**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 clous-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 ($\Delta U$) 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 $S_c$.
   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 $S_c = S_l + S_v$, 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 $\Delta U$ 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.

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 ~ 1 μm whereas Nd was measured for droplets larger than 1.5 μm, 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.

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.

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.

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.

- 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.
- $\Delta U$, $\Delta U_{ws}$ and $Q$
  In Table 1 and Eqs 18 and 19, $\Delta U_{ws}$ and $\Delta U$ appear to be 2 different variables as $\Delta U_{ws}$ includes $W_s$ and $\Delta U$ does not. But in the caption of Figure 2, the author states "$\Delta U_{ws}$ (indicated as $\Delta U$ for simplicity)". This is very confusing. So which inferred variable is actually shown in Figure 2? Please clarify the difference between $\Delta U_{ws}$ and $\Delta U$.
  Furthermore, if $\Delta U_{ws}$ already includes $W_s$ , then what is the difference between $\Delta U_{ws}$ and $Q$?
- 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?

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.
   - 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.

- 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.
- Supplementary information
    - 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.
    - The simulations in S1is 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.

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.
    - Figure 2: in panels a and b, there are no color legends.
    - Table 1 can be shown much earlier in the paper.
    - 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 …"
    - 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.
    - 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 convectional $S_c$ , 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.
    - Figure 5: what's the difference between CCNa and CCNb? Where were they first defined?

8. 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.

**Minor comments:**

1. Line 16: “We find that …”. Did you really “find” that? Or you derived/speculated/assigned that?
2. Line 29: with respect to “liquid water” or “water vapor”?
3. Lines 38-39: are there more recent studies?
4. Line 41: $N_d$ is first defined here. Please use $N_d$ in the reminder of the paper, no need to state “droplet number concentrations” again, e.g., Lines 262 and 341.
5. Line 45: “the activation” of what?
6. Lines 45-47: “Even though …”. So? What is the issue or challenge here?
7. Line 54: $10^{-3}$ Pa? hPa?
8. Line 61: where is $p_0$ in Figure 1?
9. Line 65: $c_{p,cloud}$ is not defined yet.
10. Line 98: remove “at cloud base”.
11. Line 100: add “cloud” before “relative humidity”, if the authors decide to keep using this name.
12. Line 102: determine“s”.
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…”
14. Line 123: How is this minor uncertainty estimated/calculated?
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”?
16. Please rewrite Lines 143-144, if the authors decide to keep it. This sentence is difficult to follow.
17. Line 150: the specific volume of water vapor is defined as “$\alpha_v$”, but in Eqs 6, 10, and many other instances, it is written as $\propto_v$.
18. Line 162 and Eq 9: it is unclear to me how the authors arrived from Eqs. 1, 4, 5, and 6 to Eq 9.
19. Line 165: by “a reference level”, you mean “the cloud base”?
20. Eq10: again, it is confusing to me how the authors arrived from Eq 9 to Eq 10.
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.
22. Lines 188-189: change to “where temperature gradients typically are smaller than 1K”.
23. Line 189: the calculation“s”, and by “cloud supersaturation” you mean $S_c$?

24. Lines 190-192: please rewrite this sentence.
25. Line 226: please be consistent with the temperature forms.
26. Line 228: how is $T_0$ estimated?
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.
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?
29. Lines 243-244: what does this sentence mean?
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?
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?
32. Line 270: remove "measured". The authors need to be more clearly stating which variables are inferred, which are calculated, and which are measured.
33. Step 2 around Line 270: did the authors calculate/estimate $T_{0ws}$ from a given $W_s$? Or did the authors calculate/estimate $W_s$ from $T_{0ws}$? This is unclear to me. And this step is difficult to follow.
34. Lines 278-279: which is measured by SHARC? Which is derived?
35. Line 283: define CCP.
36. Line 293: cloud base"s".
37. Line 298: remove "water vapor supersaturation".
38. Line 299: "… exposed to a set supersaturation", you mean $S_v$?
39. Line 308: remove "rain water content".
40. Remove Lines 337-340, this is redundant.
41. Line 341: perhaps provide one-sentence justification for this specific case. And remove "droplet number concentrations", you have defined it earlier.
42. Line 344: remove "(in black)".
43. Line 344: is each dot a cloud pass?
44. Lines 345-347: "This variability arises …", again?
45. Line 347: remove "(in red)".
46. Line 352: what do you mean by "cloud elements"? and how were "stronger buoyancy and more vigorous condensational growth" inferred?
47. Lines 353-354: by "cloud supersaturation", you mean $S_c$? or which one?
48. Line 367: what kind of simulations?
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.

50. Line 405: how does the cooling of the cloud parcel reflect the relationship between LWC and Nd?
51. Lines 412-421 can be largely shortened. This part could have been stated in Methods.
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.
53. Line 434: is there really an improved agreement for Flight 17?
54. Line 440: what do you mean by "appears reliable"? and please check the grammar and rewrite this whole sentence.
55. Line 455: which "both cases"?
56. Lines 472-473: were those lapse rates used anywhere in Section 2?
57. Lines 473: "Our results suggest…", which part of results or figure exactly?