupled cases. It is possible that the difference in aerosol concentration between the coupled and decoupled boundary layer is greater than 50%. We do not have aerosol concentration measurements in the decoupled layer for this case. Also, it is possible that this case experienced some cloud top 735 entrainment. The measured lapse rate for this case was slightly higher (0.1 K km⁻¹) than the adiabatic lapse rate, however this was within instrument error, so cloud top entrainment was not explored. If the heating is offset by long wave cooling (not considered in this paper), then the effect of entrainment may be significant. Note, the two entrainment cases studied both had measured lapse rates that were 1 K km⁻¹ higher than the adiabatic 740 lapse rate.

The following text has been changed to incorporate this information:

745 “The UAV observations show that both C11Sc and D05Sc have sub-adiabatic lapse rate measurements, compared to simulated moist-adiabatic lapse rates within the cloud (Table 2). The difference between the observed and simulated lapse rates therefore suggests a source of heating in the cloud. The sub-adiabatic lapse rate is attributed to cloud-top entrainment by downward mixing of warmer air at cloud-top. The D06Cu case has a slightly sub-adiabatic observed lapse rate (Table 2), however the difference with respect to an adiabatic lapse rate is within instrument error. For this reason, cloud top entrainment is not 750 explored for this case, though it may contribute slightly to the error.”

In the conclusion it is stated that cloud-top entrainment is only observed on 2 out of 13 flight days, and a decoupled boundary layer on only 4 of 13 flight days. It might be valuable to include this in the abstract. While reading the paper, I was struggling to understanding the broader
755 implications. Is there sufficient data to draw a tentative conclusion about the overall sign and/or magnitude of errors in bottom-up forcing calculations based on the surface station data at this location? If this can be addressed in any manner by the authors, then the paper will have substantially greater importance.

After revisiting the statement (that cloud-top entrainment is only observed on 2 out of 13 flight days, and a decoupled boundary layer on only 4 of 13 flight days) we have decided to reworded this statement to more clearly what these statistics are based on:

“Based on airborne observations with UAVs, decoupling of the boundary layer occurred on four of the 13 flight days (two decoupled cloud cases were not discussed due to the lack of in-cloud measurements). However, cloud drop entrainment was only observed on two of those days, limited by the ability to make in-situ measurements. These measurements occurred during the summer, so additional measurements are needed to look at seasonal trends.”

Because the entrainment statistic is limited by measurement capabilities we have decided not to include this in the abstract.

The main broader implications of these results are that cloud-top entrainment and decoupling of the boundary layer lead to over estimation of cloud-top shortwave radiative forcing when using the adiabatic and well mixed boundary layer assumptions, respectively. While we have indicated the magnitude of these errors for the cases presented, there are only a limited number of cases in this manuscript to draw statistics on the occurrence of these scenarios. In order have a Many more case studies are needed to conclude more specific implications for the Mace Head location. Furthermore, similar studies at other locations are necessary to understand global implications.

Top-down and Bottom-up aerosol-cloud-closure: towards understanding sources of uncertainty in deriving cloud radiative flux

K. J. Sanchez^{1,2}, G. C. Roberts^{1,2}, R. Calmer², K. Nicoll^{3,4}, E. Hashimshoni⁵, D. Rosenfeld⁵, J. Ovadnevaite⁶, J. Preissler⁶, D. Ceburnis⁶, C. O'Dowd⁶, L. M. Russell¹

¹Scripps Institution of Oceanography, University of California, San Diego, CA;

²Centre National de Recherches Météorologiques, Toulouse, France;

³Department of Meteorology, University of Reading, UK;

⁴Department of Electronic and Electrical Engineering, University of Bath, UK

⁵The Hebrew University of Jerusalem, Israel;

⁶School of Physics and Centre for Climate and Air Pollution Studies, National University of Ireland Galway, Ireland;

Correspondence to: Kevin J. Sanchez (kjsanche@ucsd.edu)

Abstract. Top-down and bottom-up aerosol-cloud- shortwave radiative flux closures were conducted at the Mace Head atmospheric research station in Galway, Ireland in August 2015. This study is part of the BACCHUS (Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding) European collaborative project, with the goal of understanding key processes affecting aerosol-cloud- shortwave radiative flux closures to improve future climate predictions and develop sustainable policies for Europe. Instrument platforms include ground-based, unmanned aerial vehicles (UAV)¹, and satellite measurements of aerosols, clouds and meteorological variables. The ground-based and airborne measurements of aerosol size distributions and cloud condensation nuclei (CCN) concentration were used to initiate a 1D microphysical aerosol-cloud parcel model (ACPM). UAVs were equipped for a specific science mission, with an optical particle counter for aerosol distribution profiles, a cloud sensor to measure cloud extinction, or a 5-hole probe for 3D wind vectors. UAV cloud measurements are rare and have only become possible in recent years through the miniaturization of instrumentation. These are the first UAV measurements at Mace Head. ACPM simulations are compared to in-situ cloud extinction measurements from UAVs to quantify closure in terms of cloud shortwave radiative flux. Two out of seven cases exhibit sub-adiabatic vertical temperature profiles within the cloud, which suggests that entrainment processes affect cloud microphysical properties and lead to an overestimate of simulated cloud shortwave radiative flux. Including an entrainment parameterization and explicitly calculating the entrainment fraction in the ACPM simulations both improved cloud-top radiative closure. Entrainment reduced the difference between simulated and observation-derived cloud-top shortwave radiative flux (δRF) by between 25 W m^{-2} and 60 W m^{-2} . After accounting for entrainment, satellite-derived cloud droplet number concentrations (CDNC) were within 30% of simulated CDNC. In cases with a well-mixed boundary layer, δRF is less than no greater than 20 W m^{-2} after accounting for cloud-top entrainment, compared to less than and up to 50 W m^{-2} when entrainment is not taken into account. In cases with a decoupled boundary layer, cloud microphysical properties are inconsistent with ground-based aerosol measurements, as expected, and δRF is as high as 88 W m^{-2} , even high ($> 30 \text{ W m}^{-2}$) after accounting for cloud-top entrainment. This

¹ The regulatory term for UAV is Remotely Piloted Aircraft (RPA).

work demonstrates the need to take in-situ measurements of aerosol properties for cases where the boundary layer is decoupled as well as consider cloud-top entrainment to accurately model stratocumulus cloud radiative flux.

1

820 **1 Introduction**

One of the greatest challenges in studying cloud effects on climate are that the clouds are literally out of reach. Many ground-based measurement sites have a long historical record that are useful for identifying climatological trends, however, it is difficult to quantify such trends in cloud microphysical and radiative properties at these stations based solely on remote sensing techniques such as radar and lidar. *In-situ* aerosol measurements at the surface are often used
825 to estimate cloud properties aloft, but the simulations used to estimate above surface conditions require many idealized assumptions such as a well-mixed boundary layer and adiabatic parcel lifting. Satellites have the advantage to infer cloud properties over a much larger area than ground-based observations; however, they can only see the upper most cloud layer and satellites need *in-situ* observations to improve their retrievals. In this study, we combine ground-based and airborne measurements with satellite observations to determine cloud radiative properties and compare these
830 results to an aerosol-cloud parcel model (ACPM) to identify sources of uncertainty in aerosol-cloud interactions.

The atmospheric research station at Mace Head has been a research platform for studying trace gases, aerosols and meteorological variables since 1958 (O'Connor et al., 2008). The station is uniquely exposed to a variety of air masses, such as clean marine air and polluted European air. Over the long history of observations and numerous field-
835 campaigns held at the Mace Head research station, few airborne field experiments have been conducted. During the PARFORCE campaign in September 1998, aerosol and trace gas measurements were made to map coastal aerosol formation (O'Dowd et al., 2001). During the second PARFORCE campaign in June 1999, measurements of sea spray plumes were made on an aircraft installed with a Lidar (Kunz et al., 2002). In the NAMBLEX campaign in August 2002, flights were conducted to measure aerosol chemical and physical properties in the vicinity of Mace Head (Heard
840 et al., 2006; Norton et al., 2006; Coe et al., 2006). None of the research flights thus far have studied aerosol-cloud interactions and cloud radiative properties at Mace Head.

For ground-based observations, it is often assumed that measured species are well-mixed throughout the boundary layer. Often this assumption is valid and many observational studies have shown that models which use ground-based
845 measurements can accurately simulated cloud droplet number concentrations (CDNC) (Russell and Seinfeld, 1998; Conant et al., 2004; Fountoukis et al., 2007), making bottom-up closure a viable method for predicting cloud properties. Closure is defined here as the agreement between observations and model simulations of CDNC and cloud-top [shortwave](#) radiative flux. This well-mixed boundary layer simplification, however, has been shown to be inaccurate in many field experiments (e.g., the Atlantic Stratocumulus Transition Experiment (ASTEX) (Albrecht et

al., 1995); and the Aerosol Characterization Experiments, ACE1 (Bates et al., 1998) and ACE2 (Raes et al., 2000). Previous studies at Mace Head have shown that decoupled boundary layers were observed with scanning backscatter lidar measurements (Kunz et al., 2002; Milroy et al., 2012). Such decoupled layers often contain two distinct cloud layers, distinguished as a lower layer within the well-mixed ~~surface layer~~ surface-mixed layer and a higher decoupled ~~residual~~ layer between the free troposphere and ~~surface layer~~ surface-mixed layer (Kunz et al., 2002; Milroy et al., 2012; Stull, 1988). General characteristics associated with decoupled boundary layers are a weak inversion, a decrease in aerosol concentration relative to the ~~surface layer~~ surface-mixed layer, and more commonly occurring in relatively deep marine boundary layers (> 1400 m) (Jones et al., 2011). Dall'Osto et al (2010) showed the average height of the surface-mixed layer, over Mace Head, varies from 500 m to 2000 m, and the decoupled layers have heights ranging from 1500 m to 2500 m. Marine boundary layer decoupling is often seen in the tropics and has been attributed to processes that involve cloud heating ~~and surface cooling as cloud warming can result~~ from cloud-top entrainment, leading to decoupling of the boundary layer (Bretherton et al., 1997; Bates et al., 1998; Albrecht et al., 1995; Zhou et al., 2015; Stevens, 2002). In addition, Bretherton and Wyant (1997) have ~~suggested~~ shown that the decoupling structure is mainly driven by ~~an high increasing ratio of the surface~~ latent heat flux, ~~(i.e., evaporative cooling at the surface)~~ that results in a large buoyancy jump across the cloud base. This high latent heat flux is attributed to easterlies bringing air over increasing SST, where the boundary layer becomes deeper and more likely to decouple (Albrecht et al., 1995). The cloud layer drives the turbulent motion and a zone of negative buoyancy flux develops below cloud. The turbulent motion is driven by radiative cooling at cloud top, causing air to sink (Lilly, 1968). The zone of negative buoyancy occurs because the deepening of the boundary layer causes the lifting condensation level of the updraft and downdraft to separate. This is important because latent heating in the cloud contributes significantly to the buoyancy in the cloud (Schubert et al., 1979). If this zone of negative buoyancy flux becomes deep enough, it is dynamically favorable for the cloud layer to become decoupled from the cloud layer (Bretherton et al., 1997). Bretherton and Wyant (1997) also show that drizzle can have a substantial impact on enhancing the negative buoyancy flux below cloud, but drizzle is not necessary for decoupling mechanism they proposed. to the net radiative cooling within the cloud, while oOther factors, such as ~~drizzle~~, the vertical distribution of radiative cooling in the cloud, and sensible heat fluxes, play less important roles. Turton and Nicholls (1987) used a two-layer model to show that decoupling can also result from solar heating of the cloud layer; ~~however, only during the day. Furthermore,~~ Nicholls and Leighton (1986) ~~suggested showed observations of~~ decoupled clouds ~~ing results from~~ with cloud-top radiative cooling and the resulting in-cloud eddies do not mix down to the surface (further suggesting radiative cooling plays a less important role). ~~Zhou et al. showed that the entrainment of the dry warm air above the inversion could also be the cause. Marine boundary layer decoupling is often seen in the tropics and has been attributed to easterlies bringing air over increasing SST, which increases latent cooling and adds negative buoyancy below the cloud layer.~~ Russell et al. (1998) and Sollazzo et al. (2000) showed that, in a decoupled atmosphere the two distinct layers have similar characteristics (e.g., aerosol and trace gases composition), ~~withbut~~ different aerosol concentrations ~~thatand~~ gradually mix with each other, entraining mixing air from the ~~surface layer~~ surface-mixed layer into the decoupled ~~residual~~ layer and vice versa. These previous studies also show that aerosol concentrations in the ~~upper residual layer~~ decoupled layer are lower than those in the ~~well-mixed~~ surface-mixed layer implying an overestimation in cloud shortwave radiative flux when using ground-based aerosol measurements.

Satellite measurements of microphysical properties, such as CDNC, have the potential to be independent of ground-based measurements, and therefore be reliable for studying decoupled clouds. Satellite estimates of CDNC have only become possible recently due to the increased resolution in measurements (Rosenfeld et al., 2012; Rosenfeld et al., 2014; Rosenfeld et al., 2016; Painemal and Zuidema, 2011). Therefore, current measurements still require ground-based validation until the method is further developed.

The focus of this manuscript is on the top-down closure between satellite retrievals and airborne measurements of cloud microphysical properties, as well as, traditional bottom-up closure coupling below and in-cloud measurements of cloud condensation nuclei (CCN), updraft, and cloud microphysical properties. *In-situ* measurements of CDNC are not available so bottom-up closure is expressed in terms of cloud-top shortwave radiative flux rather than CDNC and top-down closure of satellite CDNC is compared to ACPM simulated CDNC. The methods section describes how observations were collected, as well as the methods for estimating CDNC with satellite measurements and calculating radiative flux shortwave radiative flux with the ACPM. The results section summarizes the bottom-up and top-down closure for coupled and decoupled clouds and quantifies the differences in cloud radiative flux shortwave radiative flux for cases that were affected by cloud-top entrainment.

2 Methods

The August 2015 campaign at the Mace Head research station (Galway, Ireland; 53.33°N, 9.90°W) focused on aerosol-cloud interactions at the north eastern Atlantic Ocean by coupling ground-based *in-situ* and remote sensing observations with airborne and satellite observations. This section summarizes the measurements used for this study and the model used to simulate the observations.

2.1 Ground-based measurements

At the Mace Head research site, aerosol instruments are located in the laboratory at about 100 m from the coastline. They are connected to the laminar flow community air sampling system, which is constructed from a 100 mm diameter stainless-steel pipe with the main inlet at 10 m above ground level, so that samples are not impacted by immediate coastal aerosol production mechanisms, such as wave breaking and biological activity (Norton et al., 2006; O'Dowd et al., 2004; Coe et al., 2006; Rinaldi et al., 2009; O'Dowd et al., 2014). The performance of this inlet is described in Kleefeld et al. (2002). Back trajectories during the period of the experiment show that the origin of air masses is predominantly from the North Atlantic; therefore, the air masses sampled at Mace Head generally represent clean open ocean marine aerosol. Mace Head contains a variety of aerosol sampling instrumentation, spanning particle diameter range of 0.02 μm and 20 μm . Size spectral measurements are performed at a relative humidity < 40% using Nafion driers. Supermicron particle size distributions were measured using an Aerodynamic Particle Sizer (APS, TSI model 3321, 0.5 < D_p < 20 μm). The remaining submicron aerosol size range was retrieved from a scanning mobility particle sizer (SMPS, 0.02 < D_p < 0.5 μm), comprised of a differential mobility analyzer (DMA, TSI model 3071), a condensation particle counter (TSI model 3010, D_p > 10 nm), and a Kr-85 aerosol neutralizer (TSI 3077).

Cloud condensation nuclei (CCN) measurements were performed with a miniature Continuous Flow Stream-wise Thermal Gradient Chamber, which measures the concentration of activated CCN over a range of supersaturations (Roberts and Nenes, 2005). During this study, the supersaturation range spanned 0.2% to 0.82%. Aerosol hygroscopicity was calculated using κ -Köhler theory (Petters and Kreidenweis, 2007) with the sampled CCN concentrations at a particular supersaturation and corresponding integrated aerosol number concentration at a critical diameter (Roberts et al., 2001). Figure 1 shows time series of CCN spectra and aerosol number size distributions throughout the campaign. The ground-based remote sensing measurements utilized in this study are the MIRA36, 35.5 GHz Ka-band Doppler cloud radar (Melchionna et al., 2008;Goersdorf et al., 2015) to obtain vertical velocity distributions at cloud-base and the Jenoptik CHM15K ceilometer (Heese et al., 2010;Martucci et al., 2010) to obtain cloud base height.

2.2 UAV vertical profiles

The UAV operations were conducted directly on the coast about 200 meters from the Mace Head research station. UAVs were used to collect vertical profiles of standard meteorological variables, temperature (IST, Model P1K0.161.6W.Y.010), pressure (Bs rep GmbH, Model 15PSI-A-HGRADE-SMINI), and relative humidity (IST, P14 Rapid-W), as well as aerosol size distributions with an optical particle counter (OPC, Met One Model 212-2), cloud droplet extinction (Harrison and Nicoll, 2014), updraft velocity at cloud base with a 5-hole probe. A list of the various UAV flights and their instrumentation is given in Table 1. Measurement errors for the relative humidity and temperature sensors are $\pm 5\%$ and $\pm 0.5^\circ\text{C}$ respectively. As RH sensors are not accurate at high RH ($> 90\%$), the measured values have been scaled such that RH measurements are 100% in a cloud. At altitudes where the UAV is known to be in-cloud (based on *in-situ* cloud extinction measurements) the air mass is considered saturated (RH $\sim 100\%$). The temperature and relative humidity sensors are protected from solar radiative heating by a thin-walled aluminum shroud positioned outside of the surface layer of the UAV. A helical cone, mounted in front of the sensors, ejects droplets to protect the sensors. The temperature measurements for both cases in which cloud-top entrainment is explored (see section 3.2) are verified to remain in stratocumulus clouds throughout the ascents and descents, and are not affected by evaporative cooling. The temperature and relative humidity measurements were used to initialize the ACPM below cloud. The UAVs were flown individually in separate missions up to 1.5 hours and each UAV was instrumented to perform a specific science mission (referred to here as aerosol, cloud, 3D winds).

The OPC measured aerosol number size distributions in eight size bins between 0.3 and 10 μm diameter. Aerosols were sampled via a quasi-isokinetic shrouded inlet mounted on the nose of the UAV. Aerosols samples were heated upon entering the UAV ($\Delta T > 5\text{ K}$ due to internal heating by the electronics), reducing the relative humidity of the sampled air to less than 60% and decreased with height ($< 50\%$ above 150 m) before aerosol size was measured. Figure 2 shows a two-instrument redundancy cross check between ground-based APS and UAV OPC measurements (collected between 40 m agl and 80 m agl) of aerosol sizes are in agreement ($r^2 = 0.48$).

In-cloud extinction was measured in-situ using a miniature optical cloud droplet sensor developed at the University of Reading (Harrison and Nicoll, 2014). The sensor operates by a backscatter principle using modulated LED light

which is backscattered into a central photodiode. Comparison of the sensor with a Cloud Droplet Probe (DMT) demonstrate good agreement for cloud droplet diameters $>5\mu\text{m}$ (Nicol et al., 2016). The extinction measurements were used to calculate cloud-top shortwave radiative flux and is further discussed in section 2.4.

Finally, a 5-hole probe for measuring 3-dimensional wind vectors was mounted on a third UAV. The 3D wind vectors are determined by subtracting the UAV motion given by an inertial measurement unit (IMU) from the total measured flow obtained by differential pressures in the 5-hole probe (Wildmann et al., 2014; Lenschow and Spyers-Duran, 1989; Calmer et al., 2017). UAV 5-hole probe measurements were collected along 6 km long straight and level legs at cloud base. Normalized cloud radar vertical velocity distributions are compared to vertical wind distributions obtained from the UAV in Figure 3. The positive updraft velocities in Figure 3 are used to initialize the ACPM to produce simulated cloud droplet size distributions throughout the depth of the cloud. The droplet distributions for each updraft velocity are averaged and weighted by the probability distribution of the measured positive velocities. Differences in results when using the cloud radar updrafts versus the UAV 5-hole probe updrafts (Figure 3) are discussed in section 3.1.2.

2.3 Satellite measurements

Research flights with the UAV were conducted in conjunction with satellite overpasses to compare retrieved CDNC and maximum supersaturation (S_{max}) with ACPM simulated values using the Suomi NASA Polar-orbiting Partnership satellite. The satellite estimations of CDNC and S_{max} are based on methods described by Rosenfeld et al. (2012; 2014; 2016), which are briefly summarized in the following paragraph. The case selection criteria for satellite observations required the overpass to occur at a zenith angle between 0° and 45° to the east of the ground track, to have convective development that spans at least 6 K of cloud temperature from base to top (~ 1 km thick), and to not precipitate significantly. In-situ observations were often of thin clouds (< 1 km thick), and the satellite observations consist primarily of the more developed clouds in the same system.

To obtain CDNC, cloud droplet effective radius profiles were extracted from the Suomi NASA Polar-orbiting Partnership satellite. Figure 4 shows an image from the Suomi visible infrared imaging radiometer suite on 21 August overlapped on a map of western Ireland. The vertical profile in figure 4 shows satellite retrieved and ACPM simulated effective radius. To estimate the CDNC, the satellite effective radius (Figure 4) is first converted to mean volume radius (r_v) using a linear relationship (Freud et al., 2011). Next, it is assumed that any mixing that occurred between the cloud and cloud-free air was inhomogeneous; this implies that the actual r_v is equal to the adiabatic r_v . CDNC can be calculated by dividing the adiabatic water content in the cloud by r_v (Rosenfeld et al., 2012; Beals et al., 2015). The cloud base height and pressure was used to calculate the adiabatic water content. Cloud base height and pressure were obtained from the height of the NCEP reanalysis of the cloud base temperature, as retrieved from satellite. The cloud base height was validated against the ceilometer. Freud et al. (2011) showed that the inhomogeneous assumption resulted in an average over-estimate in CDNC of 30%, so the CDNC is reduced by 30% to account for the bias with the assumption. Finally, to calculate S_{max} the cloud base updraft velocity, from the UAV or cloud radar, is needed and

when paired with the CDNC, it can be used to empirically calculate S_{\max} (Rosenfeld et al., 2012; Pinsky et al., 2012). The methodology was validated by Rosenfeld et al. (2016).

2.4 Aerosol-cloud parcel model simulations

A detailed description of the aerosol-cloud parcel model (ACPM) is presented in Russell and Seinfeld (1998) and Russell et al. (1999). The ACPM is based on a fixed-sectional approach to represent the (dry) particle size domain, with internally mixed chemical components ~~and externally mixed types of particles~~. Aerosols are generally internally mixed at Mace Head ~~owing to because there were no immediate strong sources of pollution~~ ~~lack of aerosol sources~~. The model employs a dual moment (number and mass) algorithm to calculate particle growth from one size section to the next for non-evaporating compounds (namely, all components other than water) using an accommodation coefficient of 1.0 (Raatikainen et al., 2013). The dual moment method is based on Tzivion et al. (1987) to allow accurate accounting of both aerosol number and mass, and incorporates independent calculations of the change in particle number and mass for all processes other than growth. The model includes a dynamic scheme for activation of particles to cloud droplets. Liquid water is treated in a moving section representation, similar to the approach of Jacobson et al. (1994), to account for evaporation and condensation of water in conditions of varying humidity. In sub-saturated conditions, aerosol particles below the cloud base are considered to be in local equilibrium with water vapor pressure (i.e., relative humidity < 100%).

Coagulation, scavenging, and deposition of the aerosol were included in the model but their effects are negligible given the relatively short simulations used here (<2 h) and low marine total aerosol particle concentrations (<500 cm³; Dp > 10 nm). ~~Feingold et al. (2013) showed that autoconversion and accretion rates are negligible for the modeled values of LWC and CDNC except for the C21Cu case, which had LWC > 1 g m⁻³. Thus, droplet number loss by collision coalescence can be neglected for all cases except for the C21Cu case.~~ Aerosol hygroscopicity as a function of size (and supersaturation) is determined from CCN spectra and aerosol size distributions as mentioned in Section 3.1, and is used as model input. The ACPM is also constrained by measured temperature profiles, cloud base height, and updraft velocity distribution (Figure 3). The in-cloud lapse rate is assumed to be adiabatic, unless specified otherwise, so simulation results represent an upper bound on CDNC and liquid water content that is unaffected by entrainment. To account for release of latent heat in the cloud, the vertical temperature gradient is calculated as $dT = -(gwdt + Ldq_l)/c_p$, where dT is change in temperature for the vertical displacement of an air parcel, g is acceleration due to gravity, w is updraft velocity at cloud base, dt is time step, L is latent heat of water condensation, q_l is liquid water mixing ratio, and c_p is specific heat of water (Bahadur et al., 2012). A weighted ensemble of positive updraft velocities measured with the cloud radar and UAV 5-hole probe were applied to the ACPM [Sanchez et al. 2016].

The simulated cloud droplet size distribution is used to calculate the shortwave cloud extinction. Cloud extinction is proportional to the total droplet surface area (Hansen and Travis, 1974; Stephens, 1978) and is calculated from,

$$\sigma_{ext} = \int_0^{\infty} Q_{ext}(r) \pi r^2 n(r) dr \quad (1)$$

where r is the radius of the cloud droplet, $n(r)$ is the number of cloud droplets with a radius of r , and $Q_{ext}(r)$ is the Mie efficiency factor, which asymptotically approaches 2 for water droplets at large sizes ($r > 2 \mu\text{m}$).

Finally, the ~~radiative flux~~shortwave radiative flux (RF) is calculated as $RF = \alpha Q$, where Q is the daily-average insolation at Mace Head and α is the cloud albedo. α is estimated using the following equation (Geresdi et al., 2006;Bohren and Battan, 1980)

$$\alpha = \frac{(\sqrt{3}(1-g)\tau)}{(2+\sqrt{3}(1-g)\tau)}; \quad (2)$$

where τ is the cloud optical depth defined as

$$\tau = \int_0^H \sigma_{ext}(h) dh; \quad (3)$$

and H is the cloud height or thickness and g , the asymmetric scattering parameter, is approximated as 0.85 based on Mie scattering calculations for supermicron cloud drops. RF is calculated for both, simulated cloud extinction and measured UAV extinction.

3 Results/Discussion

3.1 Closure of CDNC and cloud-top ~~radiative flux~~shortwave radiative flux

For this study, closure is defined as the agreement between observations and model simulations of CDNC and cloud-top ~~radiative flux~~shortwave radiative flux. In-situ measurements of clouds were made by UAVs on 13 days during the campaign. Of these, a subset of six are chosen here for further analysis, which includes comparison with satellite CDNC as well as simulation of cloud properties with the ACPM (Table 2). The remaining days with UAV measurements did not contain sufficient cloud measurements for analysis. A satellite overpass occurred on each of the six days, however only 4 of the days contained clouds that were thick enough to analyze with the satellite. The 10 August cases experienced a light drizzle, so ACPM simulations were not conducted for this case, however analysis with satellite imagery was still conducted. On 5 August, two cloud layers were examined, for a total of 7 case studies shown in Table 2. Aerosols were occasionally influenced by anthropogenic sources, however, the cases shown consist of aerosol of marine origin with concentrations under 1000 cm^{-3} (Figure 1).

3.1.1 Ground-based measurement closure

The columns in Table 2 represent the different cases for both clouds that were (a) coupled with and (b) decoupled from the surface BL (“C” and “D” in case acronym, respectively). The first row in Table 2 includes the state of atmospheric mixing, the date, the type of cloud present, and the acronym used for each case. The top portion of Table 2 consists of *in-situ* airborne measurements, the bottom portion presents ACPM simulation results and their relation to *in-situ* cloud extinction and satellite-retrieved observations. The ground-based *in-situ* measurements in Table 2 include the Hoppel minimum diameter² (D_{\min}), as well as the aerosol concentration of aerosol with diameters greater

² The Hoppel minimum diameter is the diameter with the lowest aerosol concentration between Aitken mode and accumulation mode.

than the Hoppel D_{\min} and the inferred in-cloud critical supersaturation (S_c) (Hoppel, 1979). The dry aerosol particles with diameters greater than the Hoppel D_{\min} have undergone cloud processing and are used here to estimate the CDNC.

For each of the case study days, Figure 5 demonstrates the aerosol size distribution measurements, from the SMPS and APS, that are used to find the Hoppel D_{\min} , Hoppel CDNC and used to initialize the ACPM. The Hoppel CDNC is calculated by integrating the SMPS and APS combined size distributions for aerosol sizes greater than Hoppel D_{\min} .

Figure 6 shows Hoppel-based CDNC estimates are within 30% of simulated CDNC for the 7 cases. The presence of the Hoppel minimum occurs on average at 80 nm diameter throughout the campaign (Figure 1b, 5) implying in-cloud supersaturations near 0.25 % using a campaign averaged hygroscopicity (K) of 0.42, which is in agreement with K values observed in the North Atlantic marine planetary boundary layer in Pringle et al. (2010).

3.1.2 UAV measurements closure

Figure 7 displays vertical profiles of meteorological parameters, as well as OPC aerosol number concentration (N_{OPC} ; $D_p > 0.35 \mu\text{m}$) and cloud extinction from two flights (23 and 27) on 11 August. The UAV used on flight 23 (conducted between 12:00 UTC and 12:47 UTC), contained the cloud sensor for cloud extinction measurements and flight 27 (conducted between 16:58 UTC and 17:33 UTC) contained the OPC for droplet size distribution measurements. During this time period the cloud base reduced from 1200 m on flight 23 to 980 m on flight 27, but cloud depth remained approximately the same. In the OPC vertical profiles, in Figure 7d, an aerosol layer is shown above the cloud at ~1400 m. OPC measurements are removed inside cloud layers (as aerosol data is contaminated by cloud droplets), hence the gap in OPC data in Figure 7d. The OPC and temperature measurements, in Figure 7a and d, are used to show if the boundary layer was coupled (well-mixed) or if it was decoupled. The state of the boundary layer and the OPC and temperature measurements are further discussed at the end of this section. The observed temperature and relative humidity profiles, in Figure 7a and b, are also used to initialize the ACPM. In-situ cloud extinction measurements, in Figure 7c, are then compared to the ACPM simulated cloud extinction (Figure 8c).

Figure 8a, c and e present the observed and simulated adiabatic cloud extinction profile for three of the case studies (C11Sc, D05Sc and C21Cu)³. The measurements are binned into in-cloud, cloud-free, and cloud-transition (or cloud-edge) samples. Many clouds had a small horizontal extent making it difficult for the UAVs to remain in cloud as they ascended and descended in a spiral pattern. Also, high horizontal winds ($10 - 15 \text{ m s}^{-1}$) will generally move the cloud outside the field of measurement of the aircraft very quickly. For cases where the UAV did not remain in-cloud throughout the ascent or descent, the in-cloud samples are identified as the largest extinction values at each height and are seen in the measurements as a cluster of points (Figure 8e). Since lateral mixing with cloud-free air exerts an influence near the cloud edges, the cloud-transition air is not representative of the cloud core and adiabatic simulations. The amount of sampling within individual clouds varied from case to case, but the UAVs were generally able to make multiple measurements of the same cloud during each vertical profile. C11Sc was unique in that it involved stratocumulus clouds with a large horizontal extent, allowing the UAV to remain entirely in-cloud during the upward and downward vertical profiles around a fixed waypoint. Figure 8f shows how the difference between simulated and

³ C/D – coupled / decoupled; xx – date in August 2015; Sc / Cu – stratocumulus / cumulus cloud

observed extinction ($\delta\sigma_{\text{ext}}$) is calculated throughout the cloud based on a discrete sampling of in-cloud measurements. It is not certain that the UAV measured the cloud core for cumulus cases so $\delta\sigma_{\text{ext}}$ is an upper limit (Table 2).

1100 All ACPM simulation results, including those in Table 2, use the cloud radar updraft velocity as input and not the 5-hole probe updraft velocity because 5-hole probe updraft velocities are not available for all cases. Nonetheless, the differences in ACPM simulated ~~radiative flux~~shortwave radiative flux between using the 5-hole probe and cloud radar updraft velocities (Figure 3) is less than 3 W m^{-2} for the four cases that had both measurements.

1105 The integrated effect of $\delta\sigma_{\text{ext}}$ leads to a difference in cloud observed and simulated ~~radiative flux~~shortwave radiative flux (δRF) for both clouds that were coupled with and decoupled from the surface boundary layer (Table 2). Figure 9, presents a vertical profile of N_{OPC} and equivalent potential temperature. OPC measurements within a thin cloud layer at $\sim 2000 \text{ m}$ are removed. N_{OPC} and equivalent potential temperature (θ_e) clearly illustrate this decoupling as shown in an example vertical profile (Figure 9) at 900 and 2200 m.asl, with the latter representing the inversion

1110 between the boundary layer top and free troposphere. N_{OPC} decreases from an average of 31 cm^{-3} to 19 cm^{-3} at the same altitude as the weak inversion (700-1000 m). In this study, decoupled boundary layers are often observed and aerosol number concentrations ($D_p > 0.3 \mu\text{m}$) in the decoupled layer were $44\% \pm 14\%$ of those measured at the ground. While N_{OPC} are not directly representative of CCN concentrations, a reduction in aerosol number with height (and potential differences in hygroscopicity) will nonetheless affect aerosol-cloud closures, and ultimately, the cloud

1115 radiative properties. Similarly, Norton et al. (2006) showed results from the European Centre for Medium-Range Weather Forecasts (ECMWF) model re-analysis in which surface winds at Mace Head are often decoupled from synoptic flow and, therefore, the air masses in each layer have different origins and most likely different aerosol properties. Consequently, the CCN number concentrations measured at the surface do not represent those in the higher decoupled cloud layer, which ultimately dictates cloud ~~radiative flux~~shortwave radiative flux in the region and δRF

1120 in Table 2. While aerosol profiles were not collected by UAVs for the decoupled cases presented in Table 2, the θ_e profiles and ceilometer measurements show evidence of boundary layer decoupling. These two decoupled cases have larger $\delta\sigma_{\text{ext}}$ than the coupled boundary layer cases in this study, leading to larger cloud-top δRF as well. ACPM simulations were conducted using aerosol concentrations based on the approximate average decoupled to coupled aerosol concentration ratio (50%, Figure 9) to estimate the difference in ~~radiative flux~~shortwave radiative flux. For

1125 the D05Sc case, simulations with 50% decreased cloud-base aerosol concentrations show only slight differences in δRF of 2 Wm^{-2} and decreases in CDNC of 10%. The decrease in aerosol concentration resulted in increased supersaturation due to the low water uptake from fewer activating droplets. The increased supersaturation caused smaller aerosols to activate (Raatikainen et al., 2013) and therefore, little change in CDNC. The D05Sc case has very low updraft velocities ($0\text{-}0.3 \text{ m s}^{-1}$). At low updraft velocities, the CDNC is often updraft limited (Reutters et al., 2009). This means the CDNC is very sensitive to the updraft velocities and less sensitive to aerosol concentration. Small errors in updraft velocity and low modeled updraft resolution (0.1 m s^{-1}) likely contributes significantly to the error in this case. The D06Cu was not influenced as much by low water uptake because the CDNC was much higher at 171 cm^{-3} compared to 86 cm^{-3} for D05Sc. The D06Cu the CDNC decreased by 42% and δRF decreased by 18 Wm^{-2}

1130

². The updraft velocity range for the D06Cu case is significantly higher than the D05Cu case (0-1.6 m s⁻¹). The higher velocities for the D05Sc and greater sensitivity to aerosol concentration suggest this case is aerosol limited (Reutters et al., 2009). Both decoupled cases still have a δRF greater than the coupled cases.

3.1.3 Satellite measurements closure

The satellite and simulated CDNC and S_{max} measurements are presented in the bottom of Table 2. The method for satellite retrieval of cloud properties could not be used for cases when cloud layers were too thin – which, unfortunately was the situation during the flights with the decoupled cloud layers. Nonetheless, Figure 4 shows the satellite image used to identify the clouds to calculate CDNC for C11Sc. Satellite retrieved cloud-base height and temperature are verified by ground-based ceilometer and temperature measurements. Figure 6 shows the top-down closures demonstrate that satellite-estimated CDNC and simulated CDNC are within a $\pm 30\%$ expected concentrations, which is limited by the retrieval of effective radius (Rosenfeld et al., 2016). The stratocumulus deck at the top of a well-mixed boundary layer (C11Sc) shows evidence of cloud-top inhomogeneous entrainment (see section 3.2). Freud et al. (2011) found that the inhomogeneous mixing assumption used to derive CDNC from satellite measurements resulted in an average over-estimate in CDNC of 30% (considering an adiabatic cloud droplet profile). Consequently, satellite-retrieved CDNC is reduced by 30% to account for the inhomogeneous entrainment assumption, which does not necessarily reflect the actual magnitude of entrainment in the clouds. For the C11Sc case, before the correction, proposed by Freud et al. (2011), is applied the satellite derived CDNC (83 cm⁻³) is within 30% of the ACPM CDNC (88 cm⁻³), similar to the other cases (Figure 6). However, if the correction is applied, the satellite derived CDNC (58 cm⁻³) is not within 30% of the ACPM CDNC. This indicates cloud top entrainment for the C11Sc case is already inhomogeneous and For example, in the C11Sc case, in-situ observations do indeed show cloud-top inhomogeneous entrainment; consequently, the usual 30% reduction in CDNC to correct for the inhomogeneous assumption does not need to should not be applied (Table 2). Both stratocumulus cases (C11Sc, D05Sc) with cloud-top entrainment (Table 2) are similar to a case studied by Burnet and Brenguier (2007), in which cloud-top entrainment resulted in inhomogeneous mixing. In the following section, C11Sc and D05Sc are reanalyzed to include the effect of cloud-top entrainment on simulated cloud properties using the inhomogeneous mixing assumption.

3.2 Entrainment

Based on the ground-based and UAV measurements, ACPM simulations over-estimate cloud ~~radiative flux~~ shortwave radiative flux significantly for three cases (C11Sc, D05Sc, D06Cu). Section 3.1.2 identified that clouds in decoupled layers (D05Sc, D06Cu) have smaller radiative effects than predicted based on ground-based observations as aerosol (and CCN) number concentrations in the decoupled layer are often smaller than in the ~~surface layer~~ surface-mixed layer. In this section, cloud-top entrainment is also shown to influence the radiative properties of two sub-adiabatic stratocumulus clouds, C11Sc and D05Sc.

The UAV observations show that both C11Sc and D05Sc have sub-adiabatic lapse rate measurements, compared to simulated moist-adiabatic lapse rates within the cloud (Table 2). The difference between the observed and simulated lapse rates therefore suggests a source of heating in the cloud. The sub-adiabatic lapse rate is attributed to cloud-top

entrainment by downward mixing of warmer air at cloud-top (e.g., Figure 7a). The D06Cu case has a slightly sub-adiabatic observed lapse rate (Table 2), however the difference with respect to an adiabatic lapse rate is within instrument error. For this reason, cloud top entrainment is not explored for this case, though it may contribute to the error.

Further evidence of cloud-top entrainment is shown through conserved variable mixing diagram analysis. In previous studies, a conserved variable mixing diagram analysis was used to show lateral or cloud-top entrainment by showing linear relationships between observations of conserved variables (Paluch, 1979; Neggers et al., 2002; Burnet and Brenguier, 2007). Paluch (1979) first observed a linear relationship of conservative properties (total water content, q_t , and liquid water potential temperature, θ_l) between cumulus cloud cores and cloud edge, to show the cloud-free source of entrained air. Paluch (1979), Burnet and Brenguier (2007), Roberts et al. (2008) and, Lehmann et al. (2009) observed decreases in CDNC and liquid water content in cumulus clouds as a function of distance from the cloud cores that indicate inhomogeneous mixing at the cloud edge. Burnet and Brenguier (2007) also show that q_t is linearly proportional to liquid water potential temperature specifically for a stratocumulus cloud with cloud-top entrainment and inhomogeneous mixing. Direct observations of CDNC and liquid water content were not measured at Mace Head, so direct comparisons of CDNC and q_t with Paluch (1979) and Burnet and Brenguier (2007) cannot be investigated here. However, UAV measurements of cloud extinction (Eq. 1), which are related to CDNC ($CDNC = \int_0^\infty n(r) dr$) and liquid water content ($LWC = \int_0^\infty \frac{4}{3} \rho \pi r^3 n(r) dr$, ρ is liquid water density), were measured and are found to be systematically lower than the adiabatic simulated cloud extinction (Figure 8).

To apply the cloud-top mixing, a fraction of air at cloud-base and a fraction of air above cloud-top are mixed, conserving q_t and θ_e . The fraction of air from cloud-base and cloud-top is determined with the measured equivalent potential temperature,

$$\theta_{e,c}(z) = \theta_{e,ent}X(z) + \theta_{e,CB}(1 - X(z)) \quad (4)$$

where $\theta_{e,c}(z)$ is the equivalent potential temperature in cloud as a function of height, $\theta_{e,ent}$ is the equivalent potential temperature of the cloud-top entrained air, $\theta_{e,CB}$ is the equivalent potential temperature of air at cloud base, and $X(z)$ is the fraction of cloud-top entrained air as a function of height (referred to as the entrainment fraction). $\theta_{e,ent}$, $\theta_{e,c}(z)$ and $\theta_{e,CB}$ are measured parameters by the UAV and are not affected by latent heating from evaporation or condensation. The equivalent potential temperature, by definition, accounts for the total water content by including the latent heat released by condensing all the water vapor. Eq. (4) takes into account latent heating caused by evaporation of droplets. By rearranging Eq. (4), the entrained fraction is calculated as

$$X(z) = \frac{\theta_{e,c}(z) - \theta_{e,CB}}{\theta_{e,ent} - \theta_{e,CB}} \quad (5)$$

Figure 10a and b present the relationships between two conservative variables measured by the UAV (water vapor content, q_v , and θ_e) for C11Sc and D05Sc. The q_v is derived from relative humidity measurements and is equivalent to the q_t for sub-saturated, cloud-free air (i.e., < 100% RH). The cloud-free air is shown in blue in Figure 10, where the

below cloud measurements have lower θ_e than in-cloud and the above cloud measurements have ~~higher~~ θ_e than in-cloud.

Figure 11 shows the relative humidity and θ_e profiles used in Figure 10. For both C11Sc and D05Sc, $\theta_{e,c}(z)$ is directly measured in-cloud, and q_v and θ_e exhibit an approximately linear relationship (Figure 10; Eq. 4). The linear relationship of q_t and θ_e (between the non-mixed sources of air indicated by orange circles in Figure 10) is assumed to be a result of the cloud reaching a steady-state, with air coming from cloud-base and cloud-top (e.g. cloud lifetime \gg mixing time). The observed in-cloud q_v in Figure 10a and b is less than the conservative variable q_t , however, the figure also includes q_t based on simulated adiabatic (marked with an 'X') and cloud-top entrainment (dashed black line) conditions. Under adiabatic conditions q_t and θ_e do not change in the cloud, which is why the adiabatic simulations only consists of one point in Figure 10. Eq. (4) is used to derive the simulated cloud-top entrainment conditions (Figure 10a and b), where the fraction entrained is used to calculate q_t and shows a linear relationship between q_t and θ_e . Measurements above cloud-top (RH < 95%), labeled entrained air, with $q_v > 5.1 \text{ g kg}^{-1}$ and $q_v > 6.5 \text{ g kg}^{-1}$ are used to represent the properties of the entrained air for C11Sc and D05Sc, respectively (Figure 10). These conditions were chosen because these values are on the mixing line, between the non-mixed sources identified by the orange circles.

Figure 12 shows the sensitivity of the simulated cloud extinction profile, for the 11 August case, based on measurement uncertainties related to the entrained q_v and θ . The key variable for identifying the entrained fraction (Eq. 5), $\theta_{e,ent}$, is a function of q_v and θ , so a decrease in either parameter results in a proportional decrease in $\theta_{e,ent}$. Eq. (5) shows that entrainment fraction becomes more sensitive to the uncertainty related to the measurement of θ_e as the difference between $\theta_{e,ent}$ and $\theta_{e,CB}$ approaches zero. This is also shown in Figure 12 where σ_{ext} is more sensitive to lower entrained q_t and θ values.

Table 3 shows $\delta\sigma_{ext}$, δRF , and CDNC for two cases with cloud-top entrainment (C11Sc and D05Sc) using two methods of accounting for the cloud top entrainment. One method (labeled the 'inhomogeneous mixing entrainment method' in Table 3) applies the entrainment fraction calculated in Eq. (5) and the other an entrainment parameterization, presented by Sanchez et al. (2016). The entrainment parameterization constrains the ACPM simulation to use the observed in-cloud lapse rate instead of assuming an adiabatic lapse rate. This is labeled the 'measured-lapse-rate lapse rate adjustment' entrainment method in Table 3. In the sub-adiabatic cloud cases (C11Sc and D05Sc), the measured in-cloud lapse rate is lower than the adiabatic lapse rate, which leads to the condensation of less water vapor and subsequent activation of fewer droplets in the ACPM simulation. Similarly, when applying the inhomogeneous mixing entrainment method, the dryer and warmer entrained air (from above cloud-top) leads to evaporation of liquid water in the cloud. Previous observations of stratocumulus cloud-top mixing suggest the entrainment is inhomogeneous (Burnet and Brenguier, 2007; Beals et al., 2015), which implies that time scales of evaporation are much less than the time scales of mixing, such that a fraction of the droplets are evaporated completely and the remaining droplets are

unaffected by the entrainment. The net decrease in CDNC subsequently results in less extinction of solar radiation compared to the purely adiabatic simulation.

The inclusion of inhomogeneous ~~cloud-top~~mixing entrainment improved the ACPM accuracy for both C11Sc and D05Sc using the measured lapse-rate and entrainment fraction methods (Figure 8, Table 3). After accounting for inhomogeneous entrainment, δRF decreased from 88 Wm^{-2} to ~~47-33~~ Wm^{-2} and 48 Wm^{-2} to ~~20-14~~ Wm^{-2} for D05Sc and D11Sc, respectively, using the entrainment fraction method. D05Sc simulations still yields significant δRF even after accounting for inhomogeneous mixing entrainment, likely because the cloud is in a decoupled BL, as noted in Section 3.1.2 to exhibit lower aerosol concentrations than those measured at the surface. The CDNC presented in Table 3 represents the CDNC at cloud base and did not change after applying the entrainment fraction method, however, the CDNC decreases with height for the entrainment fraction method rather than remain constant with height. Finally, the ~~measured-lapse-rate-entrainment~~lapse rate adjustment entrainment method [Sanchez et al., 2016] does improve ACPM accuracy between in-situ and satellite-retrieved cloud optical properties relative to the adiabatic simulations, but has greater $\delta\sigma_{\text{ext}}$ throughout the cloud than the ~~entrained-fraction-mixing~~inhomogeneous mixing entrainment method. For the ~~measured-lapse rate~~ adjustment entrainment method δRF decreased from 88 Wm^{-2} to ~~6-18~~ Wm^{-2} and 48 Wm^{-2} to 32 Wm^{-2} for D05Sc and D11Sc respectively. The ~~measured-lapse-rate~~lapse rate adjustment entrainment ~~parameterization-method~~ resulted in lower δRF than the purely adiabatic simulations, however, δRF was minimized by directly accounting for the entrainment fraction.

4 Conclusions

This work presents measurements conducted in August 2015 at the Mace Head Research Station in Ireland, from multiple platforms including ground-based, airborne and satellites. As part of the BACCHUS (Impact of Biogenic versus Anthropogenic emissions on Clouds and Climate: towards a Holistic UnderStanding) European collaborative project, the goal of this study is to understand key processes affecting aerosol-cloud-shortwave radiative flux interactions. Seven cases including cumulus and stratocumulus clouds were investigated to quantify aerosol-cloud interactions using ground-based and airborne measurements (bottom-up closure), as well as cloud microphysical and radiative properties using airborne measurements and satellite retrievals (top-down closure). An aerosol-cloud parcel model (ACPM) was used to link the ground-based, airborne and satellite observations, and to quantify uncertainties related to aerosols, cloud microphysical properties, and resulting cloud optical properties.

ACPM simulations represent bottom-up and top-down closures within uncertainties related to satellite retrievals for conditions with a coupled boundary layer and adiabatic cloud development. For these conditions, the difference in ~~radiative-flux~~shortwave radiative flux between simulations and *in-situ* observed parameters is ~~no greater~~less than 20 W m^{-2} . However, when entrainment and decoupling of the cloud layer occur, the ACPM simulations overestimate the cloud ~~radiative-flux~~shortwave radiative flux. Of the seven cases, two of the observed clouds occurred in a decoupled layer, resulting in differences in observed and simulated ~~radiative-flux~~shortwave radiative flux (δRF) of 88 Wm^{-2} and 74 Wm^{-2} for the decoupled stratocumulus case on 5 August (D05Sc) and the decoupled cumulus case on 6 August

(D06Cu) cases respectively. Adiabatic ACPM simulations resulted in a maximum cloud-top δRF value of 20 W m^{-2} for coupled boundary layer cases and 74 W m^{-2} for the decoupled boundary layer cases, after accounting for cloud-top entrainment. The reduction in aerosol concentrations in the decoupled layer compared to ground-based measurements is a factor in overestimating decoupled cloud-top ~~radiative flux~~_{shortwave radiative flux} with the ACPM, however simulations with 50% decreased aerosol concentrations show only slight differences δRF of ~~23~~ W m^{-2} and decreases in CDNC of 10% for D05Sc. For D06Cu δRF decreased by 18 W m^{-2} and the CDNC decreased by 42%. Even after decreasing the aerosol concentration by 50% both decoupled cases have δRF values significantly higher than the coupled boundary layer cases ($< 20 \text{ W m}^{-2}$).

For the cases with cloud-top entrainment, D05Sc and the coupled stratocumulus case on 11 August (C11Sc), liquid water content is one of the major factors in overestimating cloud-top ~~radiative flux~~_{shortwave radiative flux} with the ACPM. For these cases, the measured in-cloud lapse rates are lower than adiabatic lapse rates, suggesting a source of heat due to entrainment of warmer, drier air from above the cloud. Furthermore, linear relationships between conservative variables, simulated total water vapor, q_t , and equivalent potential temperature, θ_e , also suggest mixing between air at cloud-base and cloud-top. For D05Sc, after accounting for cloud top entrainment by applying the entrainment fraction δRF decreased from 88 W m^{-2} to ~~33~~₄₆ W m^{-2} . For the coupled boundary layer case with entrainment (C11Sc) the δRF decreases from 48 W m^{-2} to ~~20~~₄₄ W m^{-2} after accounting for cloud top entrainment with the entrainment fraction.

Based on airborne observations with UAVs, decoupling of the boundary layer occurred on four of the 13 flight days (~~two decoupled cloud cases were not discussed due to the lack of in-cloud measurements~~). However, cloud drop entrainment was only observed on two of those days, limited by the ability to make in-situ measurements. These measurements occurred during the summer, so additional measurements are needed to look at seasonal trends. These cases illustrate the need for *in-situ* observations to quantify entrainment mixing and cloud base CCN concentrations particularly when the mixing state of the atmosphere is not known. ~~Even greater discrepancies between the surface and decoupled layer CCN concentrations will occur in the presence marine biogenic sources such as tidal regions and local anthropogenic (O'Dowd, 2002).~~ Using ground-based observations to model clouds in decoupled boundary layers and not including cloud top entrainment are shown to cause significant differences between observations and simulation radiative forcing and therefore, should be included in large scale modeling studies to accurately predict future climate forcing.

UAV measurements were coordinated with 13 days of satellite overpasses and cloud microphysical properties were retrieved for four of the cases. When accounting for entrainment, the differences between simulated and satellite-retrieved CDNC are within the expected 30% accuracy of the satellite retrievals (Rosenfeld et al., 2016). However, in-situ measurements are necessary to refine satellite retrievals to allow cloud properties to be studied on larger spatial scales.

Data availability

All data is available by contacting the corresponding author or through the following link: www.umr-cnrm.fr/spip.php?article1040

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Table 1. UAV research flights conducted at Mace Head, Ireland and measured parameters in 2015. Flight start and end times are in UTC. Suomi NASA Polar-orbiting Partnership satellite overpasses occurred at approximately 13:00 UTC. Measurements include relative humidity (RH), temperature (T), pressure (P), 3-dimensional wind vectors (3D Winds), optical particle counter (OPC) and cloud sensor measurements of cloud droplet extinction.

Date	Flight	Start Time	End Time	RH	T	P	3D Winds	OPC	Cloud
30-Jul	4	12:41	13:19	x	x	x		x	
30-Jul	5	14:00	14:44	x	x	x			x
30-Jul	6	16:04	16:42	x	x	x		x	
01-Aug	7	11:30	12:13	x	x	x		x	
01-Aug	8	12:35	13:16	x	x	x			x
01-Aug	9	14:00	15:20	x	x	x	x		
01-Aug	10	15:54	16:43	x	x	x		x	
05-Aug	11	11:47	12:29	x	x	x			x
05-Aug	13	13:36	14:26	x	x	x	x		
05-Aug	14	14:42	15:29	x	x	x			x
06-Aug	16	11:55	12:37	x	x	x			x
06-Aug	17	13:51	15:16	x	x	x	x		
10-Aug	19	13:41	14:10	x	x	x			x
10-Aug	20	14:42	15:45	x	x	x	x		
10-Aug	21	16:00	16:45	x	x	x			x
11-Aug	23	12:00	12:47	x	x	x			x
11-Aug	24	13:11	14:05		x	x	x		
11-Aug	25	14:25	15:10	x	x	x			x
11-Aug	26	15:29	16:22		x	x	x		
11-Aug	27	16:58	17:33		x	x		x	
15-Aug	29	12:19	13:03	x	x	x		x	
15-Aug	30	13:46	14:31		x	x	x		
15-Aug	31	15:08	16:14	x	x	x			x
16-Aug	32	12:30	13:20	x	x	x		x	
16-Aug	33	13:40	14:00	x	x	x		x	
17-Aug	34	11:30	12:24	x	x	x			x
17-Aug	35	13:45	14:34	x	x	x		x	
21-Aug	36	12:21	13:12		x	x		x	
21-Aug	37	13:40	14:25	x	x	x			x
21-Aug	38	15:17	16:26	x	x	x	x		
21-Aug	39	16:53	17:27	x	x	x			x
22-Aug	40	9:29	10:12	x	x	x			x
22-Aug	41	10:47	11:37	x	x	x		x	
22-Aug	42	12:52	13:53	x	x	x	x		
22-Aug	43	14:22	14:59	x	x	x		x	
27-Aug	45	10:21	11:10	x	x	x		x	
27-Aug	46	11:27	12:13	x	x	x			x
27-Aug	47	13:11	13:45			x			x
27-Aug	48	15:09	15:23	x	x	x	x		
27-Aug	49	17:20	17:50	x	x	x		x	
28-Aug	50	14:25	14:49	x	x	x		x	

Table 2. UAV observations of cloud heights and temperatures and cloud property estimates based on ground measurements. Ground-based Hoppel minimum diameter (D_{\min}) is used to estimate CDNC. ACPM simulation and satellite results are also presented, as well as differences between simulated and observation-derived cloud-top extinction and cloud-top radiative flux. Case abbreviations include if they are coupled (C) or decoupled (D), the day of the month and cloud types, cumulus (Cu) or stratocumulus (Sc).

	Coupled BL					Decoupled BL	
	01Aug Cumulus (C01Cu) ^a	05 Aug Cumulus (C05Cu)	10 Aug ^b Cumulus (C10Cu)	11 Aug ^c StratoCu (C11Sc)	21 Aug ^d Cumulus (C21Cu)	05 Aug ^c StratoCu (D05Sc)	06 Aug Cumulus (D06Cu)
<i>In-situ</i> Ground-based and UAV Measurements							
Cloud-base height (m)	800	430	650	1200	460	1490	2180
Cloud-base temperature (°C)	7.4 ±0.1	10.6 ±0.2	8.1 ±0.1	3.7 ±0.1	10.4 ±0.1	6.5 ±0.2	-2.1 ±0.2
Cloud-top height (m)	1040	710	1720	1460	960	1630	2400
Cloud-top temperature (°C)	5.7 ±0.1	8.7 ±0.2	1.8 ±0.1	2.4 ±0.2	7.6 ±0.1	5.8 ±0.2	-3.1 ±0.4
Measured lapse rate in-cloud (K km ⁻¹)	5.7	6.1	5.1	4.7	6.0	4.1	6.3
Number of cloud layers	1	2 ^g	1	1	1	2 ^h	2 ^h
Hoppel D_{\min} (nm)	74 ±6	78 ±16	73 ±8	83 ±7	83 ±5	78 ±16	80 ±9
Hoppel D_{\min} CDNC (> Hoppel D_{\min} , cm ⁻³)	129 ±5	69 ±8	105 ±11	87 ±5	94 ±12	69 ±8	164 ±13
Measured cloud τ	-	11.7	-	8.3	29.1	1.3	4.9
Hoppel minimum critical supersaturation (S_{crit})	0.43 ±0.03	0.61 ±0.10	0.37 ±0.11	0.37 ±0.05	0.41 ±0.10	0.61 ±0.10	0.31 ±0.06
ACPM Simulation and Satellite-derived Cloud Properties ^e							
Simulated moist adiabatic lapse rate (K km ⁻¹)	5.0	4.5	4.9	5.7	4.5	5.1	6.4
Simulated κ Cloud-top droplet r_e (μm)	10.3 ±0.1	14.4±0.3	-	11.3 ±0.2	14.2 ±0.4	10.0 ±0.1	8.2 ±0.2
Simulated cloud τ	-	13.2 ±1.9	-	18.7 ±2.7	42.1 ±11.2	4.4 ±0.5	9.0 ±1.1
Cloud-top extinction difference ($\delta\sigma_{\text{ext}}$, km ⁻¹)	-	11 ±25	-	36 ±12	52 ±42	37 ±6	34 ±7
Cloud-top <u>shortwave</u> radiative flux difference (δRF , W m ⁻²) ^f	-	11 ±26	-	48 ±11	20 ±6	88 ±8	74±12
Simulated CDNC (cm ⁻³)	135 ±16	60 ±12	105 ±18	88 ±12	105 ±31	86 ±10	171 ±17
Satellite estimated CDNC (cm ⁻³)	109	-	85	58 (83) ⁱ	104	-	-
Simulated S_{max} (%)	0.45 ±0.09	0.45 ±0.18	0.36 ±0.15	0.36 ±0.09	0.40 ±0.20	0.76 ±0.04	0.33 ±0.06
Satellite estimated S_{max} (%)	0.34	-	0.27	0.48	0.34	-	-

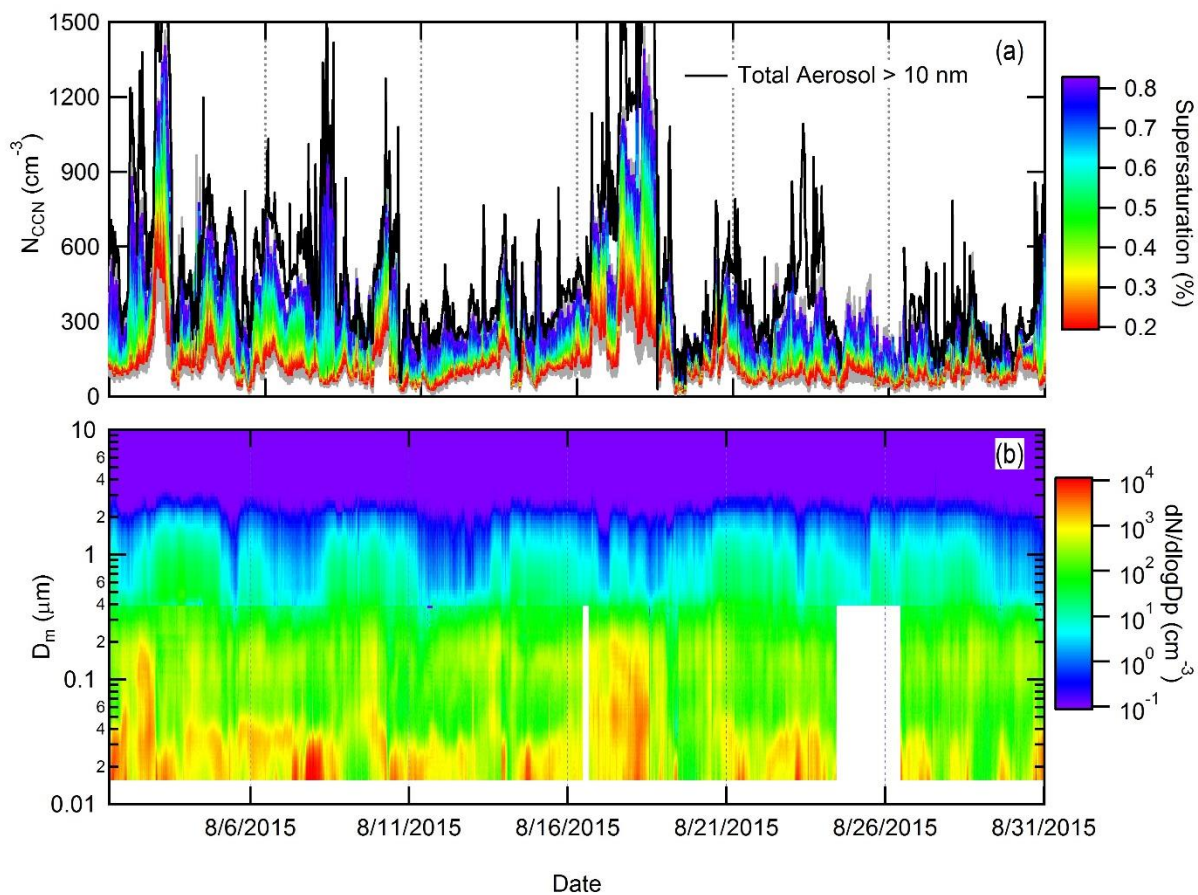
1575^a C/D – coupled / decoupled; xx – date in August 2015; Sc / Cu – stratocumulus / cumulus cloud
^b Precipitation occurred on 10 Aug.
^c Accounting for entrainment improves model / measurement closure (Table 2).
^d The C21Cu case is susceptible to droplet coalescence due to its high liquid water content (Feingold et al., 2013).
^e The error includes the potential error of $\pm 20\%$ in updraft velocity and the standard error of the CCN concentration measurements.
1580^f The difference between the observed (calculated from UAV extinction measurements) and simulated radiative flux. The error includes the potential error of $\pm 20\%$ in updraft velocity and the standard error of the CCN concentration measurements.
^g The measurements and results in this column represent the lower of the two clouds.
^h Altitude of top cloud level that is used to calculate cloud radiative flux.
ⁱ Excluding the correction for the inhomogeneous entrainment assumption in parentheses

Table 3. Results of the application of entrainment fraction and the measured lapse rate entrainment parameterization for two clouds with observed cloud-top entrainment.

Entrainment method	Coupled BL (C11Sc)		Decoupled BL (D05Sc)	
	Homogeneous mixing entrainment	Lapse rate adjustment	Homogeneous mixing entrainment	Lapse rate adjustment
Cloud-top extinction difference ($\delta\sigma_{\text{ext}}$, km^{-1})	16 \pm 10	23 \pm 11	16 \pm 5	26 \pm 6
Simulated <u>cloud</u> τ	10.1 \pm 1.5	10.3 \pm 1.6	2.2 \pm 0.3	3.5 \pm 0.5
Cloud-top <u>shortwave</u> radiative flux difference (δRF, W m^{-2})^a	20 \pm 16	32 \pm 17	33 \pm 9	61 \pm 12
Cloud base simulated CDNC^b	88 \pm 12	83 \pm 12	86 \pm 10	68 \pm 10

^a The difference between the observed (calculated from UAV extinction measurements) and simulated shortwave radiative flux. The error includes the potential error of $\pm 20\%$ in updraft velocity and the standard error of the CCN concentration measurements.

^b The simulated CDNC is unchanged at the cloud base for the entrainment fraction method, however the CDNC decreases with height.



1590 **Figure 1.** Time series for the month of August 2015 at Mace Head Ireland of ground-based CCN concentrations (top) and merged SMPS and APS number size distributions (bottom).

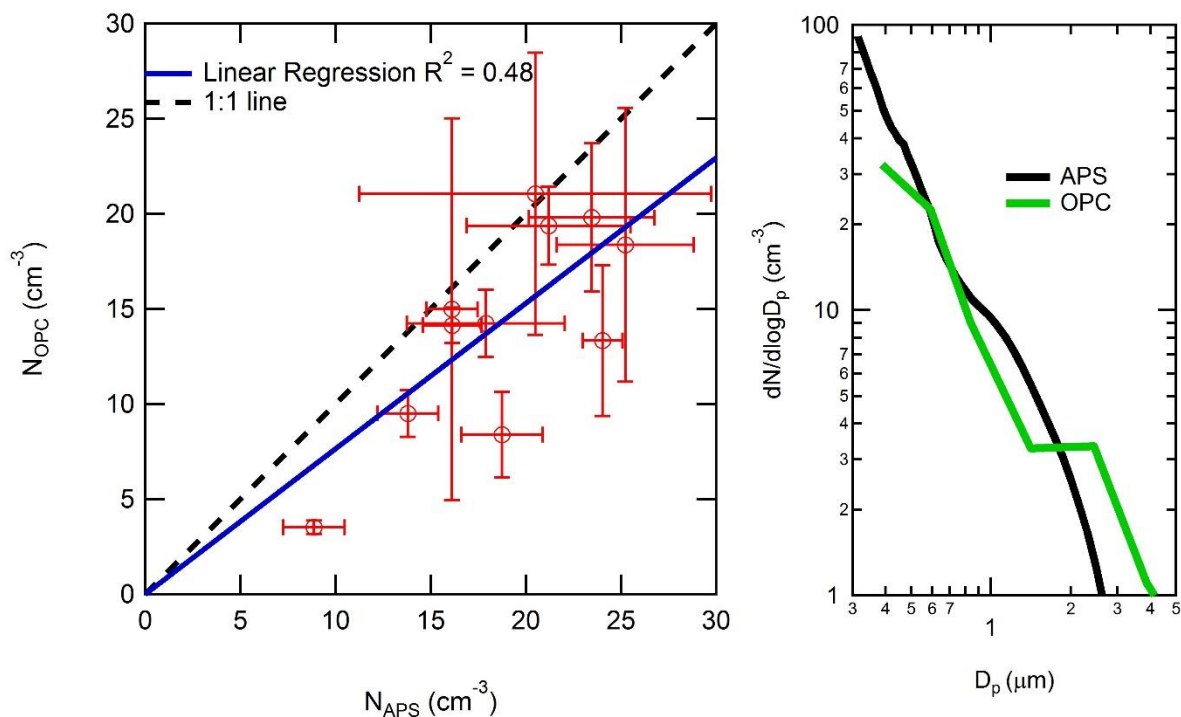


Figure 2. OPC concentrations with particle diameters (D_p) greater than 0.35 μm (left) from 11 UAV research flights, listed in Table 1, plotted against APS concentrations ($D_p > 0.35 \mu\text{m}$) at Mace Head Research Station (red circles). Error bars represent ± 1 standard deviation. The points are fit with a linear regression (blue line). OPC data was averaged between 40 and 80 m asl. Averaged OPC and APS number size distributions averaged for the 11 flights (right).

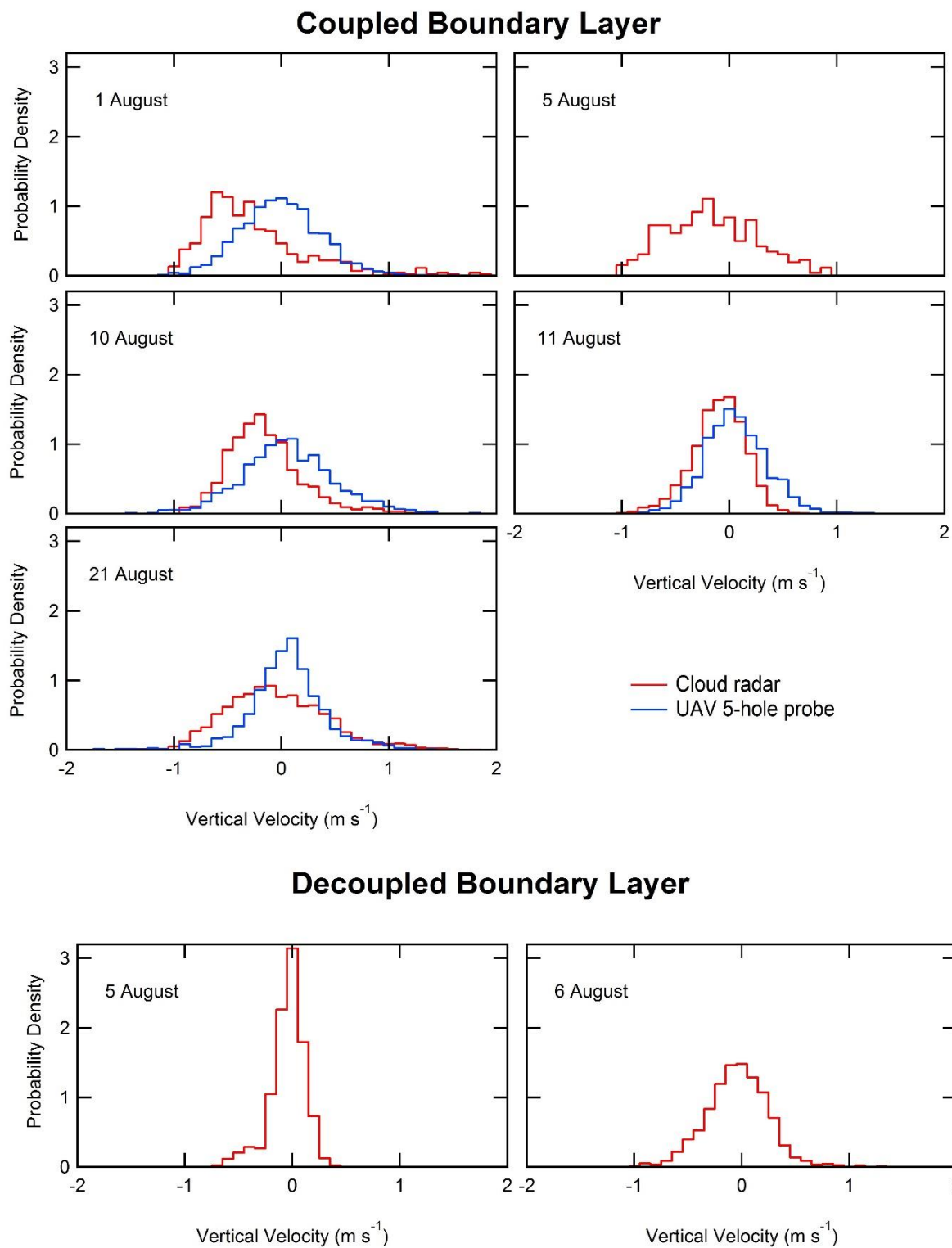
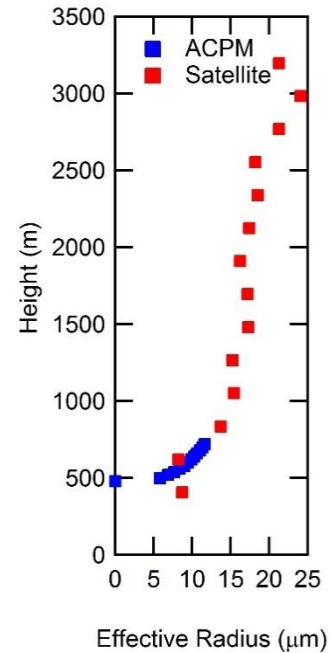
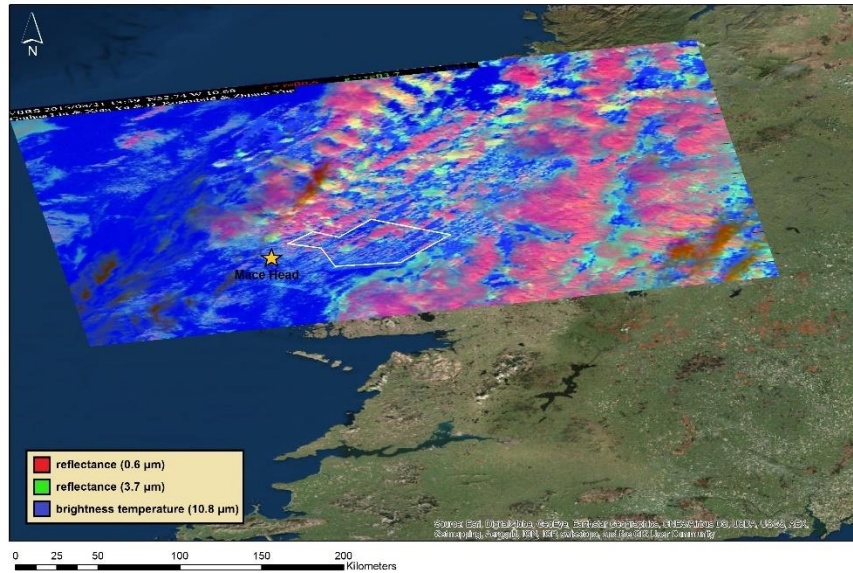


Figure 3. Normalized observed vertical velocity distributions measured by the cloud radar and UAV for each case presented in Table 2.



1605 **Figure 4.** Suomi NPP satellite RGB composite image for 21 August 2015 (left). Mace Head Research Station and UAV flight location are indicated by the yellow star. The white polygon represents the zone for retrieving cloud properties – which is represented by the profile of cloud effective radius (right). Effective radius profiles are presented for both the Suomi NPP satellite (red) and the ACPM (blue).

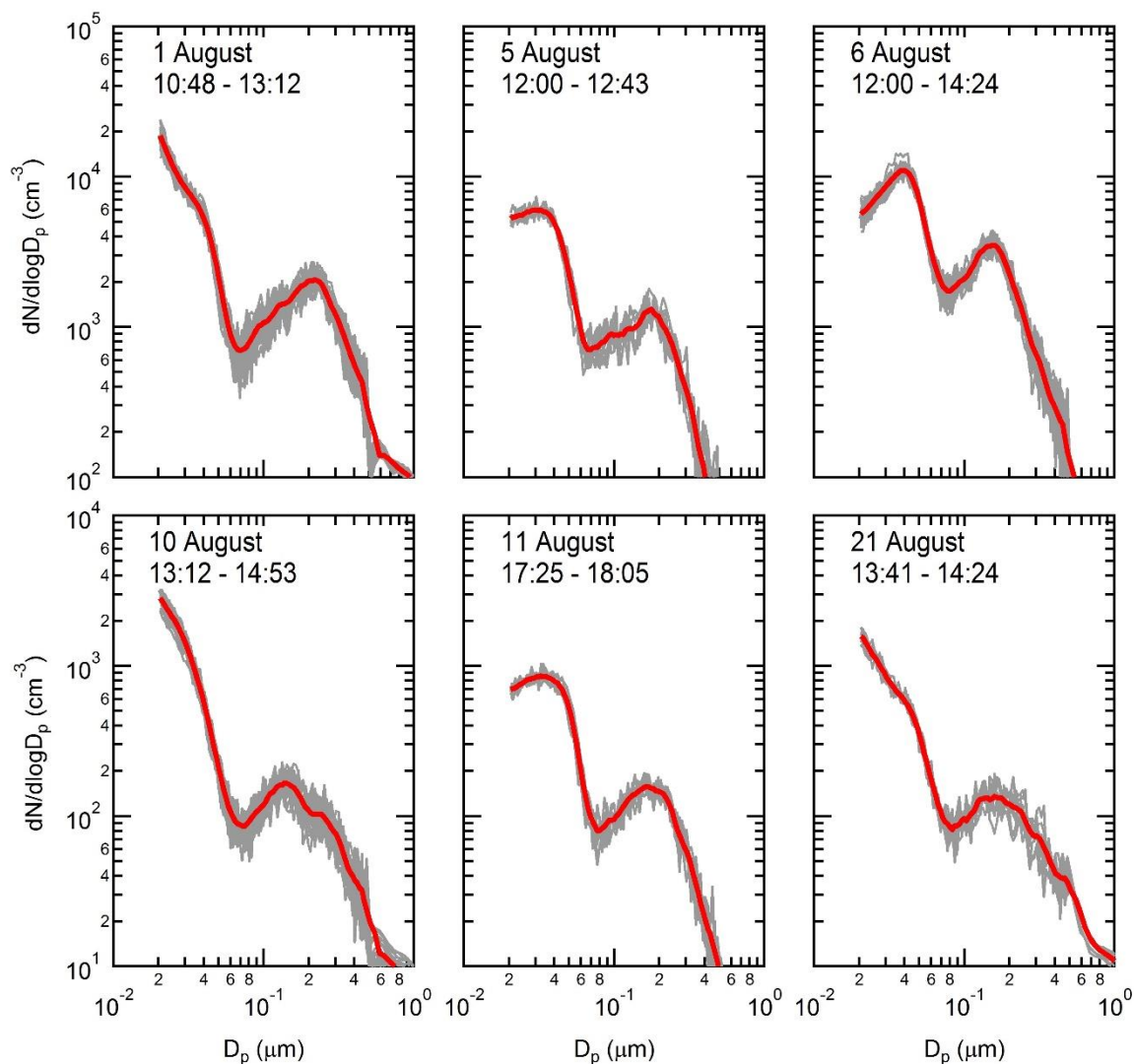
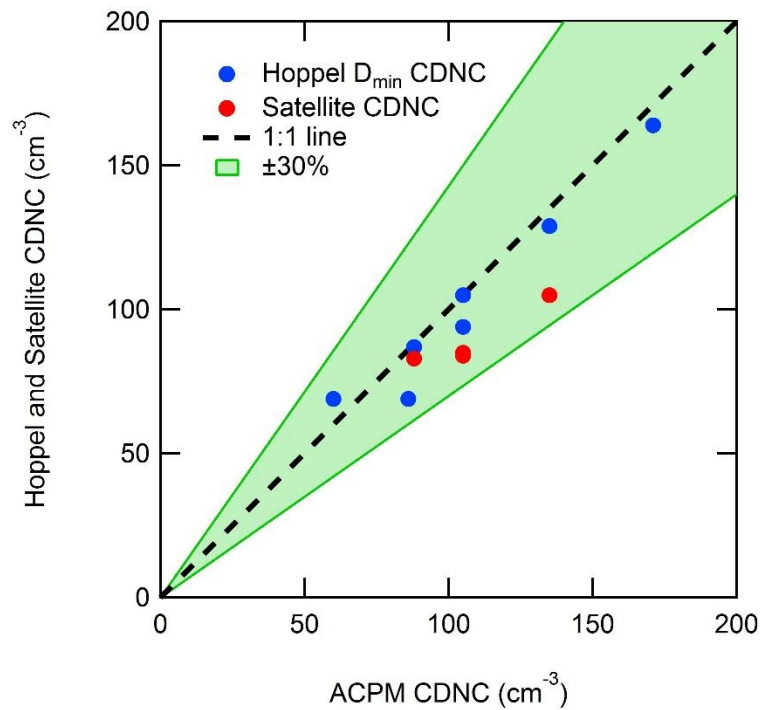


Figure 5. SMPS and APS derived size distributions used for each case study in Table 2. The 5 August size distribution is used for both the coupled and decoupled case. Individual distributions (grey) are from the indicated time ranges in the figure. The time ranges are in UTC. Average distributions are shown in red.



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Figure 6. Comparison of simulated CDNC from ACPM with both Hoppel minimum diameter (D_{\min}) derived CDNC (blue) and satellite estimated CDNC (red). CDNC plotted are from the listed cloud cases in Table 2. The green shaded region represents Hoppel and Satellite CDNCs within 30% of ACPM simulation CDNC.

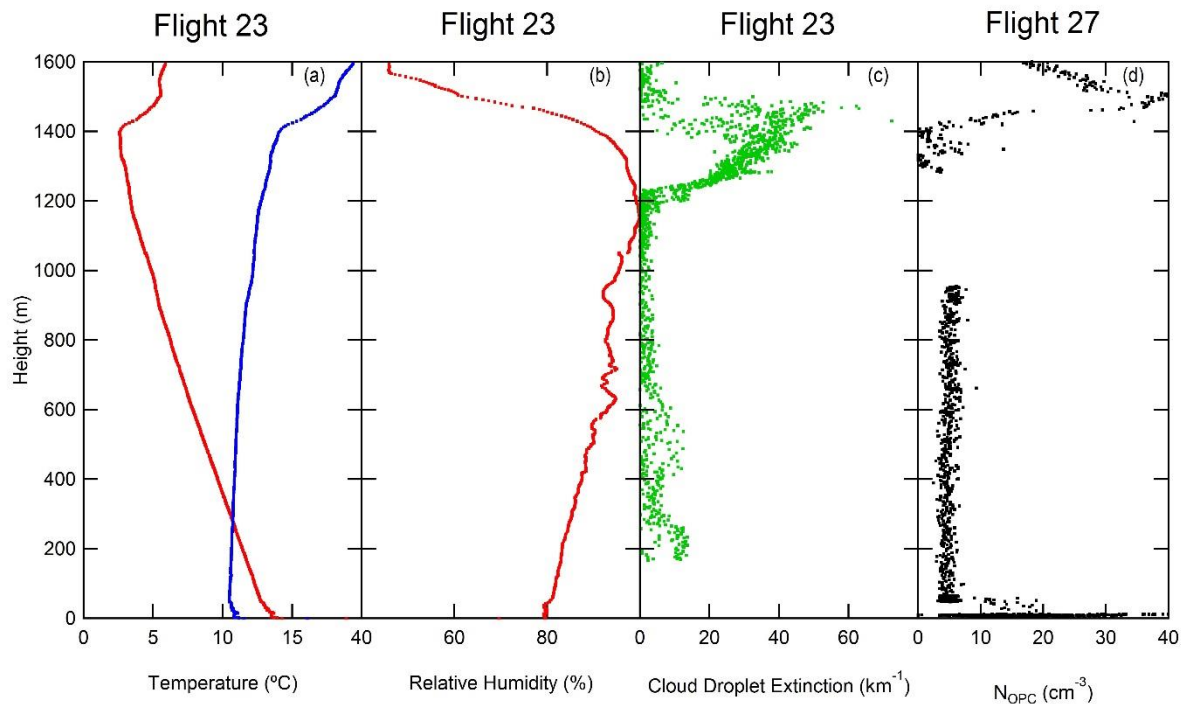
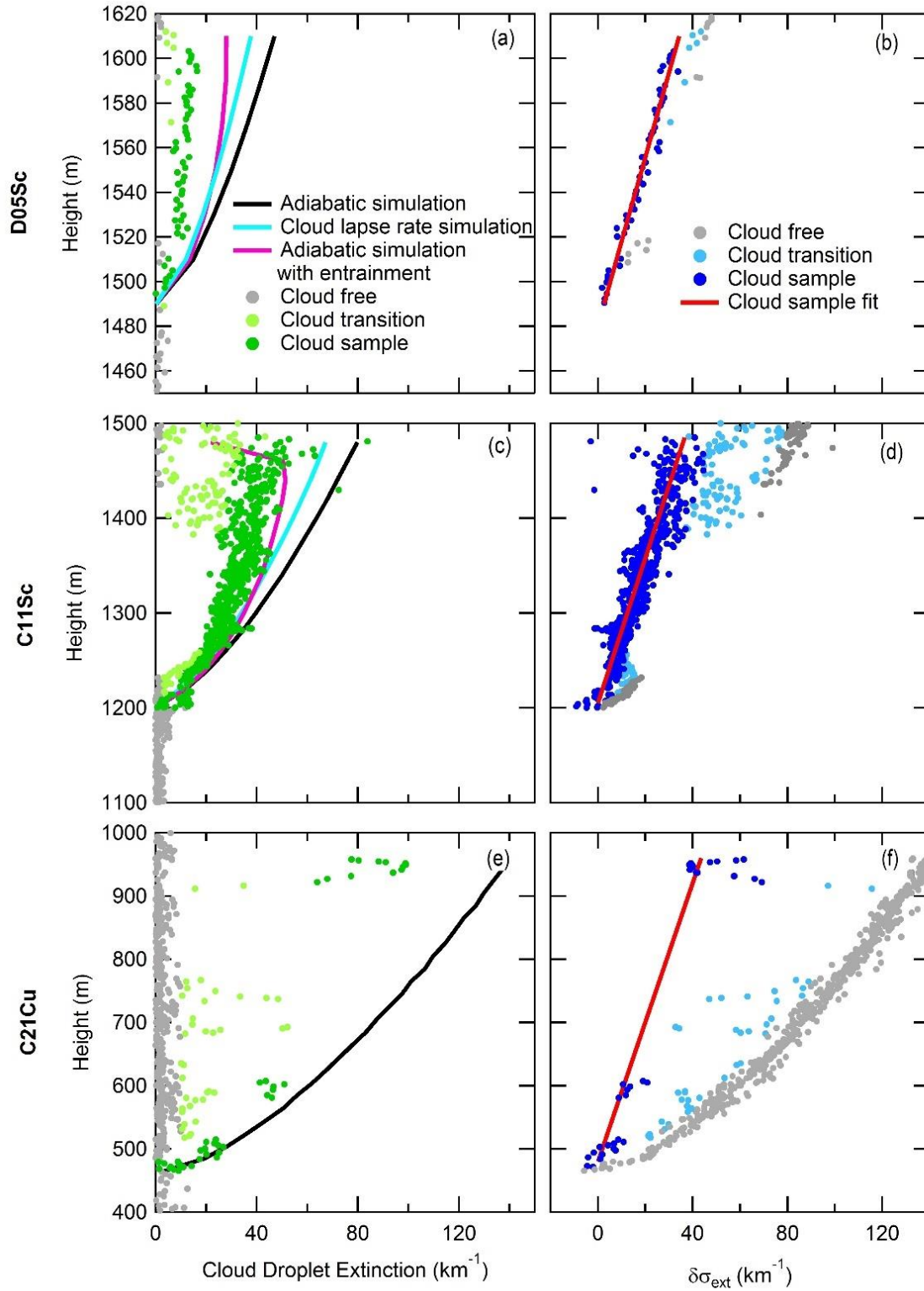


Figure 7. Vertical profiles of temperature, virtual potential temperature (θ_v), relative humidity, cloud droplet extinction and OPC total aerosol concentration. The figure consists of measurements collected from flights 23 and 27 on 11 August 2015 between 12:00 - 12:47 and 16:58 -17:33 respectively. The cloud level is between 1200 m to 1480 m in flight 23, and lowered to approximately 980 m to 1280 m in flight 27. OPC measurements that occurred in the cloud have been removed.



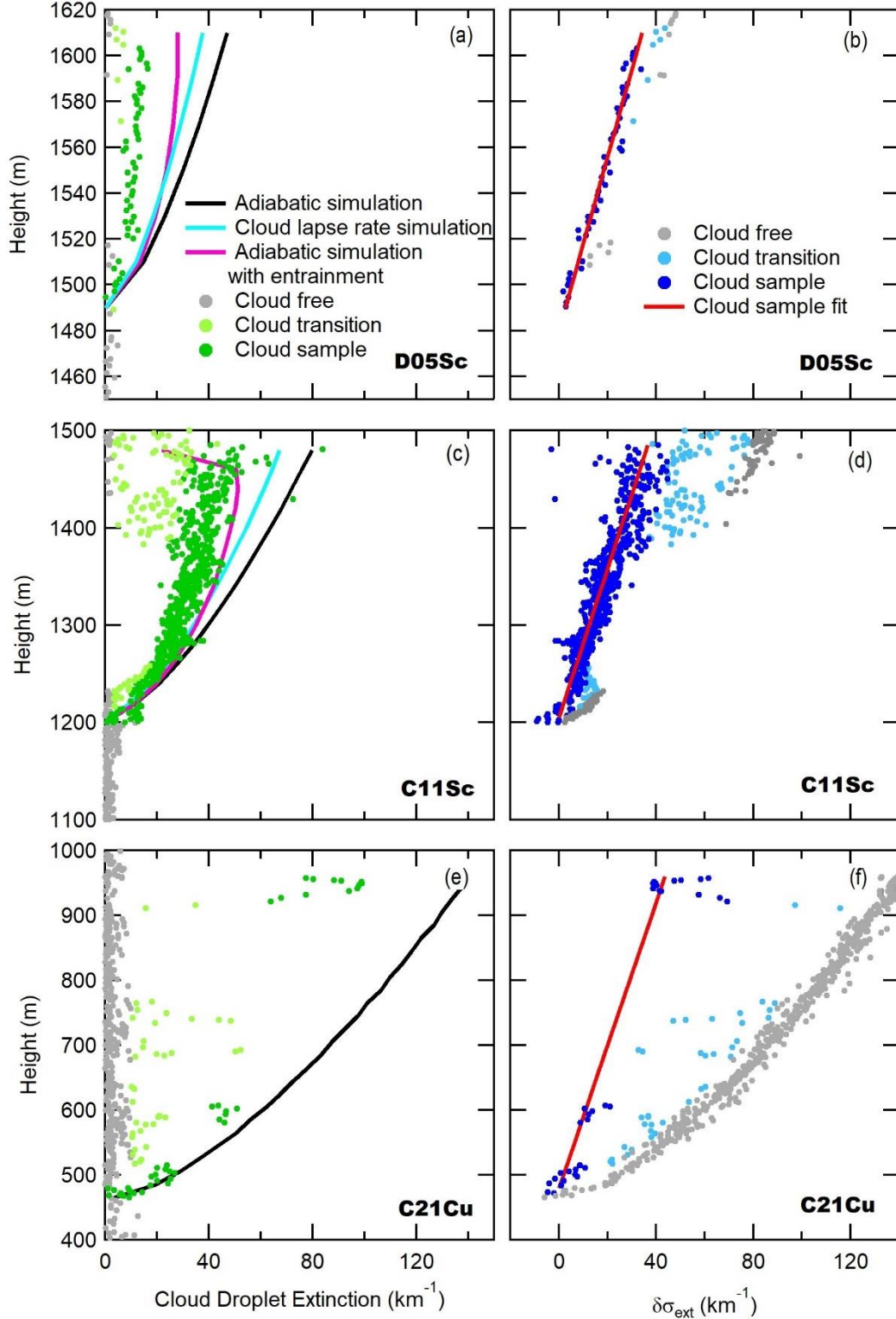


Figure 8. Vertical profiles of measured and simulated cloud extinction from flights D05Sc, C11Sc and C21Cu (left figures a, c, e; Table 2). *In-situ* measurements are classified into cloud, cloud-transition and cloud-free observations. The difference between UAV-observed (green measurements) and ACPM-simulated cloud extinction (black line) on left figures (a, c, e) are used to calculate ($\Delta\sigma_{\text{ext}}$) as a function of altitude in the right-hand side figures (b, d, f). The slope of the best fit through in-cloud measurements (red line) represents the increase in $\Delta\sigma_{\text{ext}}$ as a function of cloud thickness.

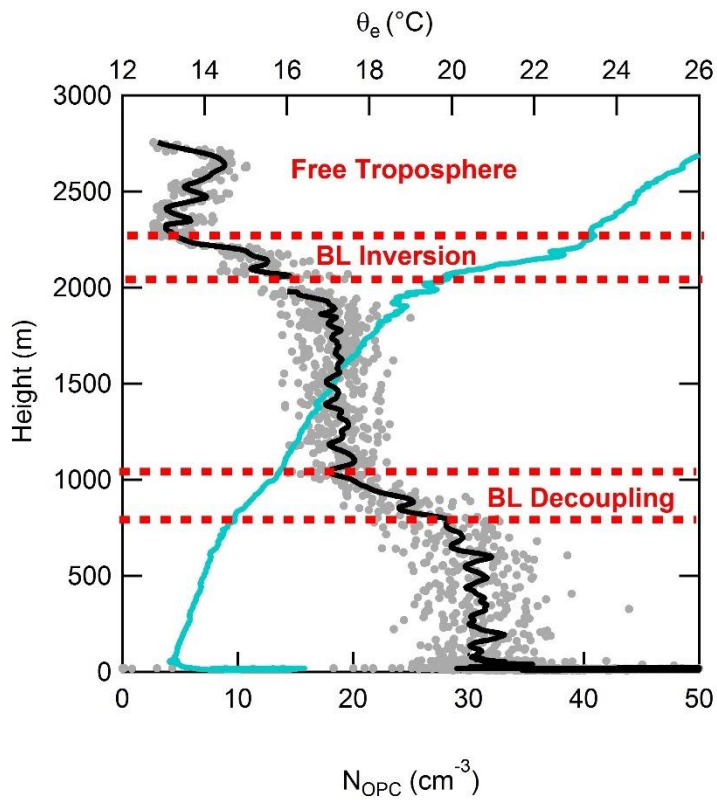
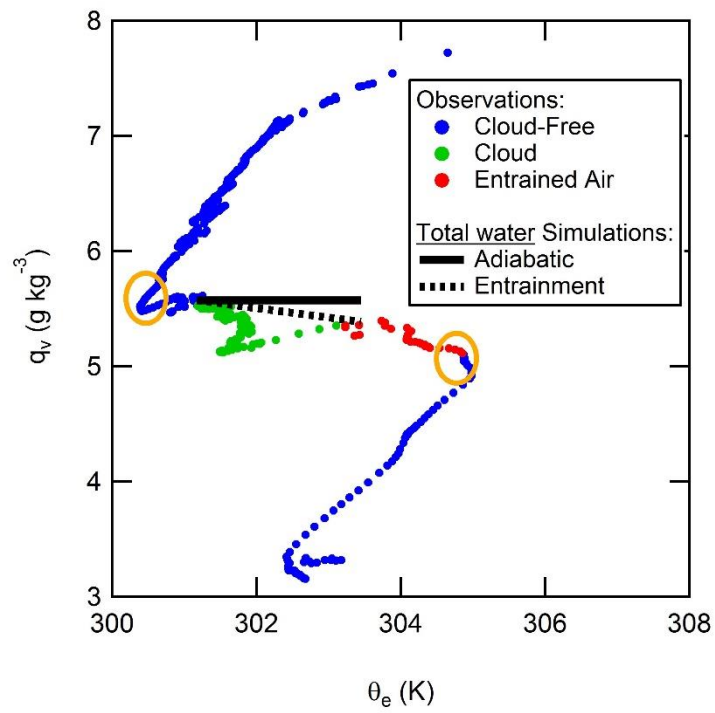
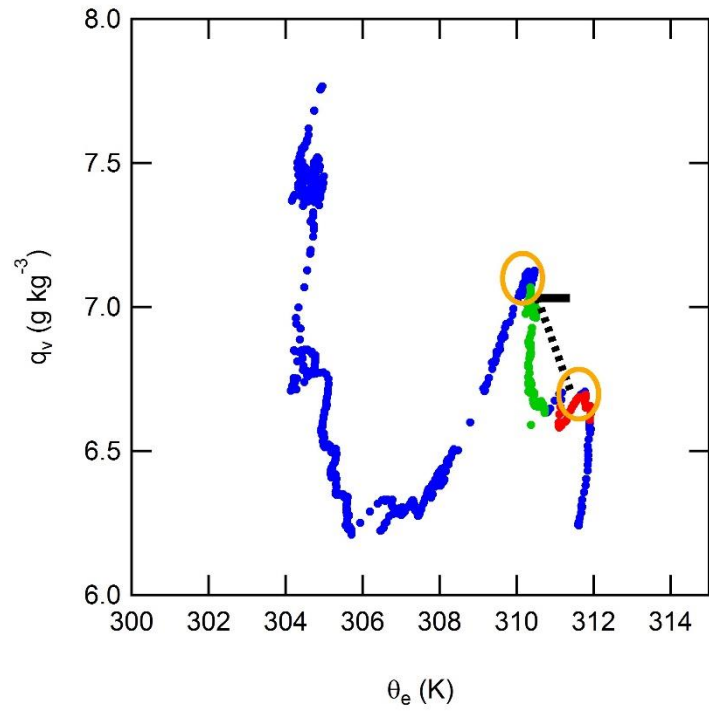


Figure 9. Flight 10 UAV vertical profile of OPC aerosol number concentrations ($D_p > 0.35 \mu\text{m}$) (grey) with a 20 second running mean (black) and equivalent potential temperature (θ_e , light blue) illustrate decoupling of the boundary layer. In-cloud OPC measurements (2000 m- 2050 m) have been removed.



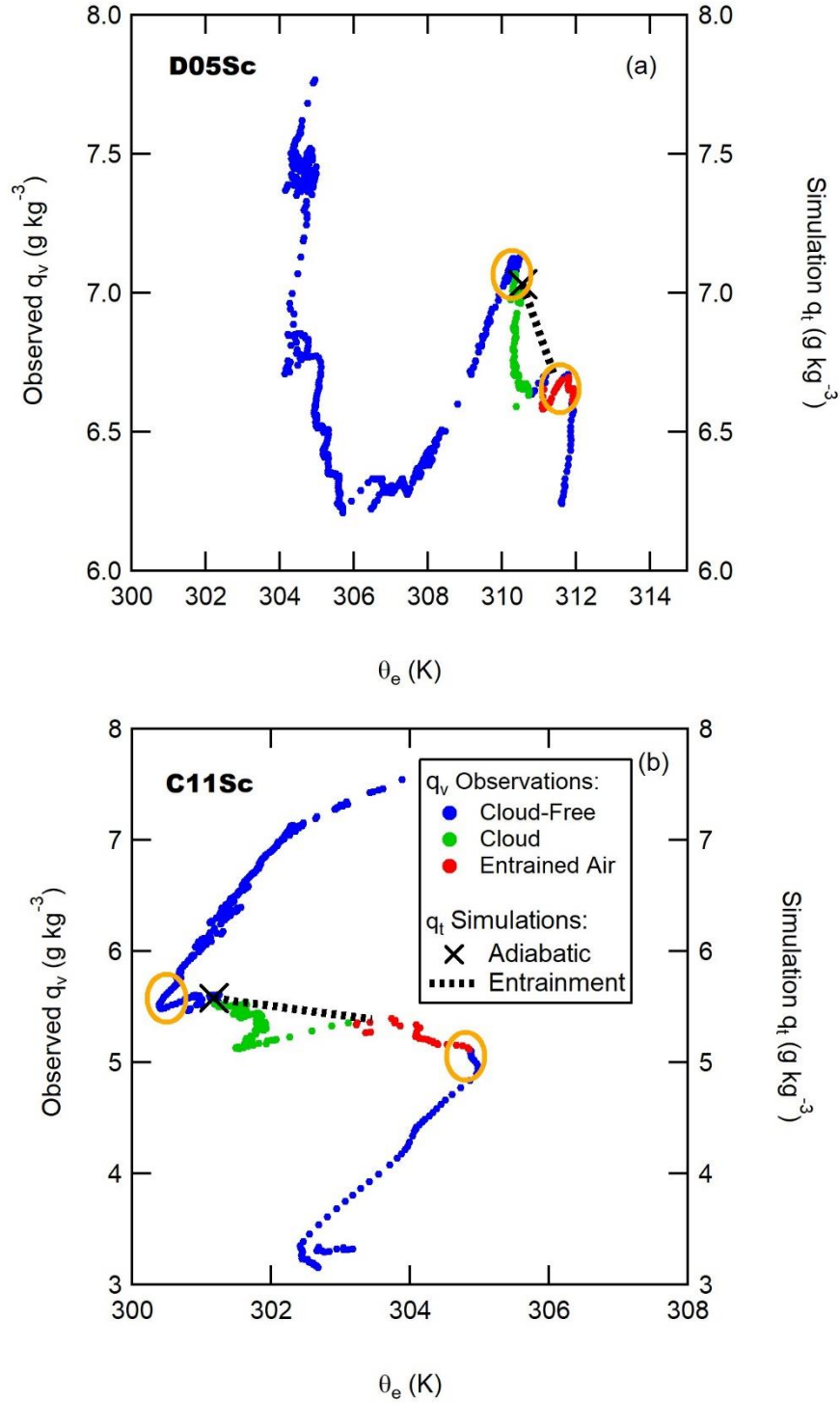
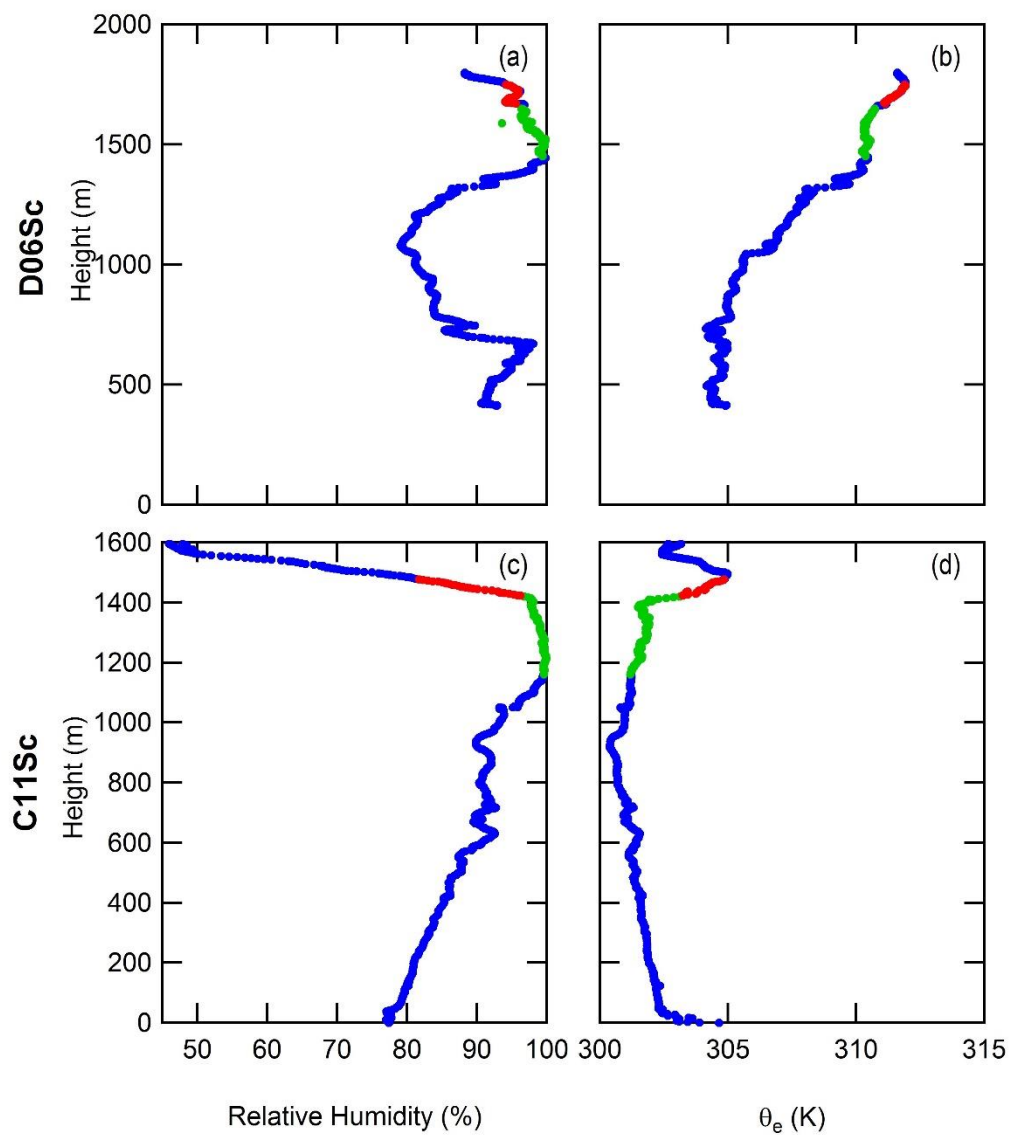


Figure 10. Conservative variables, water vapor content (q_v , conservative in subsaturated conditions and derived from RH measurements) and equivalent potential temperature (θ_e) identify mixing between cloud air and entrained air for flights D06Sc (top) and C11Sc (bottom). Measurements are defined as cloud-free (blue), in-cloud (green) or entrained air sources properties used in simulations (red). The orange circles highlight what is suggested to be the non-mixed sources of air.



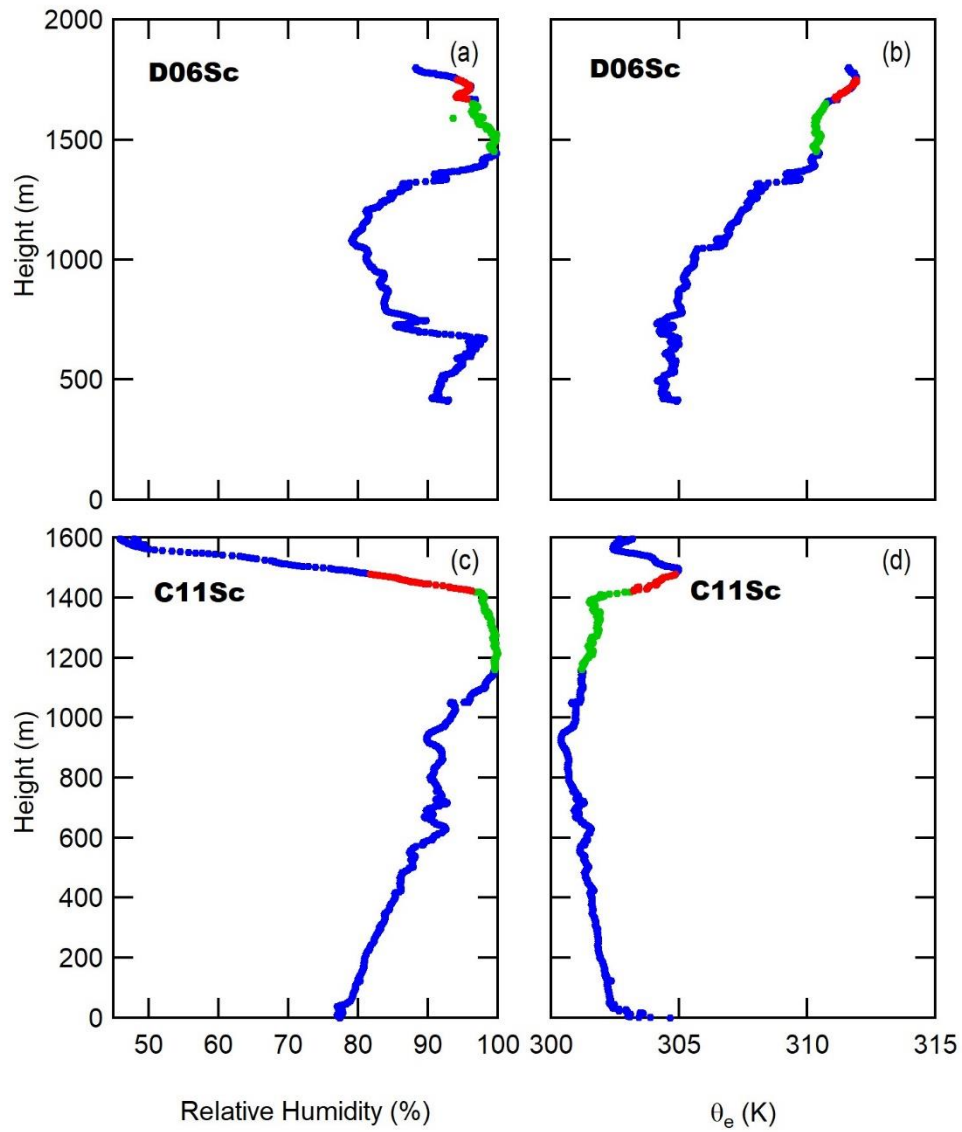


Figure 11. UAV vertical profiles of relative humidity (a, c) and θ_e (b, d) for flights D06Sc and C11Sc, used in Figure 910. Profiles are defined as cloud-free (blue), in-cloud (green) or entrained air sources (red).

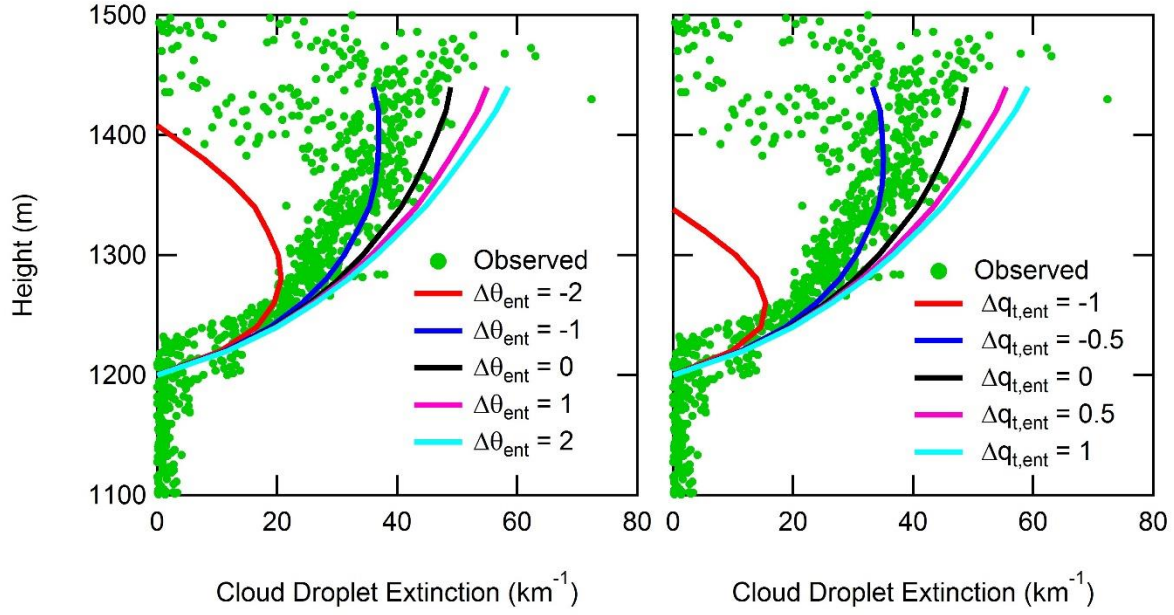


Figure 12. Sensitivity of simulated cloud extinction based on variability of entrained air potential temperature ($\Delta\theta_{\text{ent}}$, K) and entrained air total water mixing ratio ($\Delta q_{\text{t,ent}}$, g kg⁻¹) for the C11Sc case. Black lines are equivalent to the adiabatic simulation with entrainment from Figure 7c. The $\Delta\theta_{\text{ent}}$ and $\Delta q_{\text{t,ent}}$ terms define the change in the entrained θ and q_t values where no change ($\Delta\theta_{\text{ent}} = 0$ and $\Delta q_{\text{t,ent}} = 0$) is equivalent to the adiabatic simulation with entrainment from Figure 8c.