Review of manuscript nb. os-2009-62 "Thermodynamic properties of sea air"

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Referee report (anonymous)

December 9, 2009

1 Overall evaluation

The presented paper is of excellent scientific quality. I recommend the editor to accept the paper and to publish it after MINOR (more technical) revision.

2 Evaluation according to OS publishing criteria

1. Does the paper address relevant scientific questions within the scope of OS? Referee (R): Yes.

2. Does the paper present novel concepts, ideas, tools, or data? R: Yes.

3. Are substantial conclusions reached? R: Yes.

4. Are the scientific methods and assumptions valid and clearly outlined? R: Yes. Vide infra for specific comments.

5. Are the results sufficient to support the interpretations and conclusions? R: Yes.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)? R: Yes. Vide infra.

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution? R: Yes.

8. Does the title clearly reflect the contents of the paper? R: Yes.

9. Does the abstract provide a concise and complete summary? R: Yes.

10. Is the overall presentation well structured and clear? R: Yes. Vide infra for specific comments.

11. Is the language fluent and precise? R: Yes.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? R: Yes.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated? R: No.

14. Are the number and quality of references appropriate? R: Yes.

15. Is the amount and quality of supplementary material appropriate? R: Yes.

3 Rationale

At first I would like to apologise to the authors and to the editor for not having been able to come up with my referee report earlier to stimulate the open discussion. But independent of the subject and the method of the paper, the review of a manuscript of 133 pages is a very challenging endeavour within the 8-week window of the Ocean Science format. In the present case I wish to add, that the authors employed very comprehensive and expensive calculus, which is a central part of the paper. Without understanding of the details it is impossible to push a discussion. Just the understanding of these details demanded me some hard days work, which I have spent willingly owing to the large gain of knowledge about this very sophisticated approach.

The authors presented a generalised theoretical approach to determine the thermodynamic equilibrium properties for the seawater−humid air system called "sea air" for applications in meteorology, oceanology, and industry. The notion "seawater" is a generic term inclosing saltwater, freshwater, or ice in mutual thermodynamic equilibrium. The need for this approach is motivated as follows:

- The considered sea air equilibrium is a physical approximation of the air-sea interface, which consists of the ocean surface mixed layer in equilibrium with the marine atmospheric boundary layer. This interface provides a large geographical contribution to the planetary boundary layer.

- Equilibrium properties play a key role in determining the thermodynamic Onsager forces, which ensure a relaxation of thermodynamic non-equilibrium system states to equilibrium ones.

- The multitude of relations for selected quantities is puzzling, relying on different simplifications, frequently without warranty or clear statements with respect to the range of validity, self-consistency etc.

- The salinity is usually not considered in meteorological equations.

The approach follows a straightforward (but non-trivial) concept:

- Take new standard formulations for the thermodynamic potentials of the sea-air system:

    - Seawater: (i) a Gibbs free energy function for seasalt, (ii) a Gibbs free energy function for fluid (i.e., liquid and vaporous) water (derived from a Helmholtz free energy function), (iii) a Gibbs free energy function for hexagonal ice Ih

    - Humid air: (i) a rigorous epression for the Helmholtz free energy of dry air; (ii) a rigorous epression for the Helmholtz free energy of water vapour; (iii) a virial expansion of the Helmholtz free energy function of the humid-air system with consideration of up to three-particle vapour-air interactions.

    This data provide the "phenomenological" input into the approach, which can be updated independent from the subsequent derivations ("building-block concept").

- Take the definitions of the thermodynamic potentials, their differentials, the Maxwell relations, and others from a suitable textbook on thermodynamics.

- Introduce a very sophisticated and self-consistent system of notions and annotations including sub- and superscripts etc.

- Follow a rigorous agenda to extract all possible thermodynamic information about the system under consideration from the thermodynamic potentials.

The outcome of this approach is amazing: a large pool of self-consistent thermodynamic equations for a comprehensive characterisation of the seawater−humid air system in thermodynamic equilibrium. Both the method and the results have textbook character, which alone is a nutritive result. Additionally, for practical application the authors supplemented their theoretical results by a numerical source-code library for easy use on various platforms.

The paper provides sufficient information for proper application by users of the source-code library. However, trying to rederive the complete calculus I realised, that one can easily run into troubled waters. In other words: it was a hard nut to crack it. The authors employed some elegant concepts, such as the use of the latency operator, but presented (understandably) only the final expressions. Despite of the length of the paper (it is not a short one!) the manuscript is written in a very condensed style, reflecting the strong theoretical background of the authors. Because I failt in a complete rederivation of the calculus in a first attempt, I have considered to demand more details and intermediate steps of the derivation. But after getting a hint to the employment of Jacobians for some key relations, I could completely rederive Eqs. (1)-(140), (H1)-(H29), and all expressions given in Tables (14)-(16) on a step-by-step basis. (It sounds curious, but the product of the paper length and its reception time is a constant, i.e., an extension of the manuscript by including intermediate steps would reduce the time for the few to follow the calculus step by step. Even it is not a hard fact to assess the challenge: the execution of the intermediate steps of the calculus in a traceable manner took me 120 handwritten pages (plus a few dozens for the shredder).)

Anyway, without consideration of the required time, the description of the calculations allow their reproduction by fellow scientists (vide infra for some specific comments). I want to appreciate the elegancy of the whole approach.

4 Specific comments

Below, I want to give some specific minor comments, mostly technical ones, in the hope they were found to be helpful. It is not necessary for the authors to reply step-by-step. The technical recommendations do not need any reply, the comments only inasmuch as my statements are wrong. The authors can focus on the few questions.

4.1 Manuscript text

1. P. 2197, line 4: The authors distinguished between fluid water and seawater. According to p. 2205, lines 13-14, fluid water is used as a generic term for liquid and gaseous water (water vapour). Thus, seawater is also fluid water. Please check notions.

2. P. 2198, line 2: The authors wrote, that the bulk formulas for the ocean-atmosphere latent heat flux are expressed in terms of the "sea-air specific humidity" rather than of saturated humid air (ref. to King-Hele, 1990). In the context of the previous discussed Onsager forces it is more correct to say "equilibrium specific humidity of sea-air" (as done on p. 2218, line 21).

3. P. 2200, line 12: The symbol P without subscript is the absolute pressure (cf. p. 2205, line 19). In meteorological texts the symbol e is used to denote the (partial) water vapour pressure, but not the total pressure.

4. P. 2200, lines 20-23: The careful distinction between "dry adiabatic", "moist adiabatic", "wet adiabatic", and "ice adiabatic" is very helpful and avoids notional confusion from the beginning. The same is true for notions on p. 2215, lines 4-6.

5. Section 2: The review is comprehensive and good with respect to content. However, the internal itemisation should follow the order in the caption: dry air, humid air, water, seawater, ice (rearrangement of paragraphs).

6. P. 2203, line 4: Millero [...] provides an equilibrium vapour pressure formula for seawater.

7. P. 2204, line 9: Be more specific, which "properties of marine aerosol" are meant in the present context. Write "by O'Dowd et al. ...".

8. P. 2205, line 6: Do you mean "high-speed applications" in the sense of "real-time applications" such as in numerical weather predictions models etc.?

9. P. 2205, lines 12-13: "[...] describing the salinity correction to pure water in combination with a Helmholtz potential for fluid [...]water" (instead of "to be combined", maybe).

10. P. 2205, line 12: I recommend to introduce the thermodynamic potentials already here as (mass) specific values [J/kg].

11. P. 2206, line 15: Just to know: Is the enthalpy of a sample seawater, $H_{\mathrm{w}}^{\mathrm{SW}}$, a measurable field quantity?

12. P. 2208-2209, Eqs. (9), (10): Considering Appendix D it is more plausible to start with Eq. (10) with reference to Eq. (D14) and to arrive at Eq. (9).

13. P. 2209, line 10: Fig. 1 depicts the compressibility factor before it has been introduced (cf. Eq.(11)).

14. P. 2209, line 15: Considering that the water vapour with the mass fraction q is the minor fraction in humid air, the employment of the mass fraction of dry air A instead of $q = 1 - A$ needs getting used to (at least with respect to atmospheric applications.) I mean, the property q corresponds better to salinity S_{A}. Is the use of A motivated by technical applications, as mentioned in line 24?

15. P. 2212, Eqs. (21)-(23): Maybe it is helpful to give a reference to the Gibbs-Duhem relation (Eq. (21)) and the Duhem-Margules relation to arrive at Eqs. (22), (23).

16. Potential temperature, Eq. (29): In meteorological textbooks, the temperature-pressure relation for an adiabatic process reads (cf. Iribarne and Godson 1973, Eq. (54) therein):

$$\frac{T_0}{T} = \left(\frac{P_0}{P}\right)^{\kappa} , \quad \kappa = R/c_p .\tag{1}$$

If we choose P_0 = 1000 hPa, T_0 becomes by definition, the potential temperature θ:

$$\theta = T \left(\frac{P_0}{P}\right)^{\kappa} .\tag{2}$$

This formulation is equivalent to Eq. (29), apart from the meaning of T_0. To ensure formal consistency with meteorological interpretation maybe it is better to write (cf. Eq. (28)):

$$\begin{aligned} s^{\mathrm{AV}}\left(A, \theta^{\mathrm{AV}}, P_0\right) &= s^{\mathrm{AV}}\left(A, T, P\right) , \\ \theta^{\mathrm{AV}} &= T_0 + \Delta\theta^{\mathrm{AV}} , \\ T_0 &= 273.15\ \mathrm{K} . \end{aligned}\tag{3}$$

The same is valid for Eqs. (29), (30), (79), (81). Maybe it is sufficient to insert a footnote to retain author's annotation.

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17. P. 2215, Eq. (34): Please refer here to Tab. 15.

18. P. 2216, line 9: The reference to Eq. (20) for the pressure is not clear to me. I would have expected reference to Eqs. (2) or (15).

19. P. 2216, Eq. (39), line 19 (first appearance): Just a comment: The use of subscript v to denote the gas constant of water vapour would better fit into the system of annotations.

20. P. 2217, Eq. (40): This equation has already been introduced by Eq. (1). Instead of Eq. (40), for a later use the authors could have introduced here a representation of the Gibbs free energy of seawater via the Gibbs-Duhem and Duhem-Margules relations in terms of the chemical potentials of the solute and solvent, similar to Eqs. (21)-(23) for humid air.

21. P. 2219, line 1: The definition of a phase-transition latency operator $\Lambda_{\mathrm{AS}}[z]$ is a powerful concept, allowing for a very compact codification of the thermodynamic information. As employed in the calculus, one could add $\Lambda_{\mathrm{AS}}[-z] = -\Lambda_{\mathrm{AS}}[z]$. I am curious about the root of the notion "latency" operator.

22. P. 2219, 2220, Eqs. (47), (48): Are the chemical coefficients D_{A} and D_{S} observable properties or do some empirical proxies exist?

23. P. 2222, line 14: Using Eqs. (52), (54), and (44) [...]

24. P. 2222, line 16: Last term of Eq. (61): "latent part" or "latent heat part" of the heat capacity? The same on p. 2238, line 3.

25. P. 2223, line 1: I recommend to consistently use displaystyle within a line, e.g.: [...] isothermal compressibility $\kappa_{\mathrm{T}}^{\mathrm{SA}} = -(v^{\mathrm{SA}})^{-1} \left(\partial v^{\mathrm{SA}} / \partial P \right)_{w^{\mathrm{A}}, w^{\mathrm{S}}, T}$ (appearing several times throughout the manuscript).

26. P. 2223, Eq. (64): Please add reference to Eq. (33) for derivation.

27. P. 2223, line 11: From Eqs. (57), (59), (60), (61), and (63) we get [...]

28. P. 2224, Eq. (67): I arrive at this equation only by employing Eq. (43). Maybe, add this reference.

29. P. 2224, line 20-23: The authors defined "wet air" (cloudy air) as saturated humid air combined with liquid water. Thus, I guess the generic notion "saturated humid air" is sufficient in the caption.

30. P. 2226, lines 1-2: Please refer here to Fig. 8 showing $A_{\mathrm{AW}}^{\mathrm{sat}}$.

31. p. 2226, line 8, and p. 2231, lines 11-17: Just a comment: In meteorological applications, the pressure coordinate is frequently replaced by the geometrical height above ground or sea level. The vertical level corresponding to the isentropic condensation level (ICL) is the lifting condensation level (LCL). This will give the base of cumulus clouds, provided that these are actually formed by air adiabatically rising from any level z_0 with the dewpoint temperature T_{d}. Typically representatives are orographic clouds, formed by forced airmass lifting on the windward side of mountains (as one component of the Foehn phenomenon). Therefrom different is the level of free convection (LFC), which is the condensation level, that a buoyant, adiabatically rising air parcel exceeds due to a local temperature excess over the ambient temperature at any initial level.

32. P. 2226, Eq. (73): The Clausius-Clapeyron differential equation can be derived as a special case of Eq. (46). Maybe, add reference to this equation.

33. P. 2227, Eqs. (74), (75): I arrived at this equation by means of Eqs. (42), (44), (51), (54), and (55). Maybe, add reference to these equations.

34. P. 2228, line 3: The entropy according to Eq. (77) [...]

35. P. 2228, line 3: Just a comment: In meteorological textbooks, the coordinates of the "tephigram" are usually $\ln\theta$ (ordinate) and T (abscissa). The name of the diagram goes back to Shaw and refers to the letters T and ϕ, the latter as a symbol of entropy. As the ordinate variable $\ln\theta$ is proportional to the specific entropy, the tephigram can also be considered as having coordinates s, T. The convention to mention first the abscissa ("te") and than the ordinate ("phi") corresponds to general use in mathematics (Iribarne and Godson 1973, cf. p. 80 therein).

36. P. 2232, lines 22 - p. 2233, line 26: The discussion of the transitive properties of multiphase equilibria is very helpful and clearly explained.

37. P. 2237, Eq. (113) and p. 2238, lines 1-11: The derivation of Eq. (113) is elegant. The authors mentioned, it is "difficult" to find in the literature a thermodynamically rigorous definition of the latent heat that occurs in conjunction with the exchange of water between seawater, humid air, and ice. I am not aware of any. In cases there is any reference, please add it.

The last term on the right-hand side of Eq. (113) representing the latent heat contribution reads:

$$c_p^{\mathrm{SIA}}\big|_{\mathrm{lat}} = T \left(\frac{\partial s^{\mathrm{SIA}}}{\partial T} \right)_{\mathrm{lat}} .$$

The isobaric latent heat of ice is by definition the ratio of the latent heat part of the heat capacity divided by the isobaric melting rate:

$$L_p^{\mathrm{SIA}} = \frac{T \left(\partial s^{\mathrm{SIA}}/\partial T \right)_{p,\mathrm{lat}}}{\left(\partial w^{\mathrm{Ih}}/\partial T \right)_{w^{\mathrm{A}},w^{\mathrm{S}},p}} = -T \left(\frac{\partial s^{\mathrm{SIA}}}{\partial w^{\mathrm{Ih}}} \right)_{\mathrm{lat}} .$$

This allows the following interpretation:

$$\Delta s^{\mathrm{SIA}} \approx -\frac{L_p^{\mathrm{SIA}}}{T} \Delta w^{\mathrm{Ih}} .$$

Thus, an increase of the ice mass fraction in the equilibrium system, $\Delta w^{\mathrm{Ih}} > 0$, is accompanied with corresponding decrease of the entropy, $\Delta s^{\mathrm{SIA}} < 0$, whereas the effect increases with decreasing temperature.

38. P. 2244, lines 13-14: [...] that humid air in equilibrium with seawater above its freezing temperature is always subsaturated with respect to pure bulk water (or freshwater).

39. P. 2245, line 10 - p. 2246, line 3: Just a comment: Considering a layer of the atmosphere between two constant-pressure surfaces (planes) with constant total mass, assuming that layer being in hydrostatic equilibrium, and supposing, that the layer is isolated from its surrounding, neither heated nor cooled by radiation nor by interaction with adjacent air (or ground), Bohren and Albrecht (1998, cf. pp. 164-169 therein) showed, that of all linear potential temperature profiles, **a constant potential temperature maximises the entropy of that isolated atmospheric layer**. The entropy maximisation requires the equilibrium temperature of the isolated layer to decrease with height at the dry adiabatic lapse rate, $\Gamma_{\mathrm{dry}} = -(\mathrm{d}T/\mathrm{d}z)_{\mathrm{adiab}} = g/c_p$, i.e., **the equilibrium temperature profile is non-isothermal**. In contrast to an isolated solid, in which all temperature gradients are eliminated by conductive energy transport without mass transport, in the atmosphere the equilibration primarily performs by convective energy transport accompanied by mass transport (diffusive heat conduction can be neglected). The adjustment of a dry adiabatic lapse rate is a direct consequence of dry convection in the atmosphere (parcels moving from one level to another and

mixing with its environment). Only in the absence of gravity the dry adiabatic lapse rate $\Gamma_{\mathrm{dry}} = -(\mathrm{d}T/\mathrm{d}z)_{\mathrm{adiab}} = g/c_p$ would be zero, i. e.,

$$\lim_{g \to 0} \Gamma_{\mathrm{dry}} = \lim_{g \to 0} \frac{g}{c_p} = 0 \ ,$$

and the equilibrium temperature isothermal. However, there is no contradiction to the author's statement, that in an external gravity field the equilibrium state is characterised by vertically constant temperature:

- The entropy maximisation in Bohren's and Albrecht's model refers to an equilibrium state of an isolated model system, in which the maximum of entropy corresponds to an "intrinsic" thermodynamic equilibrium state. The real atmosphere is characterised by the existence of inhomogeneous boundary conditions (e. g., between the bottom and the top of atmosphere), which enforce permanent forces and accompanied fluxes (e. g., according to Fourier's and Fick's laws), leading to a permanent non-equilibrium state. Thus, any vertical temperature gradient − both an adiabatic as well as a diabatic one − is associated with a vertical heat flux, regardless if accompanied by a mass flux or not. A stationary non-equilibrium state refers to a "dynamic or steady-state equilibrium", which is a cumbersome notion inasmuch as it denotes a state, which is in reality a non-equilibrium one with non-zero Onsager fluxes.

- Also in the case of a well-mixed layer with constant potential temperature, there are large energy transports: Parcels close to the surface can be heated and thus be warmer than air near the bottom of the mixed layer. As these positively buoyant parcels rise, they cool at the dry adiabatic rate. But because temperature in the mixed layer also decreases at this rate, the rising parcels remain warmer than the mixed layer. Thus as they mix with surrounding air, they increase the temperature of the mixed layer even though there is no gradient of θ (Bohren and Albrecht 1998, cf. p. 168 therein).

- In Bohren's and Albrecht's model an isothermal equilibrium temperature profile can only be adjusted in the limiting case of a vanishing gravity field, supposed that dry convection (with air parcel mixing) remains the primary transport process. (However, in such a fictive state also the vertical pressure gradient in the hydrostatic equilibrium

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would vanish and the atmosphere would escape, thus the model losing its physical meaning.) In the real atmosphere, an isothermal equilibrium temperature profile can be adjusted also in the presence of a gravity field, if the external energy source will be switched off and the system relaxes to an equilibrium state by heat conduction according to Fourier's law. In such a case the pressure and density profile obey the hydrostatic equilibrium and the ideal gas law at T(z)=const.

40. P. 2247, line 12: I recommend to include the reference to Fig. 19 right here, when the authors describe RS 37 (cf. also lines 19-21). Just a comment: The entropy computed from Eq. (24) in conjunction with the the related sea-air entropy, computed from ocean surface properties by means of the equilibrium Eq. (43), seems to be a good synoptic airmass indicator. As seen from the top panel in Fig. 19, the deviation between both measures reflect a disturbance of the seawater−humid-air equilibrium, caused by non-seasalt debrises, here Saharan dust. This opens the way to quantify the deviation from the equilibrium in terms of aerosol load and of different proxies, such as aerosol optical depth derived from spaceborne platforms or ground-based remote sensing (Lidar) etc. It would be interesting to see, how the deviation from the equilibrium is correlated with several proxies of contamination. I consider such an approach as an interesting supplementary method for the desert dust community.

41. P. 2248, line 12: Can the increasing upper-ocean salinity overcompensate the desalination, resulting from the increasing freshwater flux from melting ice? Are the net changes relevant for a displacement of the seawater−humid air equilibrium (via the Raoult effect, cf. Fig. 10) on a global scale or over geological time scales (impact on cloud formation)?

4.2 Tables

1. The tables are cited in the text and in the appendix in the following order (first citation): 2-3-1-(7...9)-6-(10...17). I recommend to numerate the tables according to the order of their citation in the text. Please cite explicitly also Tables 4-6.

2. Table 1: Please enclose all units in brackets. What is the salinity property SSS standing for and how it is related to salinity S_A employed in the

manuscript? Maybe it is useful to include SSS and the unit "psu" (practical salinity units) in the abbreviation list.

3. Appendix G, Tables 14-16: Please add the references to Guggenheim (1950, cf. §3.08 therein) and Shaw (1935) to rapidly and elegantly derive the thermodynamic identities employing Jacobians.

 - Table 14: Use displaystyle in the middle column.
 - Table 15, line 2: Please correct typo: $g = f + pv$.
 - Table 15, line 9: Please check sign. I arrived at $g_{PP} = -\kappa_T/\rho$. (cf. also Feistel/Wagner (2006, J. Phys. Chem. Ref. Data, Vol. 35, No. 2, pp. 1021-1047, Table 3), where it is correct.)
 - Table 15, line 10: Please check sign. I arrived at $g_{TP} = +\alpha/\rho$. (cf. also Feistel/Wagner (2006, J. Phys. Chem. Ref. Data, Vol. 35, No. 2, pp. 1021-1047, Table 3), where it is correct.)
 - Table 15: Use displaystyle in the middle column.
 - Table 16: Use displaystyle in the second and third column. I cannot resolve the annotations in lines 6-8 of the third column.

4.3 Figures

1. The figures are cited in the text in the following order (first citation): 1(2209) - 14(2209) - 3(2209) - 13(2209) - 21(2210) - 2(2215) - 4(2222) - 5(2224) - 6(2228) - 8(2228) - 7(2230) - 16(2231) - 9(2231) - 10(2235) - 11(2239) - 17(2240) - 14(2242) - 19(2247) - 20(2247) - 18(2250). I did not found a citation of Figs. 12, 15, 21. I understand, that this "misorder" is a result of cross-referencing throughout the paper, e.g., to antedate something, which will be explained later. Anyway, the authors should check, whether a numeration of the figures according to the order of their citation in the text is possible and better here.

2. Figure 1: When this figure is cited, the compressibility factor is not yet defined. Please add a reference to the equation.

3. The sizes of the "twin" figures 1, 3, 4, 6, 9-12, 19, 20 are too small and should be enlarged to the format size of, e.g., Fig. 2.

4. Figure 8 should be cited on p. 2226, line 2.

5. Figure 16: Please add in the legend, that the quasi-vertical thin lines refer to the adiabatic lapse rate Γ^{SA} according to Eqs. (64), (65). (To get a clue, these lines can be obtained by employment of the dry adiabatic lapse rate of $\Gamma_{\mathrm{dry}} = -(\mathrm{d}T/\mathrm{d}z)_{\mathrm{adiab}} = g/c_p \approx 1\,\mathrm{K}/100\,\mathrm{m}$ and the barometric scale factor $\partial z/\partial p \approx -8\,\mathrm{m}/\mathrm{hPa}$.)

6. Figure 17: Can the difference between the IML and IFL denoted as thermal hysteresis? Please remove once "near very" in the legend.

7. Figure 20: Please add in the legend, that the abscissa A refers to the water vapour mass fraction or specific humidity via $q = 1 - A$.

5 Final remarks

The work is well done, its outcome is very impressive! Apart from its high practical relevance it is a very instructive teaching material for education at universities and technical colleges. The authors should consider to come up with a textbook version, in which also more intermediate steps are presented. I congratulate to this manuscript.

References

Bohren, C. F. and Albrecht, B. A.: Atmospheric Thermodynamics, Oxford University Press, New York, 1998.

Guggenheim, E. A.: Thermodynamics. An Advanced Treatment For Chemists And Physicists, North-Holland Publishing Company Amsterdam, 1950.

Iribarne, J. V. and Godson, W. L.: Atmospheric Thermodynamics, vol. 6 of *Geophysics and Astrophysics Monographs*, D. Reidel Publishing Company, 1973.

Shaw, A. N.: The derivation of thermodynamic relations for a simple system, Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences, 234, 740, 299–328, 1935.