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

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Reviewer overview:

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.

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.

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 ($\delta\sigma_{ext}$) (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

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

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 bouyancy 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 ~ 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 g/m-3) for the case studies except for the C21Cu

case. The C21Cu case does have significantly high Liquid water content (>1.0 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 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 (Reff < 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 qv=qt 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 (qv) over the 1D theta-E mixing calculation done with Eq.4? 2) The dashed line is linear by design, on a qt axis. Since qt=qv at the two end points these would indeed be the end points of the dashed line but on this qv axis the line would be curved 3) It is not clear what the adiabatic line is supposed to represent. Why does theta-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 g kg^{-1} and q_v > 6.5 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 $q_v > 5.1$ g kg^{-1} and $q_v > 6.5$ 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 qv 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 qv and the right axis represents simulated qt.**

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.