The Nucleus in and the Growth of Hygroscopic Droplets

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- Experimental methods
- Physical evolution of droplet growth and the “Köhler curve”.
- Particle number, condensation, and coalescence
- Conclusions
- Discussion

Collection of Hoarfrost to measure [Cl] in fog/clouds

- Roughly 2 kg of hoarfrost was collected off of three different mountain ranges over a 4 year period and the [Cl] was measured for each.
- Köhler also performed droplet size measurements on the fogs that produced the frost.

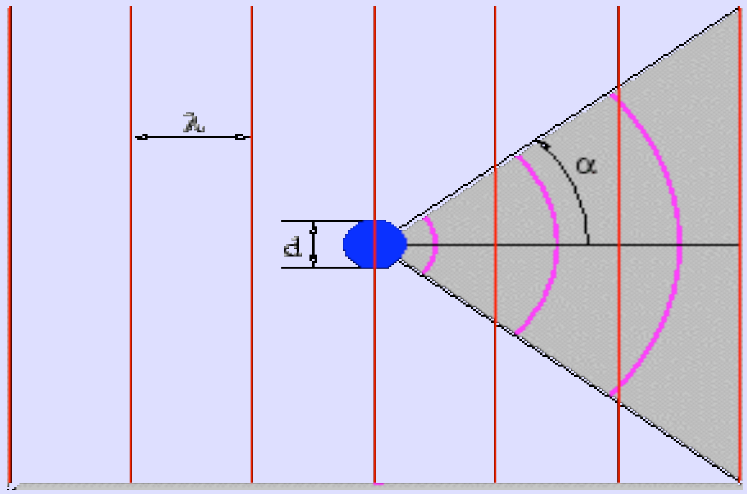
- **[Cl] was typically around 3.5mgm, although was found to vary over several orders of magnitude.**

- **[Cl] may be expressed as: C = 3.595 X 2^p.**

- **[Cl] measured in rain water (H. Isreal 1934- 3.42 X 2^p) were similar to those obtained by Köhler in clouds and fogs at altitudes around 900 meters. (Kinch 1870-86 3.81 and 3.36 mgm/l)**

- **With increasing altitude (Sonnblick and Zugspitze) Köhler observed that [Cl] decreased. (important later)**

Droplet size Measurements



r _ _/sin(_)

_ = 0.571 microns (white light)

Corona of lichen spores measured in lab(?) gives standard deviation calibration for deviation of particle size from the average.

Homogeneous droplet sizes produced sharper corona.

Experimental data tended to be based on measurements made in clouds where certain sizes dominated.

- **Average droplet sizes tended to change during a measurement period, however frequency plots showed certain maxima at specific radii. The relationship between those radii is: $r = B*2^{n/3}$ microns, where *n* is a whole negative or positive #.**

- **After calculating *n* for all series of measurements, frequency curves for *n* revealed a single maximum for whole *n*.**

- **A second maxima shifted by *n* + 0.426 was called group 2, which accounted for about 40% of the measurements.**

- **Köhler uses *n* as a proxy (log normal) for droplet radii throughout the remainder of the paper.**

The "Köhler" Curve

- Plot/Axes
- 2 regimes
  - Droplet < critical radius/concentration
  - Droplet > …
- Implications
  - Largest first
  - It's a droplet eat droplet world….

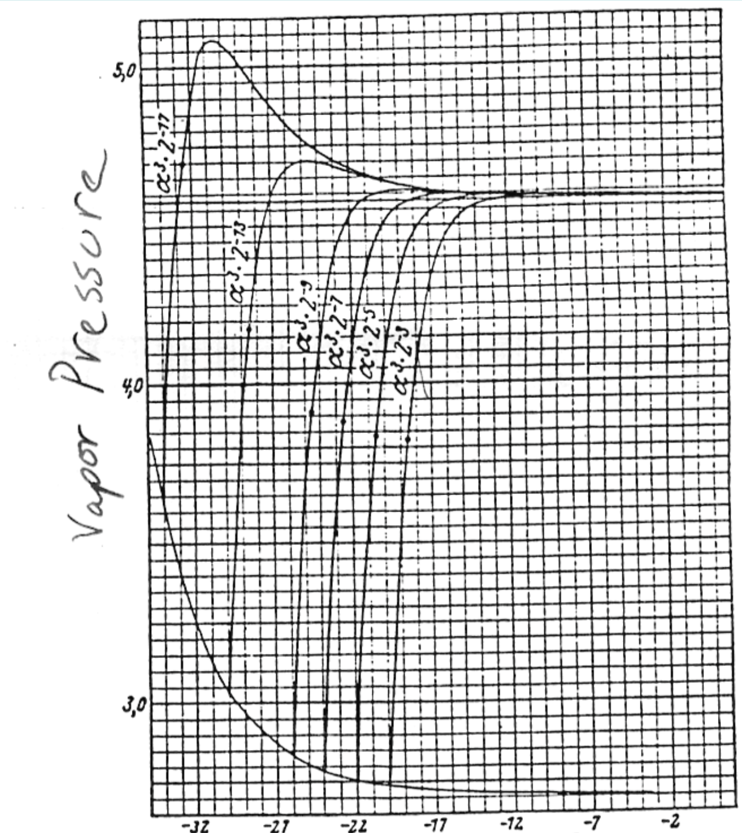


Fig. 1.

The “Köhler” Curve (cont’d)

Plot/Axes

- (assumed) Bottom is curve showing first barrier to condensation – curvature effect
- Current figures have x = radius of droplet instead of [Cl], not clear what units y is in, but 100% saturation is clear
- CCN radius is expressed as mass of nucleus instead of diameter

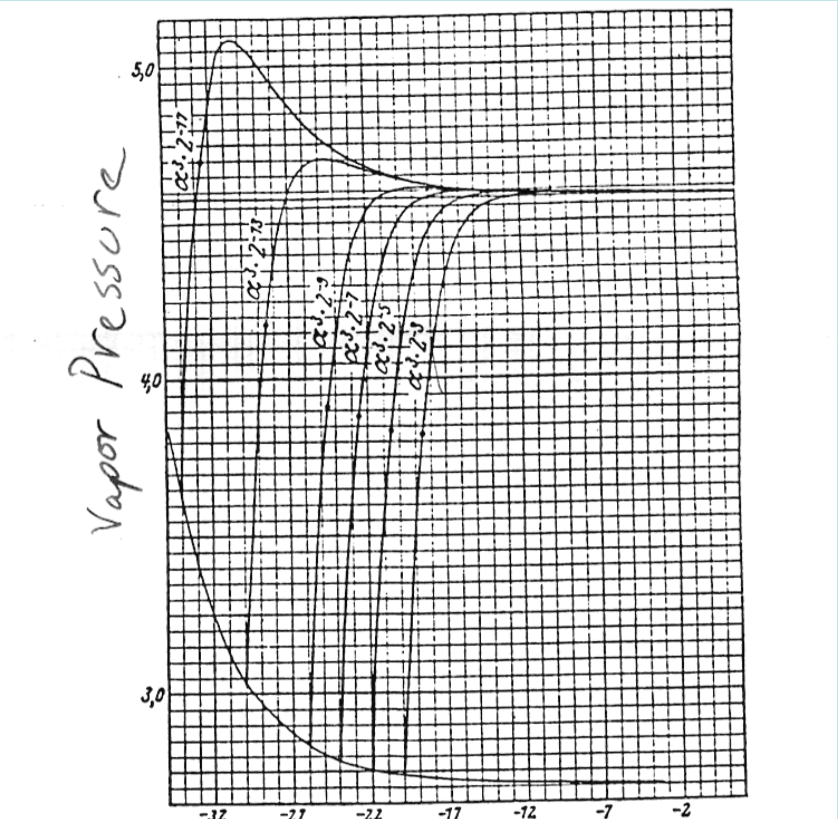


Fig. 1.

The "Köhler" Curve (cont'd)

"without detailing the calculation…"

2 regimes:

- Surface vapor pressure goes as $1-b/r^3$ due to solution effects (Rault's law) where salt lowers the surface tension of water
- Surface vapor goes as $\exp(a/r)$, due to curvature effect from surface tension
- Critical supersaturation where droplet can continue to grow

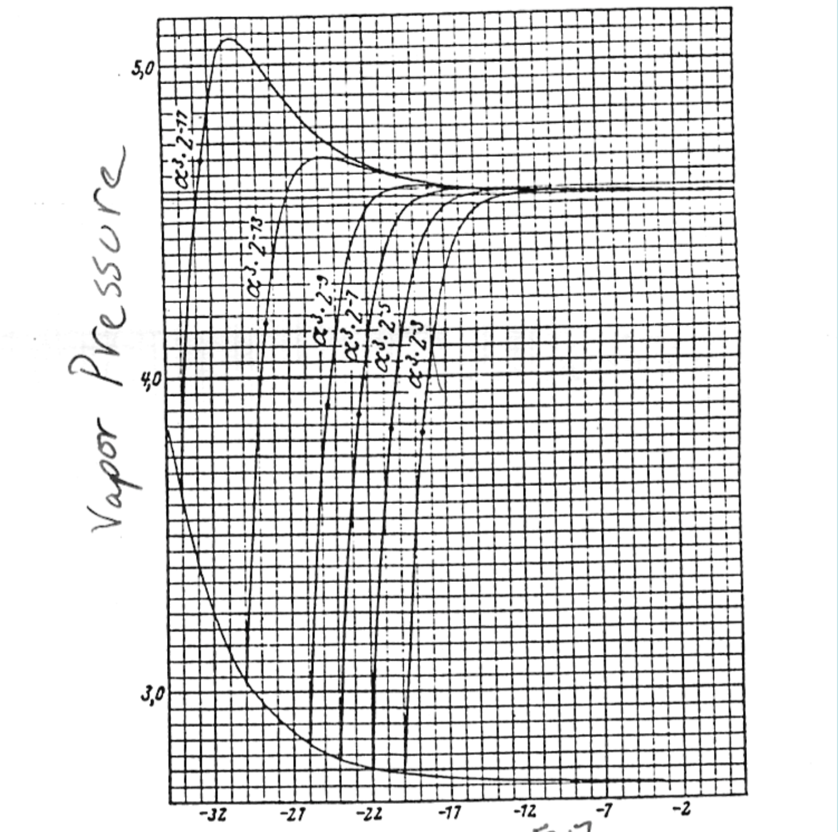
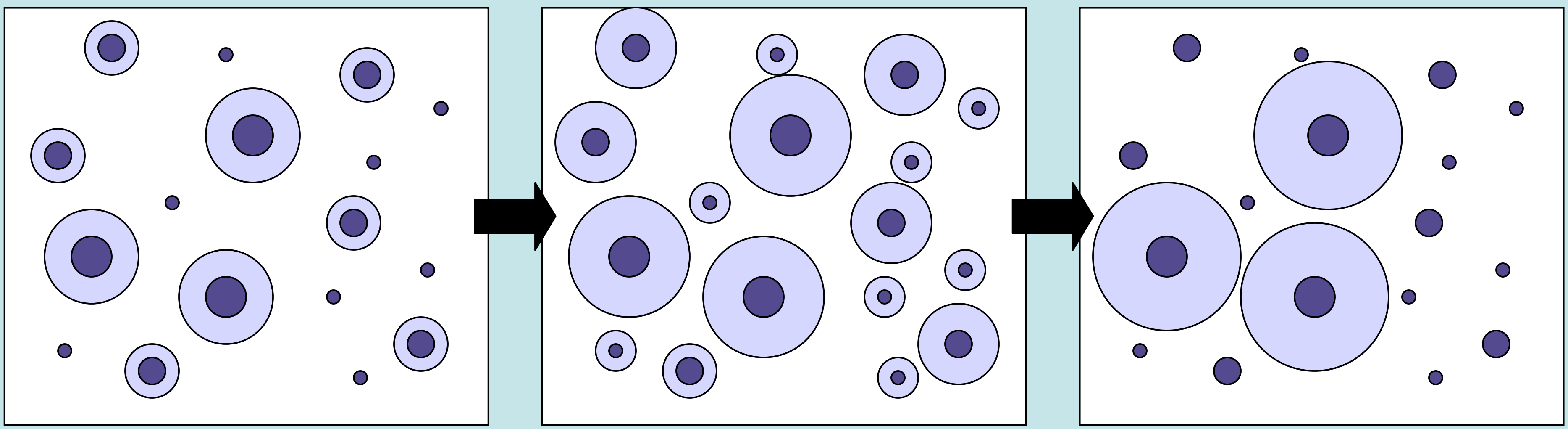


Fig. 1.

The "Köhler" Curve (cont'd)

Implications:

- As air becomes saturated, large drops deliquesce first (curvature effects)
- As pass through saturation some drops pass barrier some do not…
- As vapor continues to condense to reach equilibrium vapor pressure, drops that did not pass the radius ("concentration") barrier will shrink (evaporate) to a smaller size allowing larger particles to continue to grow…



(all deliquesced …)

Particle number condensation and coalescence

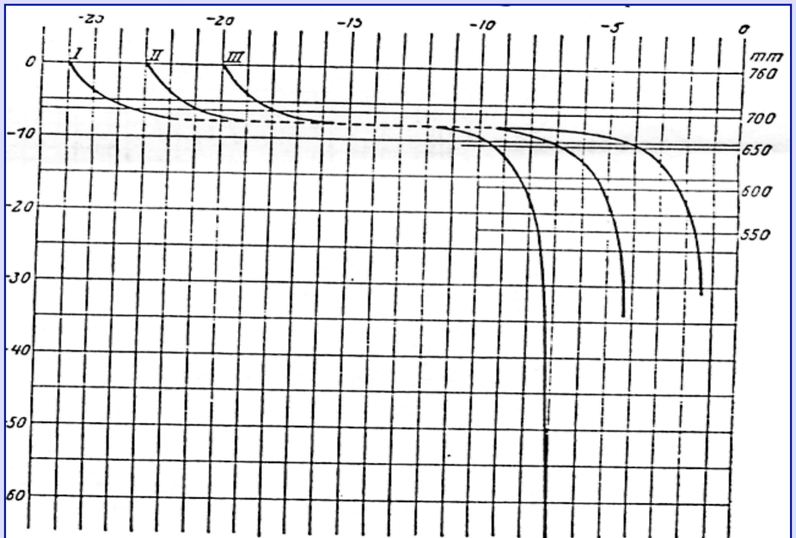


FIG. 2.

I : $a^3 = a_1{}^3 \cdot 2^{-9}$ $N = 10^5 \cdot 2^{10}$ per gm. of air
II : $a^3 = a_1{}^3 \cdot 2^{-6}$ $N = 10^5 \cdot 2^{7}$,, ,, ,, ,,
III : $a^3 = a_1{}^3 \cdot 2^{-3}$ $N = 10^5 \cdot 2^{4}$,, ,, ,, ,,

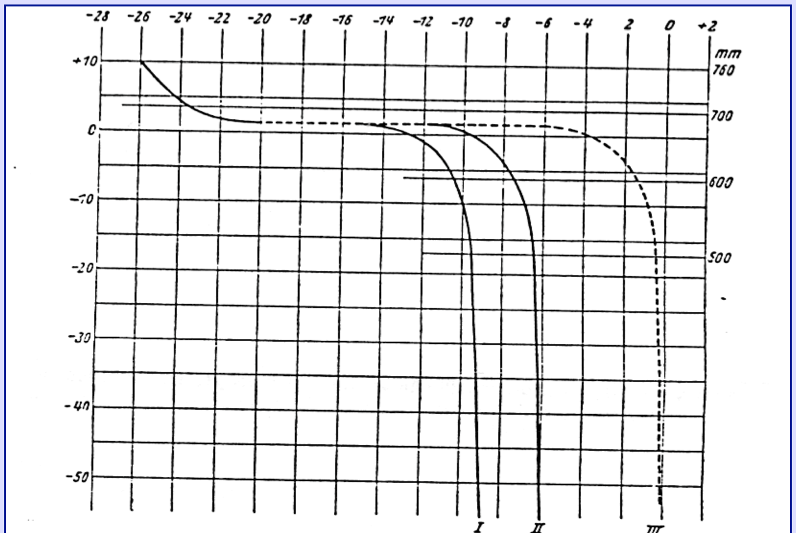


FIG. 3.

$a^3 = a_1{}^3 \cdot 2^{-9}$

I : $N = 8 \cdot 10^8$ II : $N = 10^8$ III : $N = 10^8 \cdot 2^{-6}$ per gm. of air.

- ***n*** **changes as a function of** ***p*** **(concentration) and the plot assumes constant (*p-n*) = 17**
- **Asymptote toward a specific size or the "last whole *n*".**
- **The greater N is, the smaller "LW*n*" value ie.. Average drop radius is smaller after equilibrium has been reached.**
- **This illustrates the limits on droplet growth by condensation due to available water vapor.**

C = 3.595 X 2^p *mg/L, so* for $\_^3 = \_1 \times 2^{-p}$ *than* $N = N_1 \times 2^p$ and smaller nuclei mass implies greater particle #.

(RHS – atm. pressure in mmHg); LHS – (T-273); Hor. – *n (radius)*

TABLE II.

N $(\times 10^5)$	$\alpha^3 = \alpha_1^3 . 2^{-9}$. The last whole n.	Concentration of Cl. (gm./l.).	Final Temperature.	$\alpha^3 = \alpha_1^3 . 2^{-6}$. The last whole n.	Concentration of Cl. (gm./l.).	Final Temperature.	$\alpha^3 = \alpha_1^3 . 2^{-3}$. The last whole n.	Concentration of Cl. (gm./l.).	Final Temperature.	$\alpha^3 = \alpha_1^3$. The last whole n.	Concentration of Cl. (gm./l.).	Final Temperature.
2										+1	1·798	253·7
8										−1	7·190	253·7
16							−2	1·798	253·9	−2	14·38	253·7
64							−4	7·190	253·9			
128				−5	1·798	254·2	−5	14·38	253·9			
512				−7	7·190	254·2						
1024	−7	0·899	211·0	−8	14·38	254·2						
4096	−9	3·595	211·0									
8192	−10	7·190	211·0									

- **Droplet sized achieved by condensation is independent of _^{3}, and appears to depend only on N, although [Cl] decreases with _^{3}.**
- **Larger N leads to smaller droplets, with higher [Cl] concentrations. (Twomey effect?)**
- **Smallest _^{3} must reach greater heights and [Cl] decreases. (mechanism previously explained by Jill?)**

- **The LWn reached is orders of magnitudes smaller than fog, cloud or rain drops, and since rain and cloud drops have the same [Cl], raindrops and cloud drops can not form by condensation.**

- **Evidence of this - Aitkens' nucleus counter obtains a much higher particle number than droplet # in clouds and fogs, so Kohler infers that droplets must coalesce to form cloud, fog and rain drops.**

- **Hypothesize that [Cl] is independent of drop size.**

Conclusions

- Increasing N (CCN) reduces the droplet size, to very small sizes indeed!
- The hoar frost concentrations reflect this last whole *n*, explaining the constant [Cl] for a variety of particle sizes/altitudes(Haldde?)
- Because of large N, droplets continue to grow to high altitudes (Zugspitze, Sonnblick), before reaching last whole *n*, thus explaining the decreasing [Cl]
- For certain values of a given N, different sized CCN can still give same sized droplets (last whole n)
- The last whole n concept shows that rain sized drops cannot grow to rain drops through growth, as does the observation from the Haldde (same [Cl] for rain and fog)
- Also can explain the decreasing [Cl] with altitude by the size selection due to the growth barrier.

Discussion

- What do we know now? (Twomey, Seinfeld, Nenes, Conant…)
- Was this whole study done on FROST and frozen droplets (the analysis for water was done in the late 19^{th} century)
- Unclear on some of the procedures.
- Would this paper be published today as it is presented?
- Data??? Are the curves from data?
- Assumptions? Cl is internally mixed… all droplets have the same hygroscopic properties?
- How does a heterogeneous population of CCN change the results?
- Errors?