

Response to the referee #1

Dear referee,

We appreciate your elaborate review of our manuscript. We have carefully considered your criticism and revised our manuscript accordingly, which has greatly benefited from substantial reorganization and a clearer presentation of the proposed theory and its validation. We will submit the revised paper after having received the report of reviewer # 2, in case the editor gives us the opportunity to do so. However, we would like to promptly react to your comments/suggestions. Fortunately, we show in the following comments that the core scientific conclusions of our paper remain robust.

The subject of this manuscript (water vapor supersaturation at cloud bases) was explored extensively for decades, mostly by cloud modeling (or parametrizations). There are almost no accurate in situ measurements of water vapor supersaturation at cloud bases with meteorological probes. The reason is that relative humidity (RH) is calculated from measurements of temperature and water vapor mixing ratio, and that the involved combined uncertainties result in errors of about 5-10% for the calculated RH, which is larger than the common range of values of supersaturation ($S_v = RH - 100\%$) measured by traditional cloud condensation nucleus (CCN) counters ($\sim 0.1 - 1\%$). Furthermore, additional uncertainties arise from the natural turbulence within clouds, droplet evaporation in the sensor element, and other factors. We are convinced that our study significantly contributes to the research of this important subject in cloud physics and aerosol-cloud interactions.

The main goal of the current manuscript is to present the physical basis of a new theory to quantify the energy budget at cloud base while associating it with cloud supersaturation (S_c), partitioned into liquid (Sl) and vapor (S_v) phases. We present a comparison between airborne in situ measurements of calculated S_v at cloud bases and explore the spectrum of droplet concentrations (N_d) as a function of S_v . The agreement between the calculated N_d (S_v) and measurements of $CCN(S_v)$ below cloud bases supports our proposed theory. The comparison was performed using airborne CCN counter measurements below cloud base, as in-cloud measurements are not possible. To our knowledge, this is the first time that the $CCN(S_v)$ measurements were replicated (within the uncertainty range and considering the different characteristics of the measurements) using in situ measurements at cloud bases. This comparison is discussed below, along with one of your comments. The authors had explored cloud-based supersaturation for Amazonian clouds in previous papers using modeling or parametrization techniques (see Braga et al., 2017 and 2021). We certainly believe that the theory and results presented in this study are novel.

Erratum: In the current published preprint version of the article, the authors identified an error in the schematic Figure 1 (see corrected version below). This issue was communicated to the editor on March 19 and approved by the editor on March 23. We regret if this error may have caused confusion for the referee during the review, and we sincerely apologize for the oversight.

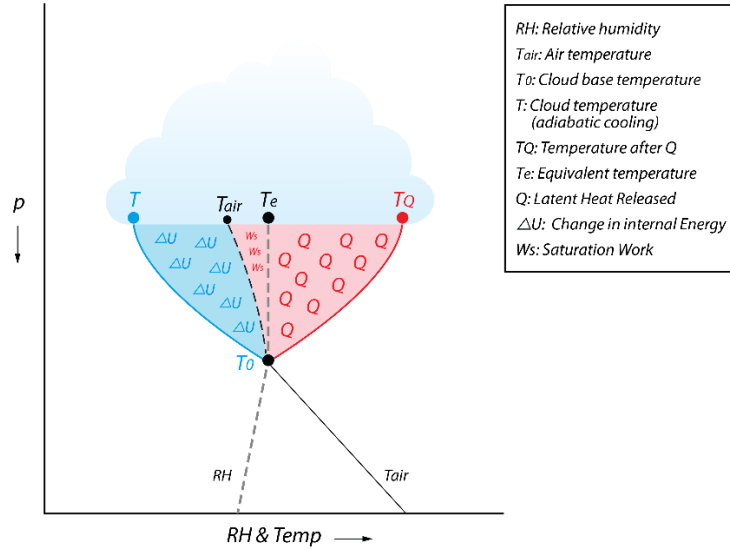


Figure 1. Energy balance of a cloud-adiabatic parcel at cloud base under the assumption of conserved isobaric enthalpy (T_e is constant). Below the cloud base, the moist air parcel ascends due to buoyancy. T_{air} decreases with altitude while RH increases up to the saturation level of water vapor (RH = 100%) at cloud base, where the temperature and pressure are T_0 and p_0 , respectively. Above this level, the ascending saturated parcel cools adiabatically as pressure (p) decreases slightly to a given temperature (T). Note that for the isobaric assumption $p = p_0$. Under the assumption of conserved enthalpy, the latent heat released by condensation (Q) is proportional to the cooling rate (ΔT) from T_0 to T (i.e., $T_Q = T_0 + \Delta T$ and $T_e = T + \Delta T$). From the first law of thermodynamics, during the condensational growth of CCN particles and droplets, Q is balanced with the decrease in internal energy (ΔU) and saturation work of water vapor (W_s), resulting in an air temperature (T_{air}) that is warmer than T and cooler than T_0 . Similarly, T is slightly warmer due to the W_s term (not shown in the figure for clarity). The energy expended on vapor expansion (W_s) during this process results in an excess of water vapor (S_v) relative to the case in which the water vapor is fully condensed.

Title: Energetically Stringent Quantification of Water Vapor Supersaturation at Cloud Base

Authors: Braga et al.

Recommendation: Rejection

Overall comments:

This manuscript proposes a new thermodynamic framework to infer cloud-base water vapor supersaturation (S_v) by treating the ascent of a saturated air parcel as a reversible cloud-adiabatic process with conserved isobaric enthalpy, and then partitioning the latent heating term (Q) into internal-energy (ΔU) and saturation-work (W_s) contributions. The authors then compare inferred $N_d(S_v)$ spectra at Amazonian cloud bases with below-cloud $CCN(S_v)$ spectra and report agreement within stated uncertainty bounds.

The manuscript is novel and potentially interesting, especially in its attempt to connect cloud-base supersaturation to an explicit energetic closure using aircraft observations. The observational dataset is also valuable. However, I am afraid that the thermodynamic interpretation is not yet sufficiently demonstrated or justified for publication. The conceptual novelty is promising, but the derivation, physical interpretation, sensitivity analysis, and validation strategy all need substantial strengthening before the main claims can be accepted. The manuscript also contains a potential overstatement in its conclusions about standard adiabatic models "overestimating" liquid water content, latent heating, buoyancy, and updraft speeds. Furthermore, the structure of the manuscript also needs substantial revision. Finally, this manuscript appears to be poorly written with many instances of unnecessary repetition.

Therefore, I do not think the manuscript is publishable in its current form. I hope my comments will be received as constructive and helpful to the authors. Considering the time and effort the authors might need to address the following concerns and comments, I recommend rejection of the manuscript.

Major comments:

1. Equation derivation and the decomposition of Q and Sc .

Even though the authors spent a lot of effort showing the equation derivations, I am afraid that it is still very difficult and not logical enough for readers to follow. My main concern is that the manuscript appears to jump too quickly from an energy-budget $Q = \Delta U + W_s$ to a new definition or decomposition of supersaturation $Sc = S_l + S_v$, where S_l and S_v are assigned in proportion to $\frac{\Delta U}{Q}$ and $\frac{W_s}{Q}$. The manuscript states this explicitly in Section 2.3 with equations 22-24. But supersaturation is a thermodynamic disequilibrium state variable defined by vapor pressure or saturation ratio, while ΔU and W_s are path-dependent energetic terms. I think the paper needs a much clearer and firmer derivation showing why this partition is plausible and unique. At present, it reads more like a proposed energetic analogy. The paper needs to prove that this decomposition really

follows the first law of thermodynamics and the Clausius-Clapeyron relation, and is not simply a convenient remapping. Therefore, the authors should either provide a firmer and cleaner derivation or explicitly reframe the method as a physically motivated diagnostic framework.

A: We thank the reviewer for this important comment. We agree that the relationship between the energy budget and the supersaturation partition can be presented more clearly and in a more structured manner. Below, you find a significantly improved introduction of the proposed method and a firm clarification of the relationship between the energy budget and the Clausius-Clapeyron relation.

In the current manuscript, we describe the ascent of a saturated parcel assuming the following thermodynamic conditions:

- Adiabatic parcel: no heat is exchanged with the surroundings ($\delta Q_{\text{ext}} = 0$);
- The energy budget is calculated for isobaric conditions: the small variability in pressure at the cloud base during the ascent ($\sim 0.1\text{-}0.3\%$) is negligible.
- For isobaric conditions, the total heat (latent heat) in the parcel results from phase change (vapor to liquid). During condensation, the latent heat released is the enthalpy difference ($Q = dH$). See section 1 below for description.
- The energy (or latent heat) budget of the parcel is calculated using the first law of thermodynamics; the change in enthalpy for isobaric conditions is given by $Q = dH = dU + Ws$. Since the air parcel is isobaric and saturated, the energy budget depends only on pressure and temperature, and thus dU and Ws are process-dependent. Sections 2 and 3 describe the proposed energy budget and the invertibility of the energetic parameters, respectively. The partition of the cloud supersaturation is described in section 4.

Important information: The “reference-state of saturation ratio”- RHc described in the submitted manuscript was long and confusing. We decided to calculate RHc based on the classical Bolton equation for $e_s(T)$, as shown below. This is more accurate and leads to minor relative differences in the absolute values of Sc, Sl, and Sv shown in the submitted version. Furthermore, it clarifies the relationship between the energy budget proposed and the Clausius-Clapeyron relation.

Below are some clarifications of the method. Some of the symbols are not available in the submitted manuscript and will be added further. Furthermore, the references mentioned here are available in the submitted version.

1) Calculation of the latent heat and cloud supersaturation

The energy (latent heat) of the saturated parcel is limited by the enthalpy difference of the phase change from vapor to liquid, $Q = dH$, and can be written in finite forms as follows:

$$Q = \Delta H \quad \therefore \quad L_v \cdot q_l = -c_{p,\text{cloud}} \cdot \Delta T = -c_{p,\text{cloud}} \cdot (T_0 - T) \quad (1)$$

During the phase change, the internal energy decreases ($T_0 > T$) as q_l (liquid water mixing ratio) and the latent heat of condensation (Q) increase. We assume the warmer temperature (T_0) in the $c_{p,\text{cloud}}$ (i.e., $c_{p,\text{cloud}} = c_{pd} + c_{pv} r_s(T_0) + c_{pl} q_l$) and L_v calculations, as it represents the initial condition of the saturated parcel. For small temperature differences ($< \sim 1$ K), the specific heat capacity of the cloudy air ($c_{p,\text{cloud}}$) varies only weakly with r_s (saturated mixing ratio) and q_l .

The phase change process leads to an increase in the air temperature, in this case the equivalent temperature (T_e), and is constant during the ascent (shown in the figure above), i.e.:

$$T_e = T + \Delta T_Q = T_0 = T_{air} \quad (2)$$

Where,

$$\Delta T_Q = \frac{L_v \cdot q_l}{c_{p,\text{cloud}}} \quad (3)$$

1.1 RHc (reference-state saturation ratio)

The formulation of relative humidity - RH describes the amount of water vapor in the air relative to the amount of water vapor in the air when it is saturated for a given temperature, i.e., the saturation ratio, and is defined as follows:

$$RH = \frac{e(T)}{e_s(T)} \quad (4)$$

Where $e(T)$ is the water vapor pressure (Pa), $e_s(T)$ is the saturation water vapor pressure (Pa), and T is the air temperature (K or °C) (Bolton, 1980; Rogers and YAU, 1989).

The values of $e_s(T)$ can be calculated for a wide range of temperatures ($-30 \text{ °C} \leq T_c \leq 35 \text{ °C}$) with minor uncertainties ($\sim 0.1\%$) as follows (Bolton, 1980):

$$e_s = 611.2 \cdot \exp\left(\frac{17.67 \cdot T_c}{T_c + 243.5}\right) \quad (5)$$

For a saturated parcel under isobaric conditions, T_c is the dew point temperature (T_d) in °C. The reference-state saturation ratio of a saturated parcel or cloud (RH_c) undergoing expansion (cooling) is calculated as follows:

$$RH_c = \frac{e_s(T)}{e_s(T_0)} \quad (6)$$

The cloud supersaturation (Sc) is formulated as follows:

$$S_c = RH_s - RH_c = 100 - RH_c \quad [\%] \quad (7)$$

where $RH_s = 100\%$ is the saturation ratio at the reference state (T_0), and Sc expresses the thermodynamic excess of water vapor relative to equilibrium with respect to liquid water at the reference state. Since the cooling process is related to the increase of q_l (shown in Eq. 1), the increase in Sc is associated with vapor-liquid conversion and Q .

2) Calculation of energy budget using the first law of thermodynamics

According to the first law of thermodynamics, the change in internal energy of a closed system is determined by the balance between heat exchange and pressure-volume work. For an isobaric process involving only pressure-volume work, the change in enthalpy (dH) can be interpreted as the sum of the change in internal energy and the expansion work performed by the system. Under adiabatic conditions, the dH equals the heat exchanged at constant pressure by a saturated parcel. Thus, for a saturated parcel undergoing condensational growth from the reference state, the latent heat released ($Q=dH$) is the source of heat in the thermodynamic system, and the energy budget can be calculated following the first law as follows:

$$Q = \Delta U + W_s \quad (8)$$

where Q represents the latent heat released by condensation, ΔU is the change in internal energy of the parcel associated with vapor-liquid conversion, and W_s is the work associated with the expansion of water vapor.

Analogous to eq. 1 above, the energy budget from eq. 8 can be calculated as follows:

$$L_v \cdot q_l = -c_{p,cloud} \cdot \Delta T_{ws} + r_{s0} \cdot R_v \cdot T_0 \cdot \ln\left(\frac{\alpha_{vs}}{\alpha_{v0s}}\right) \quad (9)$$

Or

$$L_v \cdot q_l - r_{s0} \cdot R_v \cdot T_0 \cdot \ln\left(\frac{\alpha_{vs}}{\alpha_{v0s}}\right) = -c_{p,cloud} \cdot (T_0 - T_{ws}) \quad (10)$$

Note that the W_s term assumes the parameters of the initial conditions of the saturated parcel (T_0) and depends solely on temperature (T_0 and T) for isobaric conditions. From this budget, T_{ws} , the saturated temperature, is slightly warmer than T calculated in eq. 1 due to the W_s term. Similarly, the resulting air temperature (T_{air} , shown in Figure 1 above) will be slightly cooler than T_e (see eq.3) due to the W_s term.

$$T_{air} = T + \Delta T_Q - \left(\frac{W_s}{c_{p,cloud}} \right) \quad (11)$$

The differences between $T_{ws} - T$ and $T_0 - T_{air}$ are the same.

3) Invertibility of the energy budget of the saturated parcel

The invertibility of the energy budget for isobaric conditions relies on the fact that the total latent energy ($Q=dH$) can be expressed equivalently in terms of either LWC or the temperature difference ΔT between two saturated states. In Section 1, we show that the relationship between LWC and ΔT is one-to-one since the latent energy balance $L_v q_l = c_{p,cloud} \Delta T$, assuming $c_{p,cloud} = c_{pd} + c_{pv} r_s(T_0) + c_{pl} q_l$, can be uniquely solved in either direction. In this framework, this dependence is treated explicitly through an iterative solution, ensuring consistency between condensate and heat capacity.

In section 2, we show that the total energy is partitioned according to $Q = \Delta U + W_s$, where the internal-energy component $\Delta U = -c_{p,cloud}(T_0 - T_{ws}) = L_v q_{lws}$ defines a reduced cooling relative to the case without accounting for the W_s term. Despite this partitioning, the same functional dependence between q_{lws} and ΔT_{ws} is retained, ensuring that the mapping remains one-to-one. Since the parameters at W_s , $c_{p,cloud}$ and L_v are calculated for the initial conditions of the parcel (T_0), the invertibility of the heat budget is conserved.

4) Calculation of liquid and water vapor supersaturation

Figure 1 above shows the effect of the partition of Q on the energy balance. The energy partition of S_c in the liquid phase (S_l) and vapor phase (S_v) is calculated based on the energy contributions of ΔU and W_s in the total energy of the saturated parcel (Q), respectively. S_l is considered for the liquid phase due to the vapor-liquid conversion, while S_v is associated with the water vapor expansion work.

The signs in the energy balance equations shown above describe the direction of energy transfer. However, S_l and S_v are not directional variables. They represent the partition of S_c considering the energy budget, which is defined from the absolute magnitudes of the energetic contributions, and thus, for a given latent heat released (Q), due to condensation S_l and S_v are calculated as follows:

$$S_v = S_c \cdot \frac{|W_s|}{Q} \quad [\%] \quad (12), \quad S_v = S_c \cdot \frac{|\Delta U|}{Q} \quad (13), \quad \text{and} \quad S_c = S_l + S_v \quad [\%] \quad (14)$$

The numerical differences while applying equations 1 and 8 to calculate Q are negligible ($< \sim$

0.01% on average).

5) Closure Analysis from in situ measurements

As described in the manuscript, the aircraft measured T , p , and LWC (or q_l) for cloud passes at growing convective cumuli (typically cumulus humilis and mediocris) with $N_d > 20 \text{ cm}^{-3}$ and positive vertical velocities. From these measurements, we calculate the energy budget and the Sc , SI , and S_v of the parcel for the conserved heat (Q). In these estimates, we assume the cloud temperature equals the temperature of the saturated parcel (T in the schematic figure). In the first step, the values of T_0 are calculated iteratively based on the LWC from CDP and the meteorological measurements from BAHAMAS. In the second step, the energy budget is calculated, accounting for W_s , resulting in the values of SI , S_v , and T_{air} .

We believe these clarifications address the reviewer's main concern and significantly improve the description of the derivations shown in the proposed manuscript. Below, you may find new figures that will be added to the main manuscript for clarity. Codes used in the calculations will be provided during the review process.

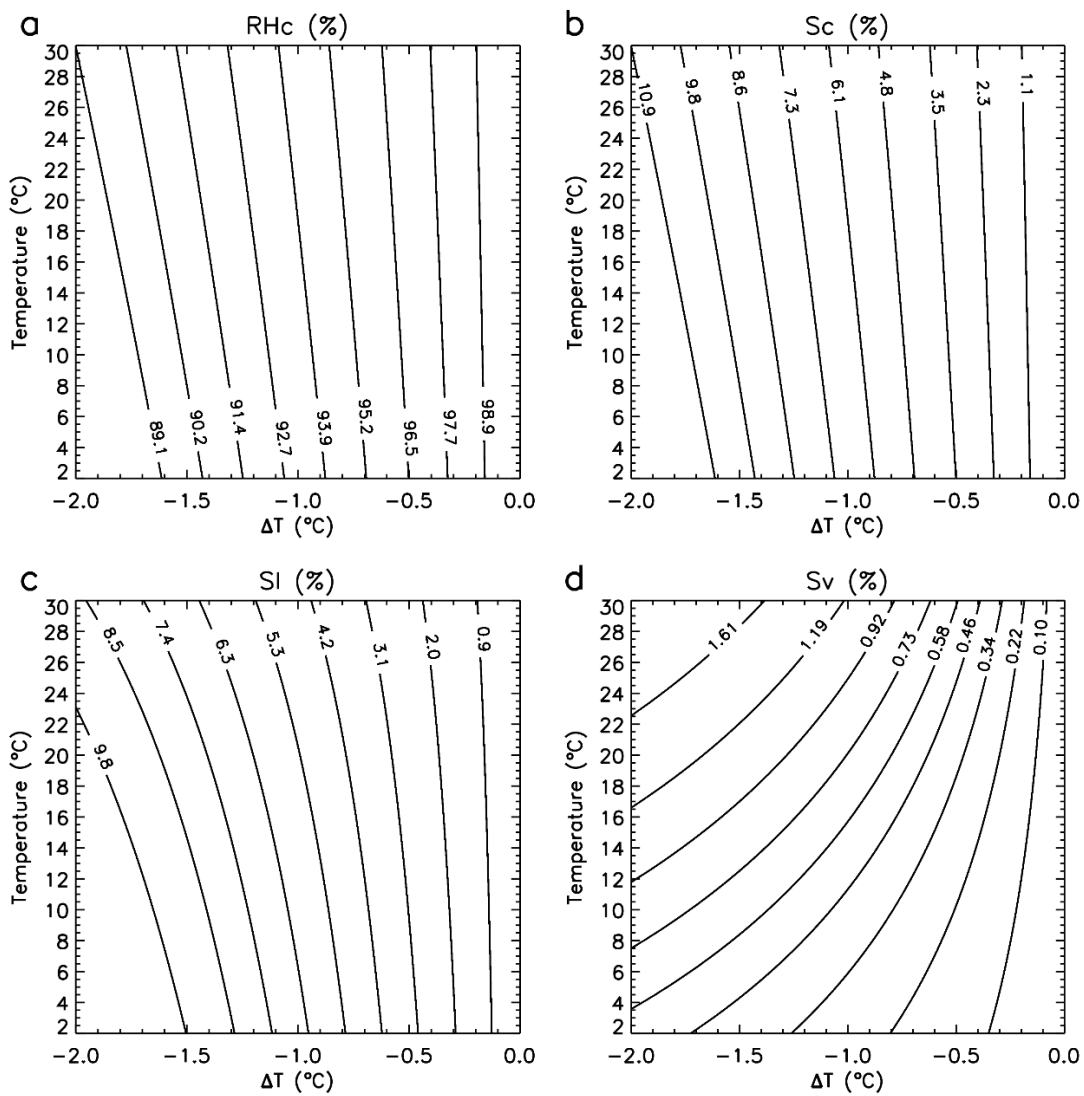


Figure 2. (a) Contours of reference-state saturation ratio (RH_c) as a function of initial temperature T_0 [y-axis] and adiabatic cooling rate (ΔT) [x-axis] for saturated parcels at 950 hPa. (b), (c) and (d) Similar to cloud supersaturation (S_c), liquid-phase supersaturation (S_l) and for water vapor supersaturation (S_v), respectively.

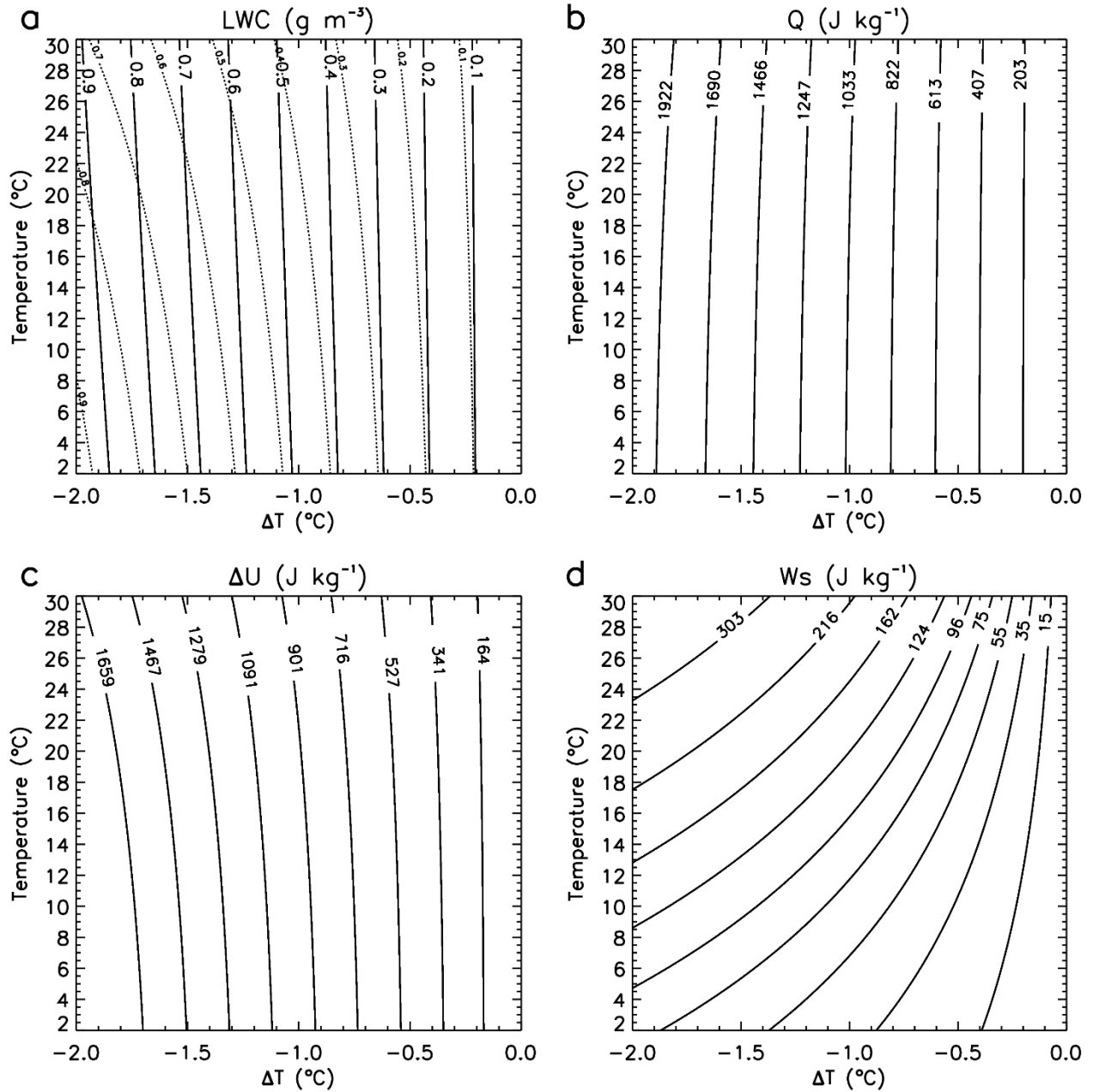


Figure 3. (a) Contours of Liquid Water Content (LWC) values and energy balance terms of cloud adiabatic parcels as a function of initial temperature T_0 (y-axis) and adiabatic cooling rate (ΔT) (x-axis) at 950 hPa. Dotted lines indicate the LWC resulting from the energy budget (W_s term leads to LWC decrease for the same ΔT). Similar for (b) Latent heat of condensation (Q), (c) Magnitude of internal-energy change ($|\Delta U|$), and (d) saturation work (W_s).

Flight AC14: Central Amazon

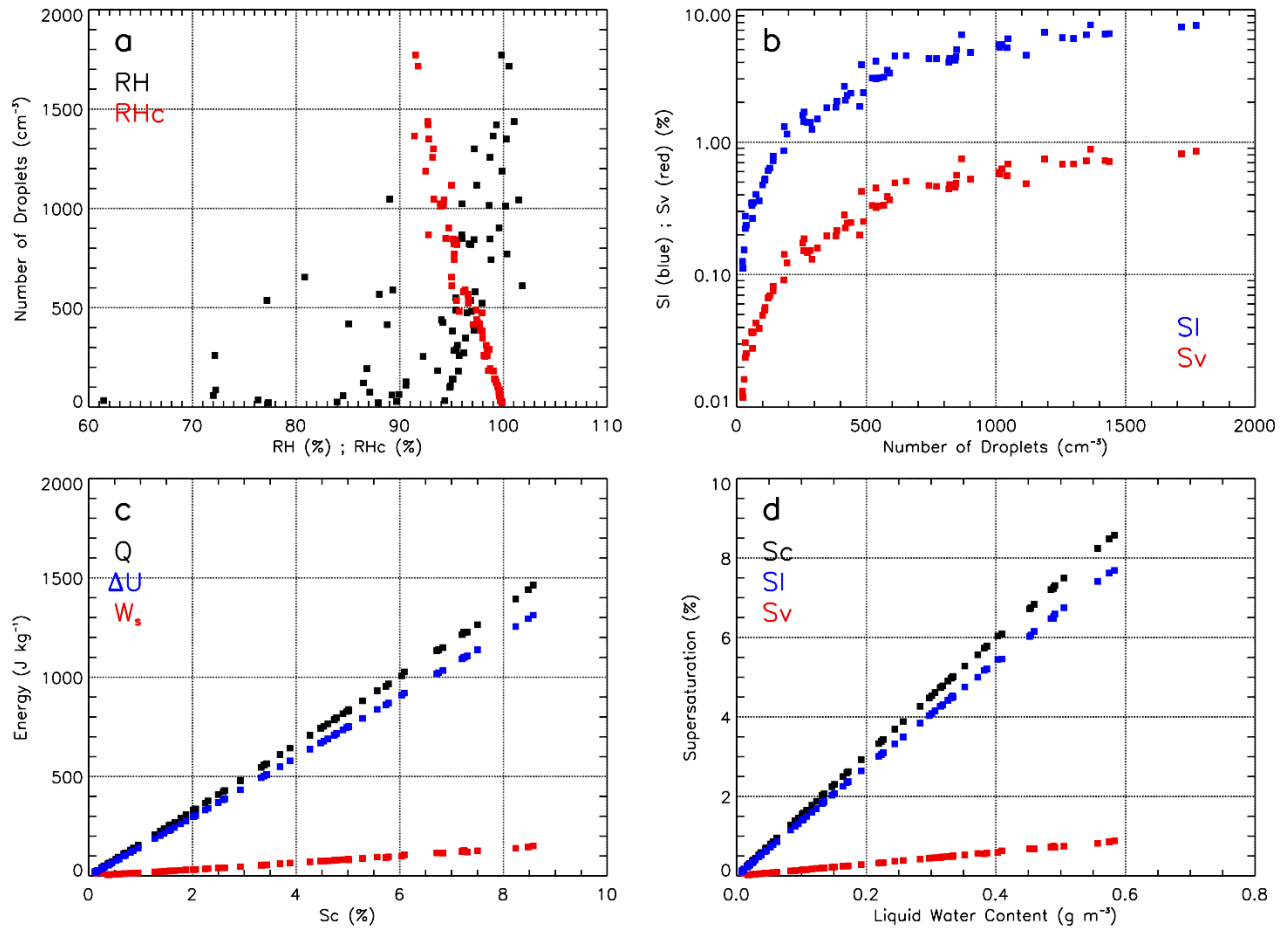


Figure 4 (former figure 2 updated). Cloud microphysical and thermodynamic properties measured at cloud bases of growing convective cumuli during Flight AC14. (a) RH versus Nd (black) and RHc versus Nd (red). (b) Nd versus SI (blue) and Nd versus Sv (red). (c) Sc versus energy magnitude of Q, ΔU , and W_s . (d) LWC versus Sc, SI, and Sv.

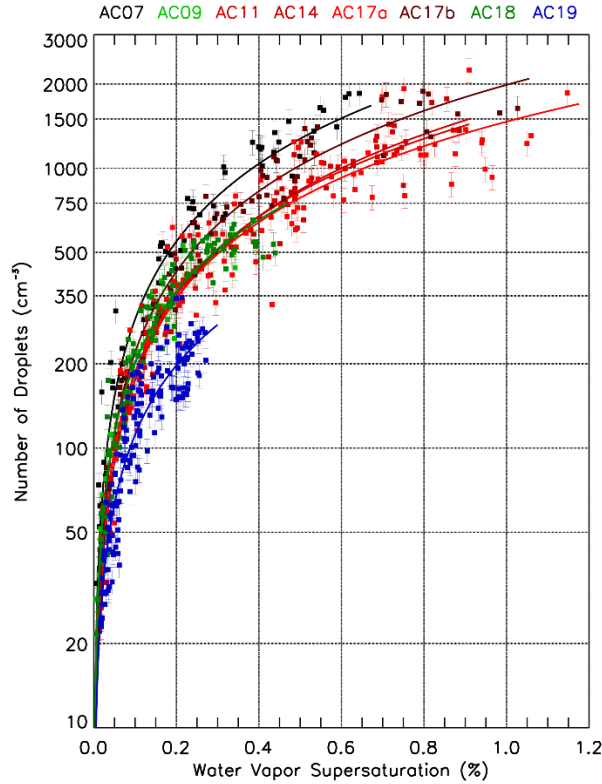


Figure 5 (former figure 4 updated). S_v vs. N_d for multiple flight segments (AC07, AC09, AC11, AC14, AC17a, AC17b, AC18, and AC19). Colored markers represent in-situ measurements, while solid curves correspond to power-law fits of the form $N_d = C \cdot S_v^k$. The fitted parameters and associated uncertainties are summarized in Table X.

Table X. Power-law fit parameters describing the relationship between cloud droplet number concentration (N_d) and supersaturation (S_v) for each flight segment, expressed as $N_d = C S_v^k$. The coefficients C (scaling factor) and k (sensitivity exponent) are reported as Monte Carlo means ± 1 standard deviation, obtained from the uncertainty propagation of S_v and N_d . The coefficient of determination (R^2) are calculated from the nominal (best) fit in log–log space. The sample size (N) indicates the number of 1 Hz cloud passes used in each regression.

Flight	C	k	R ²	N
AC07	2417.38 \pm 80.01	0.923 \pm 0.014	0.958	77
AC09	1352.97 \pm 83.48	0.828 \pm 0.022	0.952	41
AC11	1567.29 \pm 66.16	0.925 \pm 0.027	0.982	33
AC14	1644.66 \pm 51.57	0.974 \pm 0.015	0.981	73
AC17a	1467.06 \pm 33.21	0.904 \pm 0.012	0.947	121
AC17b	1985.79 \pm 63.35	0.955 \pm 0.018	0.975	64
AC18	1420.38 \pm 50.03	0.847 \pm 0.015	0.94	99
AC19	743.72 \pm 27.24	0.824 \pm 0.015	0.874	163

2. Validation

In Section 3.2, the authors compare inferred $N_d(S_v)$ at cloud base with $CCN(S_v)$ spectra measured below cloud base and concludes that they agree within uncertainty. First, how was the 10% uncertainty estimated/calculated/determined? Second, the authors note that the CCN measurements were taken roughly 700 m below cloud base in some flights and about 1400 m below cloud base in another, that CCN counters count droplets above $\sim 1 \mu\text{m}$ whereas N_d was measured for droplets larger than $1.5 \mu\text{m}$, and that spatial/temporal variability in hygroscopicity and turbulence can introduce additional uncertainty. I am afraid that those caveats really weaken the independence of the closure test. Agreement "within uncertainty" is encouraging but it is not yet a decisive validation of the thermodynamic interpretation. This appears to be more like an initial consistency check rather than a validation of the new supersaturation theory.

A: The reviewer considers that agreement between the theoretically derived estimates and those measured using instrumentation is a weak validation of the theory because of the inherent uncertainty in these measurements (explained further below), which were carefully considered and accounted for by the authors. While we agree with the reviewer that lower inherent uncertainties would be preferable, we argue that any impact due to the additional caveats mentioned are minor and that uncertainties of the range derived are common to all in-situ cloud microphysical measurements (whether acknowledged or not). We therefore propose that the validation is best practice that can be achieved with current airborne instrumentation, and that it provides a good initial validation of the theory proposed which might be further tested using a diverse range of in situ datasets by other researchers. We provide further updated information on the uncertainty analysis below.

The uncertainty shown in the figure relates to the measurement uncertainty of CCN and droplet concentrations (N_d). See item 3 for further information on the uncertainties of each set of measurements.

The comparison between $N_d(S_v)$ and $CCN(S_v)$ aims not only to check the measurement range but also to verify the robustness of the physical assumptions. Since S_v is a property of the gas (water vapor), the S_v from a CCN counter (CCN_c) is calculated based on the Kohler theory for particles with a known physicochemical composition, typically ammonium sulfate.

The $N_d(S_v)$ spectrum is representative of the $CCN(S_v)$ population below cloud bases that, after buoyant ascent, were activated into cloud droplets. Note that a smaller sample of N_d in comparison to CCN concentrations was measured in the time range in which the aircraft was at cloud base level (see Tables 2 and 3).

During the research flights, there is natural variability in CCN concentrations for a given measured S_v below cloud base. This variability is greater when flying within air masses affected by pollution plumes from biomass-burning and urban emissions. In the figure below (updated figure 5 in the former manuscript), the variability of the CCN

population is observed in the measurements with fixed S_v (CCNb), in this case, $S_v \sim 0.55\%$ shown in blue. The figure shows measurements of N_d in the same range of CCN concentrations for the same S_v . The measurements of CCNa (shown in red) show cycling S_v between 0.2% and 0.55% every 100 seconds. Since some of the measurements below cloud base were affected by pollution plumes, the typical profile of $CCN(S_v)$ (i.e., CCN concentrations increasing with S_v) is not observed for all cases. This is clearly shown in the figure (e.g., in Flight AC11) where large concentrations were measured for relatively smaller S_v . In the flight segment AC17b, a small variability of CCN concentrations is observed in the CCBn sampling, which results in CCNa(S_v) measurements more consistent with those observed in homogenous air masses.

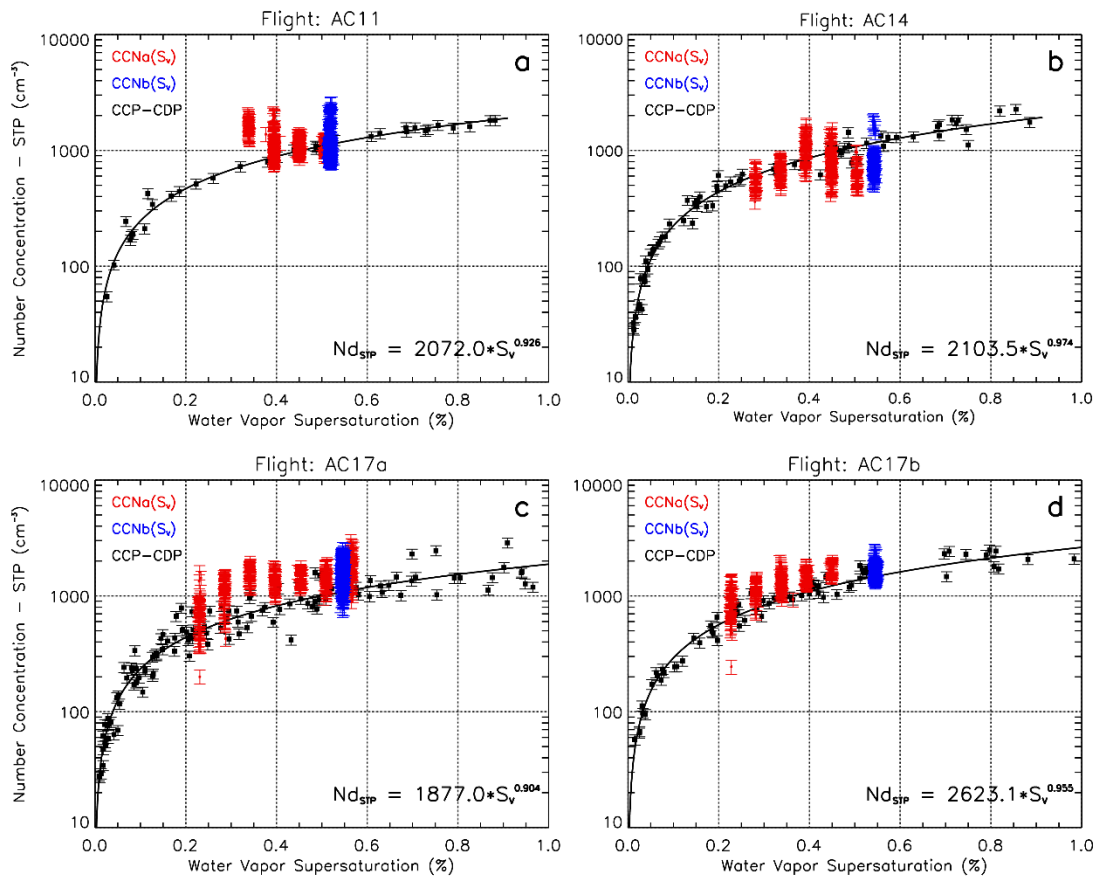


Figure 6 (former figure 5 updated). (a) S_v vs. N_d measured at cloud bases (black dots), and S_v vs. CCN concentrations measured below cloud bases (CCNa(S_v) in red and CCBn(S_v) in blue) during Flight AC11. N_d and CCN concentrations are calculated at standard temperature (0°C) and pressure (1013.25 hPa) (STP). Solid curves correspond to power-law fits of the form $N_d = C \cdot S_v^k$. (b), (c), and (d), similar data from flight legs AC14, AC17a, and AC17b, respectively. Error bars indicate the estimated uncertainties of N_d (10%) and CCN(S_v) concentrations (14%). Uncertainties of S_v from CCN counters (10%) and theoretically derived (12%) are not

shown.

The figure below shows the Nd (Sv) spectra and the CCN(Sv) measurements for time intervals in which both CCNa and CCNb had measurements, and the sampling was larger than 30 s. The CCN(Sv) measurements are shown with the same color for each cycle of CCNa measurements with fixed Sv. As mentioned before, a clear case of a pollution plume is identified in the flight AC11 data, where concentrations of up to 2000 cm^{-3} were measured in both CCN counters. The figure shows that most CCN(Sv) and Nd (Sv) measurements lie within the uncertainty range, except for cases affected by pollution plumes. For clarity, the uncertainties associated with the CCN(Sv) measurements are not shown in the figure. However, considering representative relative uncertainties of approximately 10% in Sv and 14% in CCN number concentration, the agreement between CCN(Sv) and Nd (Sv) remains within the combined uncertainty bounds, supporting the consistency of the closure analysis.

Spatial and temporal heterogeneity of the sampled air masses during flight segments AC11, AC14, and AC17a also contributes to deviations in the fitted relationships over specific supersaturation ranges (e.g., around Sv $\sim 0.55\%$ in AC14). In addition, discrepancies between inferred and measured values may arise from differences in the effective size detection ranges of the cloud droplet probe ($3 \mu\text{m} > d > 50 \mu\text{m}$) and the CCN counters ($d \geq 2 \mu\text{m}$), which sample distinct portions of the particle and droplet spectra.

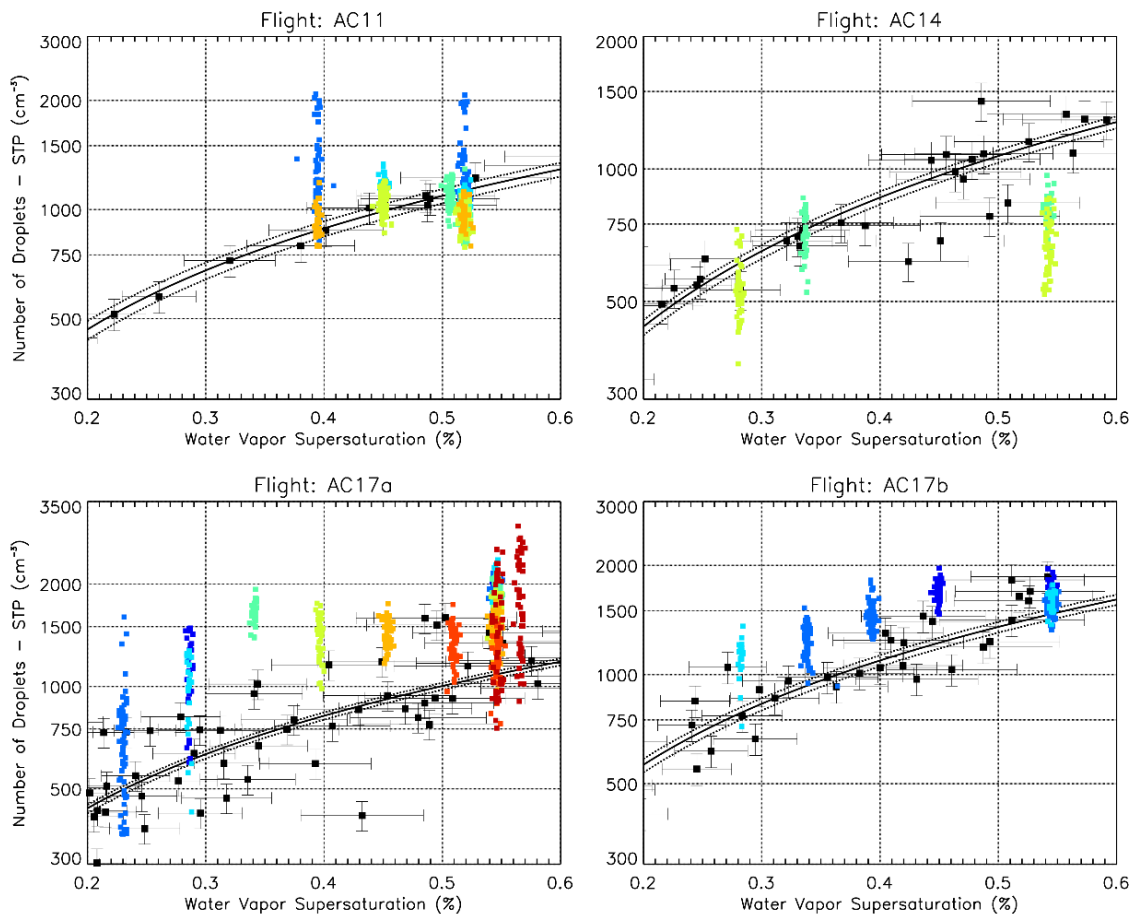


Figure 7 (new). (a) S_v vs. N_d measured at cloud bases (black dots); and S_v vs. CCN concentrations measured below cloud bases [CCNa(S_v) and CCNb(S_v)] for flight segments AC11, AC14, AC17a, and AC17b. CCN measurements are presented using consistent color coding for CCNa and CCNb, corresponding to each CCNa measurement cycle at fixed supersaturation (S_v). Each measurement period includes a minimum sampling duration of 30 s in both counters. Solid curves correspond to power-law fits of the form $N_d = C \cdot S_v^k$ (equations shown in the previous figure), dashed lines indicate the fit with $\pm 1\sigma$. N_d and CCN concentrations are calculated at standard temperature (0°C) and pressure (1013.25 hPa) (STP). Error bars indicate the estimated uncertainties of N_d and S_v , 10% and 12%, respectively. For clarity, uncertainties of CCN concentrations (14%) and S_v (10%) from CCN counters are not shown.

3. Uncertainty analysis.

The manuscript reports uncertainties for measured and inferred variables in Sections 2.5.1, 2.5.4, and 3.2. Those numbers are helpful, but it is unclear how those numbers were obtained. It is also unclear which sources dominate the uncertainty in the inferred S_v . Please provide a more detailed sensitivity analysis of S_v to temperature and LWC uncertainties, etc.

A: We thank the reviewer for this comment. We agree that the original manuscript did not describe the uncertainty analysis with sufficient detail. The measurement uncertainties of temperature (± 0.5 K), pressure (3 Pa) from BAHAMAS data, and liquid water content (10%) from CCP-CDP are analyzed and described in our previous manuscript for the Amazonian flights (Braga et al. 2017) (<https://doi.org/10.5194/acp-17-7365-2017>).

We have contacted the CCN counter PI (Dr. Mira Pohlker) about the uncertainties regarding the flights we used in this manuscript. She updated us that the uncertainty in S_v remains 10%, but the CCN concentrations were recalculated in other analyses, and they end up at about 14% in these measurements (10% from counting efficiencies and 10% due to calibration accuracy). This will be described in the new version.

Following the calculations described in the previous section, the measurement uncertainties were propagated using a Monte Carlo approach, enabling a consistent estimation of uncertainties in both Step 1 - latent energy closure and Step 2 - energy partition. Uncertainties were propagated using a Monte Carlo approach (the code and the resulting analysis is available upon request for review). For each cloud pass (1Hz data), $N = 3000$ – 5000 realizations were generated by perturbing the measured inputs:

$$T_i = T + \delta T, p_i = p + \delta p, LWC_i = LWC \cdot (1 + \delta_{LWC}) \quad (14)$$

where perturbations were sampled from Gaussian distributions:

$$\delta T \sim \mathcal{N}(0, \sigma_T), \delta p \sim \mathcal{N}(0, \sigma_p), \delta_{LWC} \sim \mathcal{N}(0, \sigma_{LWC}) \quad (15)$$

For each cloud pass, the full thermodynamic calculation (Step 1 and Step 2) was recomputed. The uncertainty for each variable X was defined as:

$$\sigma_X = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (X_i - \bar{X})^2} \quad (16)$$

and the relative uncertainty in % was calculated with respect to the measured value:

$$\epsilon_X = \frac{\sigma_X}{X_{\text{nom}}} \cdot 100\% \quad (17)$$

We recalculate the uncertainties of the relevant variables (due to the change in the RHe equation) in this manuscript using this method. Uncertainties in the meteorological measurements from the BAHAMAS system translate into an estimated average uncertainty of about 0.24% (typically < 1% for 1 Hz data) in the derived RHe when calculating using the Bolton equation. The relative uncertainties of the energy terms were about 10% for Q and ΔU , and 13% for W_s . Propagating these errors yields overall uncertainties of ~10% for Sc and SI , and ~12% for Sv . The numerical differences between the energy budget calculations performed in the first and second steps described in the section above were negligible (< ~ 0.01%) due to the invertibility of the latent energy budget.

The power-law relationship $N_d = C \cdot S_v^b$ was fitted for each flight using Monte Carlo uncertainty propagation (using 10% for N_d and 12% for S_v). Across all flights, the exponent is well constrained with uncertainties of $\sigma_b \approx 0.01\text{--}0.03$, while the C shows relative uncertainties of approximately 2–6%. This indicates a robust determination of the N_d - S_v scaling despite measurement uncertainties. The code used for that can also be provided in case of review.

Braga et al.: Comparing parameterized versus measured microphysical properties of tropical convective cloud bases during the ACRIDICON-CHUVA campaign, *Atmos. Chem. Phys.*, 17, 7365–7386, <https://doi.org/10.5194/acp-17-7365-2017>, 2017.

4. The statement that adiabatic models overestimate LWC, buoyancy, and updraft speed. This is one of the manuscript's strongest claims. However, there is no direct comparison against results from other models. Please add such a comparison, either analytically or

numerically, and identify exactly which governing-equation term convectional models omit. Otherwise this claim needs to be softened.

A: Indeed, this is not adequately covered by the content of the current manuscript and will be removed.

5. Concepts/definitions.

- Cloud relative humidity RH_c

The authors introduced a new concept of relative humidity RH_c , and for a saturated parcel or cloud, $RH_c < 1$. I would argue that this is confusing to most readers, because RH for a saturated parcel is normally 100% by definition. I suggest the authors to either rename RH_c to something like a "reference-state saturation ratio" or provide an earlier clarification/explanation of how it differs from conventional RH.

A: We agree with the referee. This was clarified above.

- Cloud supersaturation or cloud supersaturations

The authors first defined cloud supersaturation (S_c) at Line 102, then partitioned it into contributions from the liquid phase (S_l) and the vapor phase (S_v)- But for many instances (e.g., Line 188,259), the authors used "cloud supersaturations" and "cloud supersaturation" interchangeably, which is very confusing.

A: On reflection we can see how this would be confusing to the reader. We have corrected all instances to use the precise terminology in each case.

- ΔU , ΔU_{ws} and Q

In Table 1 and Eqs 18 and 19, ΔU_{ws} and ΔU appear to be 2 different variables as ΔU_{ws} includes WS and ΔU does not. But in the caption of Figure 2, the author states " ΔU_{ws} (indicated as ΔU for simplicity)". This is very confusing. So which inferred variable is actually shown in Figure 2? Please clarify the difference between ΔU_{ws} and ΔU .

Furthermore, if ΔU_{ws} already includes WS, then what is the difference between ΔU_{ws} and Q ?

A: Indeed, the term ΔU in the first step of derivation (see equation 1 above) was removed and substituted by the enthalpy term (ΔH). We consider that the revised description above clarifies any ambiguity in the notation.

- Cloud base

The authors need to provide a clearer definition of cloud base, especially for the aircraft analysis. Is it first nonzero LWC or other thresholds?

A: The theoretical calculations define cloud base when LWC exceeds 0 g m^{-3} . In the aircraft observations, cloud presence is identified using $N_d > 20 \text{ cm}^{-3}$, which in practice corresponds to an $LWC > \sim 0.01 \text{ g m}^{-3}$. This was stated as follows in section 2.4: “ In this study, we focus on the initial stage of cloud formation, and thus, cloud passes with a number concentration of droplets (N_d) greater than 20 cm^{-3} , developing in updraft conditions, are considered.”

6. The structure of the manuscript.

- Introduction

The Introduction is too short (at and before Line 50). It does not provide a thorough and comprehensive background/review of the research topic. The research motivation is unclear and objectives/questions vague. The cited studies are not up to date. Lines 51-79 (including Figure 1) should either go to Methods or at least be largely shortened.

A: We wished to respond quickly to the substantial technical critiques of our manuscript. In the updated manuscript we will provide an improved and extended review of the research topic in the introduction. Figure 1 will move to the Methods section.

- Methods

- Lines 98-115 can be largely shortened. This part seems repetitive of Lines 51-79.
- I would suggest the authors merge Sections 2.1-2.3 into 1 shorter section and show only the very necessary definitions and equations, while other derivations can be more specific and moved into an appendix.
- Section 2.4 mentions iterative estimation of T_0 and related quantities, I think the manuscript would really benefit from a compact algorithm summary: inputs, iteration sequence, criteria, and any quality-control filters applied to cloud passes. Doing so will not only make the proposed workflow more transparent but also benefit model implementation, as the authors noted many times in the paper.

A: We agree with these suggestions and will implement them in full as we revise the manuscript further.

Section 2.5 can also be shortened. I appreciate that the authors provide uncertainties of the observations, but the authors did not state how these uncertainties were estimated/calculated.

A: We believe this comment is now addressed per section 3 of this response.

- Supplementary information

- o Figures S2-S8 are not just ancillary repetitions; they show that the claimed behaviors are robust across environments. This consistency is a strength, and the authors can be more explicit about it in the main text.
- o The simulations in S1 is also interesting and supportive of the results. It shows that the proposed framework behaves smoothly and systematically across temperature, cooling rate, and pressure, which is reassuring from an internal-consistency standpoint. I would suggest the author shorten the simulation analysis and move it to the main text, with a brief section in Methods describing the model configurations.

A: Agreed, we will reorganize the manuscript to clarify the method and include these results in the main text. Most relevant figures will be included.

7. Tables and figures.

- Figure 1: it is helpful to have a schematic figure. But Figure 1 is not clear enough. It mixes physical states, inferred states, and schematic arrows in a way that is difficult to interpret. Please state more clearly which temperatures are observed, which are hypothetical, and which follow from the closure assumptions.

A: Indeed, we apologize for this mistake.

- Figure 2: in panels a and b, there are no color legends.

A: Thanks, we added that.

- Table 1 can be shown much earlier in the paper.

A: OK. This will be moved to the end of the introduction section.

- Tables 2 and 3: it is helpful to have "period of measurements", but it would be more helpful to add a column for the sample size of data collected/analyzed. Perhaps the authors can also add a column to state the location of each flight, instead of repeating it many times in the main text. Suggested captions: ""Measurements at cloud bases ... " and "CCN measurements below cloud bases ... "

A: Figure S1 shows the map with locations. Table 2 shows the 'Period of measurements', this is the sample size of cloud measurements in seconds, the initial time, and the end time are shown in UTC. CCN measurements are continuous; this is why there are more samples.

We will improve the clarity here.

- Figures 3 and 4: if the 8 cloud segments are the same in Table 2 or 3, you don't have to list them out again.

A: The colors indicate flight IDs. We will improve the clarity here.

- Figure 4 and Figures S10-S11: these figures show that N_d scales comparably well with all 3 supersaturations (S_c , S_l , and S_v), then the authors need to explain more carefully what is fundamentally gained by isolating S_v , beyond the fact that it can be interpreted as the vapor-phase fraction of the total supersaturation in the proposed framework. In other words, if N_d can already be well constrained by the convective S_c , then why partition it into 2 different components? Also, I would suggest the authors just panel the three figures together, perhaps remove the power-law fits on each panel or put them in another table if the figure gets too crowded.

A: Thanks for the suggestions. We aim in this manuscript to quantify S_v since it is the only variable that can be compared with CCN counter measurements. We will clarify better the importance of our analysis and why we focus on S_v .

- Figure 5: what's the difference between CCNa and CCNb? Where were they first defined?

A: These are the two CCNc counters (A and B), which we described above. We will clarify it better in the manuscript.

The manuscript needs a cleaner separation between what is observed, what is retrieved, and what is assumed. This distinction should be explicit throughout the manuscript and especially in the figure captions and conclusions.

A: We agree with the referee. We believe the information provided above clarifies the manuscript's message.

Minor comments:

1. Line 16: "We find that ... ". Did you really "find" that? Or you derived/speculated/assigned that? A: Derived...we changed in the new version.
2. Line 29: with respect to "liquid water" or "water vapor"? A: liquid water. The statement is correct.
3. Lines 38-39: are there more recent studies? A: We will add new references about this topic.
4. Line 41: N_d is first defined here. Please use N_d in the remainder of the paper, no need to state "droplet number concentrations" again, e.g., Lines 262 and 341. A:Ok.
5. Line 45: "the activation" of what? A: "...CCN particles..."
6. Lines 45-47: "Even though ... ". So? What is the issue or challenge here? A: The whole challenge is that S_v is not measured with accuracy in clouds. Current airborne relative

humidity instrumentation has uncertainties of about 5% in cloud. Thus, S_v is an estimate based on model assumptions, which presents variability depending on the atmospheric conditions and particles hygroscopicity. In this manuscript, we provide a new method for S_v calculations based only on the thermodynamic of the cloudy parcel. This will be better clarified in the new version of the manuscript.

7. Line 54: 10^{-3} Pa? hPa? A: hPa...thanks
8. Line 61: where is P_0 in Figure 1? A: P_0 is assumed the same as P as described in line 54.
9. Line 65: $C_{p,cloud}$ is not defined yet. A: We moved that figure to methods.
10. Line 98: remove "at cloud base". A: Ok...thanks.
11. Line 100: add "cloud" before "relative humidity", if the authors decide to keep using this name. A: Ok...thanks.
12. Line 102: determine"s". A: Ok.
13. Lines 118-119: Please rewrite this sentence, it is difficult to read. Perhaps "The formulation of RH describes the amount of water vapor in the air relative to the maximum amount of water vapor the air can hold for a given temperature ... ".
A: Ok.
14. Line 123: How is this minor uncertainty estimated/calculated? A: This is from laboratory measurements.
15. Lines 134-135: cooling can not exceed the rate of condensation. I assume the authors meant "cooling rate initially exceeds the rate of condensational heating"? A: yes thanks.
16. Please rewrite Lines 143-144, if the authors decide to keep it. This sentence is difficult to follow. A: Ok. We will provide better wording.
17. Line 150: the specific volume of water vapor is defined as " α_v ", but in Eqs 6, 10, and many other instances, it is written as $\propto v$. A: Ok. We will fix that.
18. Line 162 and Eq 9: it is unclear to me how the authors arrived from Eqs. 1, 4, 5, and 6 to Eq9. A: This derivation is removed from the manuscript...see comments above.
19. Line 165: by "a reference level", you mean "the cloud base"? A: initial temp. T_0
20. Eq10: again, it is confusing to me how the authors arrived from Eq 9 to Eq 10. A: T_e is the same in this case...this derivation is removed from the manuscript.
21. Line 170: density is an intensive property; it doesn't get lost by condensation like mass or number concentrations. I notice that term "vapor density" is used many times in the manuscript, perhaps the authors should also provide the definition. A: vapor density is the inverse of $\alpha_v=1/p_v$.
22. Lines 188-189: change to "where temperature gradients typically are smaller than 1K". A: in our analysis from cloud base measurements the maximum temperature gradients were about 1 K.
23. Line 189: the calculation"s", and by "cloud supersaturation" you mean S_c ? A: Yes.
24. Lines 190-192: please rewrite this sentence. A: Ok.
25. Line 226: please be consistent with the temperature forms. A:Ok...we will improve that.
26. Line 228: how is T_0 estimated? A: We described it above.
27. Line 236: by "these microphysical processes", you mean "expansion, condensation, and latent-heat release"? If so, I think only condensation here is a microphysical process. Expansion, either air parcel or water vapor, is more a thermodynamic process; and latent-heat release is a thermodynamic consequence of a phase-change microphysical process.

A: We agree.

28. Line 237: this is already the third appearance of "for modeling purposes". And here, do the authors refer to any specific model or all numerical models? A: [Adiabatic cloud microphysical models](#).
29. Lines 243-244: what does this sentence mean? A: [We better described it above](#).
30. Line 261: I think Table 2 just provides basic atmospheric conditions (same comment for Table 3). And which "cloud properties" were measured at cloud bases? A: [The tables provide the general atmospheric conditions during the measurements. CCP measured the DSDs from 3-960 micron. The relevant measurements to our study are Nd and LWC. We will clarify that in the new version.](#)
31. Lines 262-263: "cloud passes ... developing in updraft conditions". Please rewrite this sentence. Clouds can develop in updraft conditions, cloud passes can not. Plus, was the updraft also measured during flights? How strong updraft speeds are considered? A: [This is described in section 2.5.1. The histogram with updraft velocities are shown in the supplementary material.](#)
32. Line 270: remove "measured". The authors need to be more clearly stating which variables are inferred, which are calculated, and which are measured. A: [Ok. Thanks.](#)
33. Step 2 around Line 270: did the authors calculate/estimate T_{ows} from a given WVs ? Or did the authors calculate/estimate WVs from T_{ows} ? This is unclear to me. And this step is difficult to follow. A: [We better described it above.](#)
34. Lines 278-279: which is measured by SHARC? Which is derived? A: [Mixing ratio of water vapor.](#)
35. Line 283: define CCP. A: [Ok.](#)
36. Line 293: cloud base"s". A: [Ok.](#)
37. Line 298: remove "water vapor supersaturation". A: [Ok.](#)
38. Line 299: "... exposed to a set supersaturation", you mean S_v ? A: [Yes.](#)
39. Line 308: remove "rain water content". A: [Ok.](#)
40. Remove Lines 337-340, this is redundant. A: [Ok.](#)
41. Line 341: perhaps provide one-sentence justification for this specific case. And remove "droplet number concentrations", you have defined it earlier. A: [Ok.](#)
42. Line 344: remove "(in black)". A: [Ok.](#)
43. Line 344: is each dot a cloud pass? A: [1 Hz data.](#)
44. Lines 345-347: "This variability arises ... ", again? A: [Ok...removed that.](#)
45. Line 347: remove "(in red)". A: [Ok.](#)
46. Line 352: what do you mean by "cloud elements"? and how were "stronger buoyancy and more vigorous condensational growth" inferred? A: [Here, 'cloud elements' refers to localized in-cloud 1 Hz sampling volumes along the flight track. 'Stronger buoyancy' is not directly measured but inferred from updraft velocities shown in the supplement \(Fig.S9\). 'More vigorous condensational growth' is inferred from higher supersaturation \(\$S_v\$ \) and increased liquid water content.](#)
47. Lines 353-354: by "cloud supersaturation", you mean S_c ? or which one? A: [The sentence refers to the results in figure 2.](#)
48. Line 367: what kind of simulations? A: [We explored our calculations for a range of cooling rate](#)

and initial temperature. This is shown in the supplement (Section S1). The results of the updated simulations are shown above.

49. Lines 393-395: how was the "buoyant forcing" measured/inferred? The first sentence reads to me like "because there are lower LWC and updraft speeds in CCN-limited regions, so there is weaker buoyant forcing". But then the next sentence reads like "in polluted regions, because there is stronger buoyant forcing, so there are more droplet formation (so higher LWC?) and higher updraft speeds". The authors need to be more careful and clearer with these statements.

A: Buoyant forcing is not directly measured but inferred from updraft velocities shown in the supplement...Figure S9.

50. Line 405: how does the cooling of the cloud parcel reflect the relationship between LWC and Nd? A: We described it in section 2.2. A better explanation is shown above.

51. Lines 412-421 can be largely shortened. This part could have been stated in Methods. A: Ok. Thanks

52. Line 426: I am not sure if the Nd and CCN spectra comparisons "were within uncertainty range". For Flight 17 (Figure 5 c and d), the CCN spectra is above the Nd spectra.

A: better explanation is shown above.

Line 434: is there really an improved agreement for Flight 17? A: A better explanation is shown above.

Line 440: what do you mean by "appears reliable"? and please check the grammar and rewrite this whole sentence. A: Indeed, it is strange we will provide better wording in case of review.

Line 455: which "both cases"? A: The Sv from our calculations and those from CCN counters. We will provide better wording in case of review.

53. Lines 472-473: were those lapse rates used anywhere in Section 2? A: This is shown in the updated figure and discussions above.

Lines 473: "Our results suggest... ", which part of results or figure exactly? A: Our results show that the LWC is overestimated when Ws term is neglected in the calculations of energy budget. We will provide better wording in case of review.