

Dear Referee #2 and Referee #3,

We sincerely thank you for your constructive reviews of our manuscript. Your comments and suggestions have significantly improved the clarity of the theoretical framework and the overall quality of the manuscript.

In the present compiled response document, the authors' initial replies to the referees' comments, as published online during the discussion phase, are shown in **blue**, while modifications incorporated into the revised manuscript are indicated in **red**.

We recognized from the comments of both referees that the description of the thermodynamic framework in the previous version of the manuscript could be misinterpreted, and that Figure 1 may have contributed to this ambiguity. We have therefore updated the figure and substantially clarified the underlying assumptions of the methodology. Specifically, the derived water-vapor supersaturation (S_v) is calculated as a thermodynamic analogue of the supersaturation established in a CCN counter. The saturated air parcel is treated as an adiabatic, reversible, and closed thermodynamic system, such that the initial saturated-vapor state can be recovered from the vapor–liquid state by reversing the thermodynamic transformation.

Accordingly, the method does not calculate the energy budget associated with an extended moist-adiabatic ascent of an air parcel through the atmosphere. Instead, it evaluates the energy budget associated with a short transition between saturated thermodynamic states under adiabatic and locally quasi-isobaric conditions. This approximation is supported by the small pressure variability observed during the cloud-base passes, typically up to approximately 1 hPa (equivalent to about 10 m in altitude), corresponding to $\sim 0.1\%$ of the ambient pressure. Consequently, the temperature difference used in the diagnostic should not be interpreted as the cooling produced by a vertical displacement through the moist-adiabatic lapse rate. Rather, it represents the cooling associated with the sensible enthalpy change of the saturated parcel under approximately constant-pressure conditions within the assumptions of the proposed thermodynamic framework.

Following the referees' recommendations, the revised manuscript includes further clarification of the theoretical framework, the physical interpretation and terminology of the derived supersaturation quantities, the assumptions concerning cloud microphysics, and the relationship between the proposed diagnostic and existing approaches for estimating water-vapor supersaturation. Additional explanations and discussion have been incorporated throughout the manuscript to address the specific concerns raised by both referees.

Finally, following discussion among the authors, we revised the manuscript title to better reflect the scope and objectives of the study.

We believe that these revisions have substantially strengthened the manuscript and have addressed the reviewers' comments and concerns. We therefore hope that the revised manuscript is suitable for publication in *Atmospheric Chemistry and Physics*.

We again thank the referees for their valuable contributions to improving this work.

Sincerely,

Ramon Braga, Manfred Wendisch, Rachel Albrecht and Daniel Harrison

Referee #2:

General comments:

The paper proposes a novel method to diagnose vapor-phase supersaturation at cloud base by utilizing measured liquid water content (LWC), temperature (T), and pressure (p). The latent heat (Q) is partitioned into changes in internal energy and saturation work. This method was applied to ACRIDICON-CHUVA campaign data, and the derived N_d (S_v) spectra were successfully compared with sub-cloud NCCN(S_v). This revised manuscript adequately addressed the concerns raised during initial review and my concerns are therefore not with the goal, the data or the care taken, but with the theoretical explanation and assumptions. The manuscript had been revised significantly making it extremely difficult to follow with unflattened track-changes and duplicate equation numbers resulting in significant delay.

Since some analysis requires an important clarification, it is recommended that manuscript is reconsidered after a minor revision with cleaner formatting.

Specific Comments:

1. The entire introduction section needs to be revised to ensure a consistent flow. For example, consider moving sentence starting with “In general, ...” at lines 67-70 after discussing all the current methods in later part of introduction. Also consider removing the sentence starting with “In this study we introduce...” at line 71 since it is repeated later at line 128.

A: We have revised the Introduction and created a new section to improve the clarity of the methodology and better prepare the reader for the results presented in this study. The referee's suggestions to move or remove specific sentences were carefully considered to improve the overall flow of the section. The main changes are provided in the new version as below.

“... ”

In general, current estimates of S_v at cloud base are inferred from aerosol-activation parameterizations or cloud parcel models rather than being directly constrained by independent in situ measurements (Campos Braga et al., 2017; Pinsky et al., 2012; Rosenfeld et al., 2016; Twomey, 1959; Varghese and Prabhakaran, 2025). Although these approaches reproduce observed N_d at cloud base, direct observational validation of S_v remains challenging (Campos Braga et al., 2017; Braga et al., 2021). For in situ cloud-base conditions, the quasi-steady supersaturation (S_{QSS}) is also not applicable within the activation layer because it requires dynamical forcing and the droplet population to vary slowly relative to the supersaturation-adjustment timescale, a condition that is not satisfied during rapid aerosol activation at cloud base (Romps et al., 2023; Romps, 2025).

In this study, we propose a new method to derive S_v at cloud bases for saturated parcels considering the energy and water vapor budgets during the condensational growth process of liquid droplets. S_v is diagnosed from the thermodynamic states of the saturated parcel, and the transition from the saturated-vapor state to the vapor-liquid state is treated as quasi-isobaric over the short cloud-base transition ($\Delta z \approx 10$ m), for which the expected hydrostatic pressure change is of the order 1 hPa ($\Delta p \approx \rho g \Delta z$), corresponding to ~ 0.1 % of the ambient pressure. At cloud base, in situ measurements typically show slightly lower air pressure in cloudy than cloud-free air, with higher LWC and water vapor mixing ratios at lower temperatures for quasi-isobaric conditions. The diagnostic thermodynamic state of the saturated parcel is determined primarily by the atmospheric air pressure (p), the initial air parcel temperature (T_0) of the saturated-vapor state, and the final temperature (T) of the vapor-liquid state, which constrains the calculation of the saturation ratio. At cloud base level, the atmospheric pressure and the specific heat capacity of cloudy air ($c_{p,cloud}$) vary only slightly over the short-saturated transition. Therefore, this formulation represents a quasi-isobaric thermodynamic transition rather than an extended moist-adiabatic ascent in which the temperature difference between the two thermodynamic states is directly related to vertical displacement through the saturated adiabatic lapse rate. Within this quasi-isobaric framework, the thermodynamic transition between saturated-vapor and vapor-liquid states is then constrained by the parcel energy budget.

2. Thermodynamic framework and measurement strategy

Here, the transition of thermodynamic states is assumed to be adiabatic and reversible, such that it follows an idealized reversible thermodynamic process, and the initial saturated-vapor state can be recovered from the vapor-liquid state by reversing the energy transformation. The energy associated with the vapor-to-liquid transition is constrained by the sensible enthalpy difference (dH) between the two thermodynamic states and by the latent energy released during condensation (Q), such that $\delta Q = dH$. Q is interpreted as an internal energy redistribution within the parcel rather than as external diabatic heating. The partitioning of the condensational energy of the saturated parcel is formulated from the first law of thermodynamics, such that the latent energy associated with condensation, Q , is partitioned between changes in parcel internal energy (ΔU) and the work associated with water vapor expansion (W_s), according to $Q = \Delta U + W_s$. Within this diagnostic framework, part of the condensational energy is associated with W_s during liquid-water formation relative to the limiting case in which the entire condensational energy contributes to liquid formation ($\delta Q = dH$). Consequently, the LWC is reduced, resulting in a relative excess of water vapor. We define the cloud supersaturation (S_c) from the vapor pressures of the saturated parcel in the vapor and liquid states for the limiting case in which the entire condensational energy contributes to liquid formation. Since part of Q is associated with W_s during the vapor-to-liquid transition, we diagnostically define the liquid-phase supersaturation (S_l) and vapor-phase (S_v) as the respective fractions of Q associated with ΔU and W_s (i.e., $S_c = S_l + S_v$). The corresponding temperatures of the vapor and liquid states obtained from the energy-budget calculations define the vapor pressures of the saturated parcel during the vapor-to-liquid transition.

The derived S_v provides a thermodynamic analogue to the effective supersaturation established in a CCN counter (CCNc), where the instrument supersaturation is calibrated from the critical activation of particles with known physicochemical properties, e.g. ammonium sulfate, using Köhler theory (Roberts and Nenes, 2005; Rose et al., 2008). This correspondence allows the cloud-base N_d (S_v) relationship to be represented by the classical power-law CCN activation spectrum, $N_d = C \cdot S^k$ (Twomey, 1959), in which increasing supersaturation activates progressively larger fractions of the available CCN population. Unlike conventional parcel-model or CCN-closure approaches, which estimate S_v from prescribed w and CCN properties to reproduce the observed N_d , the present method diagnoses cloud-base S_v from a thermodynamic energy-budget closure constrained primarily by condensational energy and LWC . The method is evaluated using in situ measurements of cloud-base properties and compared with N_{CCN} (S_v) observations below cloud base. In this framework, Q is directly derived from measured LWC , air temperature (T) and pressure (p) at cloud base, while T_0 , ΔU , W_s , S_l , and S_v are diagnostically estimated. The airborne cloud base data were collected in growing convective cumuli over the Amazon Basin in September 2014 during the ACRIDICON-CHUVA campaign (Wendisch et al., 2016), encompassing polluted conditions over forested and deforested regions, and clean marine conditions over the adjacent Atlantic Ocean. ACRIDICON stands for “Aerosol, Cloud, Precipitation, and Radiation Interactions and Dynamics of Convective Cloud Systems”; CHUVA is the acronym for “Cloud Processes of the Main Precipitation Systems in Brazil: A Contribution to Cloud Resolving Modeling and to the GPM (Global Precipitation Measurement)”. The High Altitude and Long Range Research Aircraft (HALO) was used during the campaign (Stevens et al., 2019). The resulting cloud-base N_d (S_v) spectra was calculated for eight flight segments and, for the flights in the Central Amazon, were compared with the N_{CCN} (S_v) spectra measured below cloud base. This comparison tests the robustness of the underlying physical assumptions, as the cloud-base N_d (S_v) spectrum is expected to represent the N_{CCN} (S_v) population below cloud bases. Figure 1 summarizes the measurement strategy and the diagnostic thermodynamic framework used in this study. Cloud-base measurements provide N_d , LWC , T , and p while N_{CCN} (S_v) is independently measured below cloud base (Fig. 1a). From the measured cloud-base state, the initial saturated-vapor state is diagnostically reconstructed under the assumptions of an adiabatic and reversible transition treated locally as quasi-isobaric (Fig. 1b). The condensational energy is partitioned according to $Q = \Delta U + W_s$, from which the liquid- and vapor-phase contributions to cloud supersaturation, S_l and S_v , are derived. The resulting cloud-base N_d (S_v) spectrum is then compared with the independently measured sub-cloud N_{CCN} (S_v) spectrum as a closure test of the proposed framework. The following section 3 presents the proposed method and its underlying physical laws, while the results and discussion are provided in the subsequent sections. Table A1 in the Appendix lists and defines the symbols used in this manuscript.

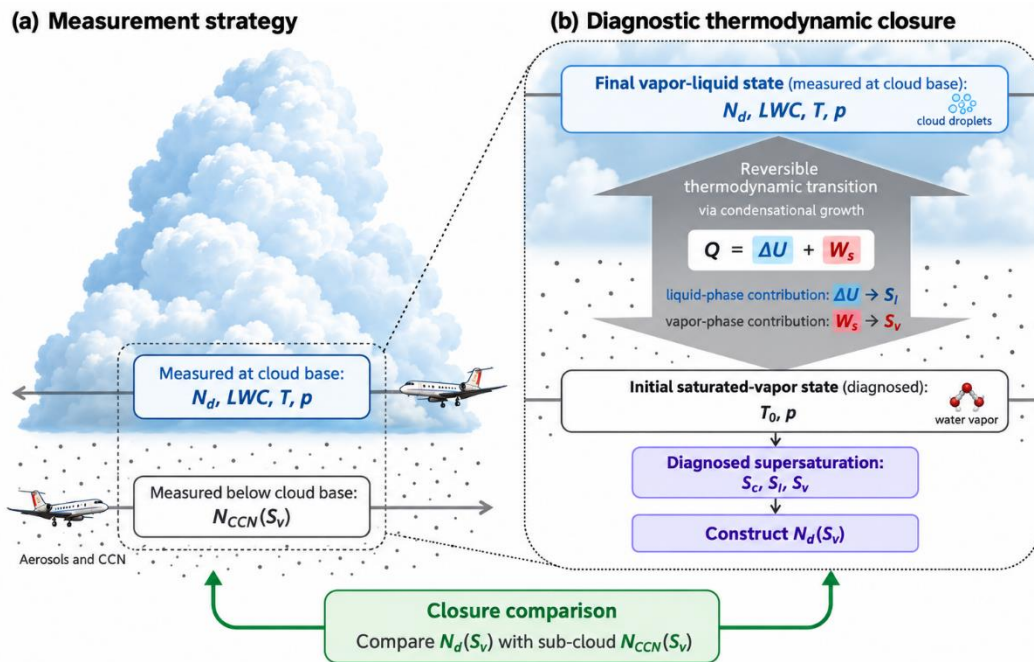


Figure 1. Measurement strategy and diagnostic thermodynamic closure used to derive water vapor supersaturation at cloud base. (a) Schematic of the airborne measurement strategy. At cloud base, the HALO aircraft measures droplet number concentration (N_d), liquid water content (LWC), temperature (T), and pressure (p). Below cloud base, cloud condensation nuclei activation spectra, $N_{CCN}(S_v)$, and pressure are measured. The highlighted cloud-base region indicates the local layer to which the thermodynamic closure is applied. (b) Schematic of the diagnostic thermodynamic framework. The measured cloud-base state is treated as the final vapor-liquid state, characterized by N_d , LWC , T , and p , while the corresponding initial saturated-vapor state, characterized by T_0 and p , is diagnosed. The transition between these states is represented as a reversible thermodynamic transformation during condensational growth. The latent heat (Q) released by condensation is partitioned according to $Q = \Delta U + W_s$, where the internal-energy contribution is associated with the liquid-phase supersaturation S_l and the water-vapor expansion work with the vapor-phase supersaturation S_v . The diagnosed supersaturations S_c , S_l , and S_v are then used to construct the cloud-base $N_d(S_v)$ spectrum, which is compared with the independently measured sub-cloud $N_{CCN}(S_v)$ spectrum as the closure test.”

2. The framework of “conserved isobaric enthalpy” justified by small pressure variability of “0.1-0.3%” (Line 209) is questionable with a physically ascending parcel and the smallness of Δp cannot defend it, please justify.

Figure 1 and its caption Line 248-262 and lines 225-226 (“undergoes a small upward displacement to slightly lower pressure”) describe a physical displacement. For displacement, the conserved quantity should be moist static energy, not enthalpy. The difference is $g \cdot dz$ and for adiabatic motion $-g \cdot dz$ equals $c_p \cdot dT$. So, the neglected term is the same size as the entire budget $Q = C_{p,cloud}(T_0 - T)$ of Eq. 2 (Line 286) at any displacement, however small. So even a small pressure change cannot hold this key approximation. Looking at your data, largest reported cooling of 1.81K for AC17a (Lines 791-792) implies ascent of ~180m considering saturated lapse rate of 4.5 K km⁻¹, which is a pressure change of ~2%, an order of magnitude beyond the 0.1-0.3% invoked at line 209. Several other statements (e.g., AC18, $\Delta T_c = 0.71$ °C, Line 788) exceed the stated bounds as well.

A: We agree with the referee that the reviewed version of the manuscript suggests this interpretation. Figure 1 was updated from the manuscript to avoid mistake as shown above.

We state in the Methods the following regarding the conserved isobaric enthalpy:

“The transition is further assumed to be adiabatic and reversible, and the total moist enthalpy approximately conserved over the short cloud-base transition, while the sensible enthalpy changes between the saturated-vapor and vapor-liquid states.”

We clarified in the new introduction (see above) that the derived S_v is calculated as a thermodynamic analogue to the supersaturation established in a CCN counter. It means that we are not calculating the energetic budget of a moist adiabatic parcel ascending in the atmosphere, but rather we are calculating the energy budget in a closed system, which is considered adiabatic and quasi-isobaric. The estimated cooling associated with the supersaturation does not express a vertical displacement, but the cooling associated with the change in the sensible enthalpy of the cloud parcel under the physical assumptions of the methodology. Thus, differences such as 1.81 K or 0.71 K should not be interpreted as implying 180 m or a corresponding moist-adiabatic displacement in the energy calculation.

3. Adiabaticity assumption for the sampled clouds may not apply to most of the observations. Section 3.2, in-cloud RH ranged ~60-102% (Lines 709-710) attributed to sensor wetting and entrainment (Lines 711-716). If entrainment affects 60-90% readings, then the reversible-adiabatic premise (Sec 2.4) fails where the method is applied. The $N_d > 20 \text{ cm}^{-3}$ / $RWC=0$ selection (Line 526-528) does not address this. Consider stratifying Fig. 6, 7 closure by an adiabaticity proxy (LWC/LWC_{ad} , or the $RH-RH_c$ residual already computed in Fig 4a) to show whether the agreement improves for near adiabatic passes. A positive result will further strengthen the method.

A: The adiabatic assumption is clarified in the new introduction section.

The method uses the measured LWC directly rather than replacing it with an adiabatic LWC; therefore, dilution that reduces the observed condensate is reflected in Q . However, entrainment or mixing occurring during the diagnosed transition can also modify T and LWC and can bias the inferred partition.

The figure 4 in Section 4.2 (previous 3.2), which the referee reference, exemplifies a case study in which the air parcel is saturated (i.e., $N_d > 20 \text{ cm}^{-3}$ or $LWC \geq \sim 0.01 \text{ g m}^{-3}$), but the vapor pressure or RH is variable due to environmental conditions and the cloud life cycle. For this case study and the other flights most of estimated vapor pressure resulting from our methodology were within the uncertainty range of the in-situ measurements (shown in the supplement material for each flight).

We agree that entrainment can affect the closed system approximation and we have clarified this as a limitation while comparing the water vapor pressures estimated vs. measured. This is described as the following:

“...Figure 4a shows that RH generally increases as a function of N_d even though in most cloud passes RH is smaller than 100 %. This variability in RH measurements arises from the characteristics of in situ temperature measurements: liquid water impinging on the sensor can induce evaporative cooling, whereas the latent heat released during particle condensational growth and buoyant air volumes may locally warm the sensor. RH_c decreases consistently with N_d associated with higher LWC . For cloud passes where $RH < RH_c$ the local vapor pressure $e(T)$ is typically smaller than expected from the cooling trajectory of the initially saturated parcel, suggesting enhanced entrainment of unsaturated environmental air and reduced condensational growth (see Figure S4). Under such conditions, smaller droplets evaporate preferentially due to the inhomogeneous nature of mixing in convective clouds. This selective evaporation reduces the local LWC , and the remaining droplet population is biased toward droplets formed on CCN with higher hygroscopicity, which are more resistant to evaporation. Conversely, cases in which $RH > RH_c$ indicate that the parcel retains a larger vapor content relative to theoretical calculations, favouring continued condensational growth, enhanced S_v , yielding higher N_d for a given CCN population.”

This is also discussed in Sect. 4.3, where the comparison between $N_d(S_v)$ and $N_{CCN}(S_v)$ is presented over the S_v range of approximately 0.2–0.55%, as follows:

“...Overall, most $N_d(S_v)$ and $N_{CCN}(S_v)$ values remain within the combined uncertainty bounds, whereas the largest deviations are associated with variability in polluted air masses and with derived $N_d(S_v)$ values from cloud passes more strongly affected by the entrainment and mixing of dry air.”

In this manuscript, we do not aim to investigate cloud adiabaticity in detail, but rather to describe the proposed methodology as applied to cloud-base measurements and to evaluate it through comparison with CCN measurements obtained below cloud base. We agree that such an analysis would be valuable in a dedicated follow-up study, particularly by comparing the conventional adiabatic liquid water content with the LWC derived from the proposed method over a broader vertical extent of the saturated parcel.

Technical corrections:

Units: Mixing ratio “kg-1kg-1” throughout text should be “kg kg-1”

A: Ok...thanks.

Line 57: Consider replacing the word "salty" droplets with "solution" droplets.

A: Ok...changed for “solution”.

Line 76: "Clayperon-Clausius relation" should be written as "Clausius-Clapeyron relation"

A: Ok...thanks.

Line 539-540: Please mention equation number used to calculate RHc and Sc.

A: Sorry we could not track that comment but Equations 10 and 11 are those used to calculate RHc and Sc, respectively (described in Section 3.2).

Line 1170: Squires (1952) is referenced but I could not find it cited in the text.

A: It is shown in the manuscript as follows.

“Rosenfeld, D., Zheng, Y., Hashimshoni, E., Pöhlker, M. L., Jefferson, A., Pöhlker, C., Yu, X., Zhu, Y., Liu, G., Yue, Z., Fischman, B., Li, Z., Giguzin, D., Goren, T., Artaxo, P., Barbosa, H. M. J., Pöschl, U., and Andreae, M. O.: Satellite retrieval of cloud condensation nuclei concentrations by using clouds as CCN chambers, *Proceedings of the National Academy of Sciences*, 113, 5828–5834, <https://doi.org/10.1073/pnas.1514044113>, 2016.

Squires, P.: The growth of cloud drops by condensation. I. General characteristics, Aust. J. Chem., 5, https://doi.org/10.1071/CH9520059, 1952.

Twomey, S.: The nuclei of natural cloud formation part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, *Geofisica Pura e Applicata*, 43, 243–249, <https://doi.org/10.1007/BF01993560>, 1959.”

Table 1, Line 635: AC19 is dated 20-Sep but is listed after AC18 (28-Sep), please explain.

A: The date is 30-Sep...thanks.

Review of Braga et al. “Energetic Derivation of Water Vapor Supersaturation at Cloud Base”

Braga et al. propose a new method for quantifying supersaturation near the cloud base in developing cumuli with aircraft observations. The method is based on energy-budget arguments, allowing it to avoid some parameterizations in conventional methods. Because cloud-base supersaturation is an important quantity for the activation of cloud-condensation nuclei aerosols, the paper and method have the potential to make a meaningful contribution to the field. However, the paper in its current form is unclear in some important ways and lacks a comparison to other studies and methods, which makes it harder to accept the argument that the current method is superior to existing methods. If these issues can be addressed, then the paper may be suitable for publication.

Major comments:

- My first major comment is that several important aspects of the theory and method are not adequately explained.

First, I find the difference between the terms “liquid supersaturation (Sl)”, “water vapor supersaturation (Sv)” and “cloud supersaturation (Sc)” quite confusing. In the cloud physics textbooks I have read, supersaturation is computed relative to an infinite flat surface of pure water. I suspect that much of the audience of this paper will have that concept in mind. It would be really helpful if the authors could explain how Sl, Sv, and Sc are related to the common textbook definition of supersaturation, and how they should be physically interpreted. It is especially confusing that Sl, Sv, and Sc are used several times before they are defined near line 260.

A: The supersaturation calculated relative to an infinite flat surface of pure water is the water vapor supersaturation (Sv). In the conventional cloud-physics definition, water-vapor supersaturation over an infinite flat surface of pure liquid water is $S_v = e/e_s(T) - 1$ (shown in the Appendix A1). Sv is an excess of water vapor in a saturated air parcel. In our method the Sv is resulting from the energy partition of the condensational energy budget, which is calculated from the energy fraction and the resulting temperatures (saturated vapor pressures) from the saturated parcel during the vapor-to-liquid transition (see the definitions of Sc, Sl and Sv in sections 3.3 and 3.5).

We edited the introduction section and created a new section to describe calculated supersaturations as follows:

“2. Thermodynamic framework and measurement strategy

Here, the transition of thermodynamic states is assumed to be adiabatic and reversible, such that it follows an idealized reversible thermodynamic process, and the initial saturated-vapor state can be recovered from the vapor-liquid state by reversing the energy transformation. The energy associated with the vapor-to-liquid transition is constrained by the sensible enthalpy difference (dH) between the two thermodynamic states and by the latent energy released during condensation (Q), such that $\delta Q = dH$. Q is interpreted as an internal energy redistribution within the parcel rather than as external diabatic heating. The partitioning of the condensational energy of the saturated parcel is formulated from the first law of thermodynamics, such that the latent energy associated with condensation, Q , is partitioned between changes in parcel internal energy (ΔU) and the work associated with water vapor expansion (W_s), according to $Q = \Delta U + W_s$. Within

this diagnostic framework, part of the condensational energy is associated with W_s during liquid-water formation relative to the limiting case in which the entire condensational energy contributes to liquid formation ($\delta Q = dH$). Consequently, the LWC is reduced, resulting in a relative excess of water vapor. We define the cloud supersaturation (S_c) from the vapor pressures of the saturated parcel in the vapor and liquid states for the limiting case in which the entire condensational energy contributes to liquid formation. Since part of Q is associated with W_s during the vapor-to-liquid transition, we diagnostically define the liquid-phase supersaturation (S_l) and vapor-phase (S_v) as the respective fractions of Q associated with ΔU and W_s (i.e., $S_c = S_l + S_v$). The corresponding temperatures of the vapor and liquid states obtained from the energy-budget calculations define the vapor pressures of the saturated parcel during the vapor-to-liquid transition.

...”

Second, the paper would benefit from an explanation about what microphysical assumptions are being made. Cloud microphysics, in particular the integral radius of the cloud droplets, affects the condensation term “Cd” in the supersaturation development equation in Eq. 1. However, the parcel model on which the authors’ method is based does not explicitly discuss or consider microphysics at all. There must be some implicit assumption about the cloud microphysical properties baked into the theory, right? It would help if the authors can explain this.

A: We thank the referee for raising this important point. Equation (1) is the conventional prognostic equation describing the temporal evolution of S_v in an ascending cloud parcel. In this formulation, cloud microphysical properties are required because the condensational sink depends on the diffusional growth rate of the droplet population and therefore on quantities such as the integral droplet radius.

In the present manuscript, however, Eq. (1) is not used to run a parcel model or to calculate S_v for comparison with our diagnostic. It is introduced to describe the conventional framework for S_v calculation and to provide context for the different thermodynamic approach proposed here. Our method does not calculate $(dLWC/dt)$, the diffusional growth rate of individual droplets, or the temporal evolution of S_v . Consequently, no integral droplet radius or explicit droplet-growth parameterization is required.

In the new version of the Introduction section, we justify and clarify our physical assumption as follows:

“In general, current estimates of S_v at cloud base are inferred from aerosol-activation parameterizations or cloud parcel models rather than being directly constrained by independent in situ measurements (Campos Braga et al., 2017; Pinsky et al., 2012; Rosenfeld et al., 2016; Twomey, 1959; Varghese and Prabhakaran, 2025). Although these approaches reproduce observed N_d at cloud base, direct observational validation of S_v remains challenging (Campos Braga et al., 2017; Braga et al., 2021). For in situ cloud-base conditions, the quasi-steady supersaturation (S_{QSS}) is also not applicable within the activation layer because it requires dynamical forcing and the droplet population to vary slowly relative to the supersaturation-adjustment timescale, a condition that is not satisfied during rapid aerosol activation at cloud base (Romps et al., 2023; Romps, 2025).

In this study, we propose a new method to derive S_v at cloud bases for saturated parcels considering the energy and water vapor budgets during the condensational growth process of liquid droplets. S_v is diagnosed from the thermodynamic states of the saturated parcel, and the transition from the saturated-vapor state to the vapor-liquid state is treated as quasi-isobaric over the short cloud-base transition ($\Delta z \approx 10$ m), for which the expected hydrostatic pressure change is of the order 1 hPa ($\Delta p \approx \rho g \Delta z$), corresponding to ~ 0.1 % of the ambient pressure. At cloud base, in situ measurements typically show slightly lower air pressure in cloudy than cloud-free air, with higher LWC and water vapor mixing ratios at lower temperatures for quasi-isobaric conditions. The diagnostic thermodynamic state of the saturated parcel is

determined primarily by the atmospheric air pressure (p), the initial air parcel temperature (T_0) of the saturated-vapor state, and the final temperature (T) of the vapor-liquid state, which constrains the calculation of the saturation ratio. At cloud base level, the atmospheric pressure and the specific heat capacity of cloudy air ($c_{p,cloud}$) vary only slightly over the short-saturated transition. Therefore, this formulation represents a quasi-isobaric thermodynamic transition rather than an extended moist-adiabatic ascent in which the temperature difference between the two thermodynamic states is directly related to vertical displacement through the saturated adiabatic lapse rate. Within this quasi-isobaric framework, the thermodynamic transition between saturated-vapor and vapor-liquid states is then constrained by the parcel energy budget.

”

In the last paragraph of the new section 2 we also state that:

“The derived S_v provides a thermodynamic analogue to the effective supersaturation established in a CCN counter (CCNc), where the instrument supersaturation is calibrated from the critical activation of particles with known physicochemical properties, e.g. ammonium sulfate, using Köhler theory (Roberts and Nenes, 2005; Rose et al., 2008). This correspondence allows the cloud-base N_d (S_v) relationship to be represented by the classical power-law CCN activation spectrum, $N_d = C \cdot S^k$ (Twomey, 1959), in which increasing supersaturation activates progressively larger fractions of the available CCN population. Unlike conventional parcel-model or CCN-closure approaches, which estimate S_v from prescribed w and CCN properties to reproduce the observed N_d , the present method diagnoses cloud-base S_v from a thermodynamic energy-budget closure constrained primarily by condensational energy and LWC.”

Third, Eq. 14 seems to assume that the process is isothermal. Does this conflict with other assumptions in the theory? A justification for this step is needed.

A: The process is not isothermal but quasi-isobaric. The Eq. 14 shown below considers the initial and final temperature in the logarithm term. The warmer temperature out of the logarithm indicates the initial temperature of the saturated parcel. The reversibility analysis shows that the changes in the energy during from saturated vapor-state to liquid-state and vice versa leads to negligible differences in the thermodynamic properties calculated (see Section 3.4).

$$W_s = r_s \cdot \int_{\alpha_{vs0}}^{\alpha_{vs}} e_s d\alpha_v = r_s \cdot \int_{\alpha_{vs0}}^{\alpha_{vs}} \frac{R_v \cdot T}{\alpha_v} d\alpha_v \approx r_{s0} \cdot R_v \cdot T_0 \cdot \ln \left(\frac{\alpha_{vs}}{\alpha_{vs0}} \right) = r_{s0} \cdot R_v \cdot T_0 \cdot \ln \left(\frac{T}{T_0} \cdot \frac{e_s(T_0)}{e_s(T)} \right) \quad (14)$$

Fourth, why is the analysis limited to cumuli at the initial stage of development? The method seems agnostic to the lifecycle stage of the cumuli. It would help to justify and explain this choice.

A: Cloud base represents the initial stage of cloud formation, where the spectrum of CCN below cloud base is activated. The analysis intended to compare the sub-cloud CCN(S_v) spectra with the cloud base N_d (S_v) spectra. Above cloud base, entrainment and mixing, secondary activation etc. can progressively alter N_d and LWC, weakening the direct correspondence with the CCN population sampled below cloud base.

We have added this explanation in the Methods section as follows:

“In situ measurements at the cloud bases of growing cumulus clouds are used to evaluate the theoretical assumptions. N_{CCN} (S_v) spectra measured just below cloud base, prior to cloud measurements, are compared with the N_d (S_v) spectra derived at cloud base during flights in the Central Amazon. The N_d (S_v) spectrum is considered representative of the N_{CCN} (S_v) population below cloud base that, after buoyant ascent, is activated into cloud droplets. The analysis is restricted to the cloud-base activation stage, where sub-cloud

CCN can be related most directly to newly formed droplets. Farther above cloud base, the effects of entrainment of dry air, evaporation, secondary activation of CCN and coagulation processes weaken this correspondence.”

Fifth, it would help if the authors can be more explicit about where in the cloud their method is applicable. The current draft just says that the supersaturation is estimated ”at cloud base”. Does this mean the lowest layer of the cloud where CCN activate, supersaturation changes rapidly with height, and supersaturation reaches a local maximum (approximately the lowest 10 m of the cloud)? Or is it valid above this layer? It seems like the method can’t be valid in the activation layer since microphysical details are very important to the supersaturation development there but are not explicitly treated in the derivation. If the method is only valid above the activation layer, then the proposed method provides the same information as the standard quasi-steady-state approximation for estimating supersaturation (Squires, 1952). A comparison with that method, including explanations of advantages and disadvantages of each method, would be really helpful.

A: As described above the diagnostic method is applied to the first cloudy samples at cloud base, i.e. the short-saturated vapor-to-liquid transition (~ 10 m) in which liquid water and newly activated droplets are detected under updraft conditions. It is not a prognostic parcel model and does not calculate the local time evolution of S_v or identify a dynamically generated maximum supersaturation (S_{max}). Therefore, the derived S_v should be interpreted as a finite-state thermodynamic analogue associated with the sampled cloud-base transition, rather than as the classical parcel-model S_{max} . This is also different from the quasi-steady approximation of Squires (1952). The quasi-steady regime occurs when supersaturation adjusts rapidly enough that its production by cooling is approximately balanced by condensational uptake onto an established droplet population. This condition is not satisfied within the rapid CCN activation layer at cloud base, where N_d and the condensational sink are still changing quickly. In the quasi-steady approach, dS/dt is set approximately to zero and S_{QSS} is inferred from the balance between the dynamical production term and the condensational sink, requiring the measured updraft velocity (w) and the droplet integral radius/size distribution. The quasi-steady approach is therefore not appropriate during rapid aerosol activation at cloud base (Romps, 2025).

Our diagnostic does not use w , an integral radius, or a droplet-growth equation but it uses the measured T , p , and LWC to construct a finite thermodynamic and energy budget. The advantage is that no explicit condensational-growth closure is required and the result can be tested independently against CCN-chamber activation spectra. This is better explained in the new schematic figure 1 and section 2 as follows:

“Figure 1 summarizes the measurement strategy and the diagnostic thermodynamic framework used in this study. Cloud-base measurements provide N_d , LWC , T , and p while $N_{CCN}(S_v)$ is independently measured below cloud base (Fig. 1a). From the measured cloud-base state, the initial saturated-vapor state is diagnostically reconstructed under the assumptions of an adiabatic and reversible transition treated locally as quasi-isobaric (Fig. 1b). The condensational energy is partitioned according to $Q = \Delta U + W_s$, from which the liquid- and vapor-phase contributions to cloud supersaturation, S_l and S_v , are derived. The resulting cloud-base $N_d(S_v)$ spectrum is then compared with the independently measured sub-cloud $N_{CCN}(S_v)$ spectrum as a closure test of the proposed framework...

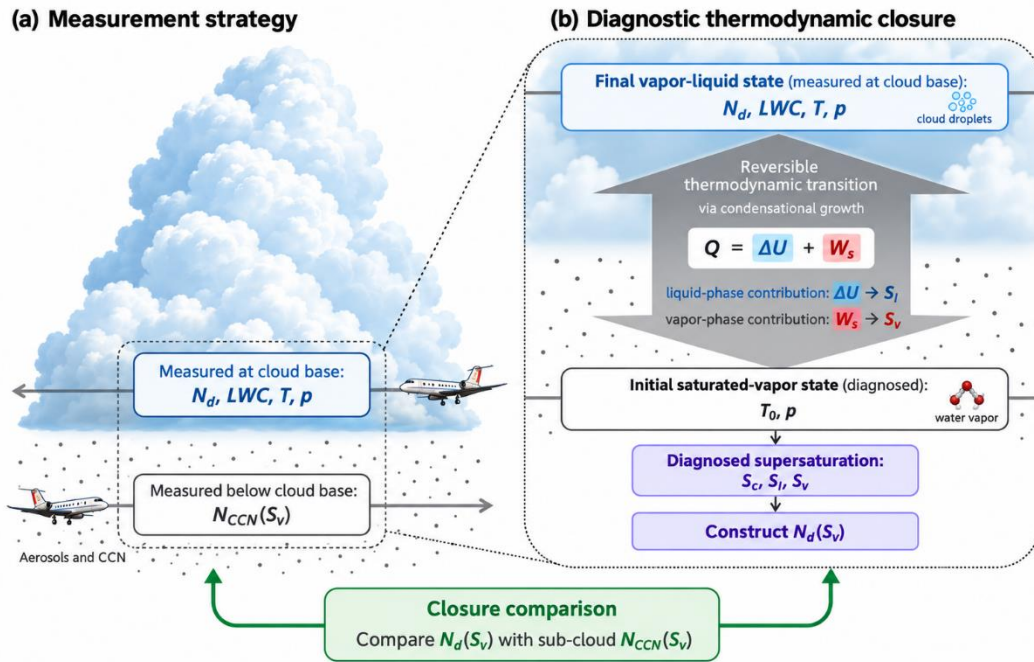


Figure 1. Measurement strategy and diagnostic thermodynamic closure used to derive water vapor supersaturation at cloud base. (a) Schematic of the airborne measurement strategy. At cloud base, the HALO aircraft measures droplet number concentration (N_d), liquid water content (LWC), temperature (T), and pressure (p). Below cloud base, cloud condensation nuclei activation spectra, $N_{CCN}(S_v)$, and pressure are measured. The highlighted cloud-base region indicates the local layer to which the thermodynamic closure is applied. (b) Schematic of the diagnostic thermodynamic framework. The measured cloud-base state is treated as the final vapor-liquid state, characterized by N_d, LWC, T , and p , while the corresponding initial saturated-vapor state, characterized by T_0 and p , is diagnosed. The transition between these states is represented as a reversible thermodynamic transformation during condensational growth. The latent heat (Q) released by condensation is partitioned according to $Q = \Delta U + W_s$, where the internal-energy contribution is associated with the liquid-phase supersaturation S_l and the water-vapor expansion work with the vapor-phase supersaturation S_v . The diagnosed supersaturations S_c, S_l , and S_v are then used to construct the cloud-base $N_d(S_v)$ spectrum, which is compared with the independently measured sub-cloud $N_{CCN}(S_v)$ spectrum as the closure test.”

”

As discussed before in previous reviews, we applied the relevant cloud-base parameterizations and a cloud parcel model to Amazonian clouds in Braga et al. (2017) and Braga et al. (2021). These approaches showed reasonable agreement in reproducing N_d at cloud base (as described in the Introduction section). Although the parcel model reproduces N_d within a few metres of cloud base, typically up to about 20 m above the level where $RH > 100\%$, the adiabatic LWC (LWC_{ad}) generally is comparable to observations only about 50–100 m above cloud base due to the implicit condensational sink in the supersaturation estimate (Eq. 1).

- My second major comment is that the paper would be much stronger if it compared the method and results to other studies. In particular, Romps et al. (2023) estimated supersaturation from the same aircraft campaign but using a different method. How do the results of their work compare with the present study, and if there are differences, then why? In particular, I find it hard to reconcile the results of Fig. 4, which shows “cloud supersaturations” up to 8-9%, with Romps et al. (2023), who found supersaturations that are almost always less than 1%. Why are the results so different? If the “cloud supersaturation” is not the correct quantity to compare with Romps et al. (2023), then what is, and why?

More generally, there are approximately a dozen studies that have used in situ aircraft

observations and the quasi-steady approximation to estimate supersaturation in liquid parts of cumulus clouds (a list is cited in Wall et al. (2026)). Comparing the current results with that literature would be helpful.

The estimated supersaturation from Romps et al. (2023) is the quasi-steady supersaturation (S_{QSS}) is a proxy of the water vapor supersaturation (S_v). The S_{QSS} requires the dynamical forcing and droplet population to vary slowly relative to the supersaturation-adjustment timescale and is therefore not appropriate during rapid aerosol activation at cloud base (Romps, 2025). The present study specifically focuses on this cloud-base thermodynamic transition using an independent finite-state energetic diagnostic. We state that in the new introduction as shown in our previous answer about this topic.

- My last major comment is that I find it hard to believe that supersaturation strongly increases with cloud-droplet number concentration (N_d), as shown in the power-law fits in Fig. 5, because the condensation rate scales with supersaturation times N_d . With all else equal, a larger N_d will increase condensation. This, in turn, would lower supersaturation. The only place that the proposed power-law relationship might make sense (to me) is at the narrow layer (order 10 m) at cloud base where CCN activate, so N_d and supersaturation both change rapidly with height. It would help if the authors can explain and justify the physical interpretation of their results, rather than just fitting a power law to the data.

A: We agree that, once a cloud-droplet population is established, increasing N_d enhances the condensational sink and tends to reduce supersaturation. The positive relationship shown in Fig. 5 is not intended to describe this quasi-steady regime. Rather, our analysis is specifically focused on the cloud-base activation transition, which is also the regime identified by the reviewer in which N_d and supersaturation can increase concurrently. As supersaturation increases during activation, progressively more CCN exceed their critical supersaturation and activate as cloud droplets, resulting in increasing N_d . The use of a power function is therefore physically motivated by the conventional representation of the cumulative CCN activation spectrum, $N_{CCN}(S) = C \cdot S^k$ (Twomey, 1959), rather than being selected solely as an empirical fit to the observations. In the cloud-base activation layer, N_d can consequently be regarded as the integrated response of the available CCN population to the supersaturation attained by the saturated parcel. We have clarified this interpretation in the revised manuscript (described above) and emphasize that the fitted relationship is applicable to the cloud-base activation regime rather than to developed cloud conditions.

Minor Comments

- As far as I know, equivalent temperature is usually denoted with and “e” subscript, but in this paper it has a subscript “Q”. Consider changing to the conventional subscript.

A: We agree that T_e is the conventional symbol for meteorological equivalent temperature.

However, the quantity denoted T_Q in this manuscript is not intended to be the conventional equivalent temperature; it is a diagnostic latent-heat-equivalent temperature defined from $Q/c_{p,cloud}$. We therefore prefer to retain T_Q to avoid implying identity with standard T_e .

- Line 336: I find it confusing to report uncertainties in supersaturation as a percent because supersaturation itself is often reported in units of percent. The way it is written now, I can't tell if the authors are reporting absolute or relative uncertainty. For example, does 10% uncertainty for S_c mean that if one calculates $S_c=1\%$, then the true S_c is $1 \pm 10\%$, or $1 \pm 0.1\%$? The former is so large that it is hard to imagine that it is useful, and the latter seems too good to be true. Because it is so challenging to accurately quantify supersaturation in clouds, it is important that the meaning of the quoted uncertainties is clear. It is also important to compare the uncertainty to uncertainties of other methods

A: We agree and have clarified the notation as follows:

“The relative uncertainties in the energy terms are about 10 % for Q and ΔU , and 13 % for W_s . Propagating these errors yields relative uncertainties of supersaturation (σ_s/S) of approximately 10 % for S_c and S_l , and ~ 12 % for S_v .”

The quoted uncertainties for S_c , S_l , and S_v are relative 1σ uncertainties, rather than absolute percentage-point uncertainties. For example, if $S_v = 1.0\%$ and its relative uncertainty is 12%, the result is $1.0\% \pm 0.12$ percentage points. For comparison, the effective S_v of the CCN measurements used here has an estimated relative uncertainty of about 14%., which we clarified in section 3.7.2 as follows:

“...Furthermore, the estimated relative uncertainty in S_v values (σ_{sv}/S_v) is about 14% on average, resulting from the uncertainties of 10% from counting efficiencies and 10% due to calibration accuracy.”

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