

Cloud microphysics

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Overview of cloud physics lecture

- Atmospheric thermodynamics
 - gas laws, hydrostatic equation
 - 1st law of thermodynamics
 - moisture parameters
 - adiabatic / pseudoadiabatic processes
 - stability criteria / cloud formation
- Microphysics of warm clouds
 - nucleation of water vapor by condensation
 - growth of cloud droplets in warm clouds (condensation, fall speed of droplets, collection, coalescence)
 - formation of rain
- Microphysics of cold clouds
 - homogeneous nucleation
 - heterogeneous nucleation
 - contact nucleation
 - crystal growth (from water phase, riming, aggregation)
 - formation of precipitation
- Observation of cloud microphysical properties
- Parameterization of clouds in climate and NWP models

Phase transitions

vapor	↔ liquid	condensation, evaporation
liquid	↔ solid	freezing, melting
vapor	↔ solid	deposition, sublimation

Changes from left to right:

- ⇒ increasing molecular order, “free energy barrier” to overcome
- ⇒ cloud forming processes

saturation = equilibrium condition for thermodynamic system consisting of vapor (ice) and liquid

Saturation vapor pressure

Clausius-Clapayron equation

$$\frac{de_s}{dT} = \frac{L_v}{T(\alpha_v - \alpha_l)}$$

- integration yields $e_s(T)$, approximate because L_v depends on T

Magnus formula (empirical)

$$\text{water}(0^\circ\text{C} - 100^\circ\text{C}) : e_s = 6.1078 \exp\left(\frac{17.0809T}{234.175+T}\right)$$

$$\text{subcooled water} : e_s = 6.1078 \exp\left(\frac{17.8436T}{245.425+T}\right)$$

$$\text{ice} : (-50^\circ\text{C} - 0^\circ\text{C}) : e_s = 6.1071 \exp\left(\frac{22.4429T}{272.44+T}\right)$$

T in °C and e_s in hPa.

Why do droplets form?

- at equilibrium (saturation):
rate of condensation = rate of evaporation
- **energy barrier** of small droplets: generally no phase transition at saturation (**homogeneous nucleation** unlikely)
- when air parcels ascent without condensation \Rightarrow supersaturation
- energy barrier may be decreased by **cloud condensation nuclei**
 \Rightarrow **heterogeneous nucleation**
 - hygroscopic particles serve as centers of condensation
 - supersaturation in clouds not much larger than 1%
- when air parcel including cloud droplets ascend to $T < 0^\circ$
 - droplets become supercooled
 - freeze when ice nuclei are present

Energy difference due to formation of droplet

ΔE = surface energy of droplet - Gibbs free energy due to condensation

$$\Delta E = 4\pi R^2 \sigma - \frac{4}{3}\pi R^3 nkT \ln \frac{e}{e_s}$$

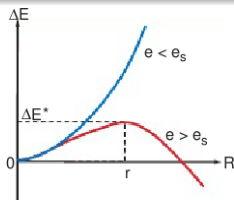
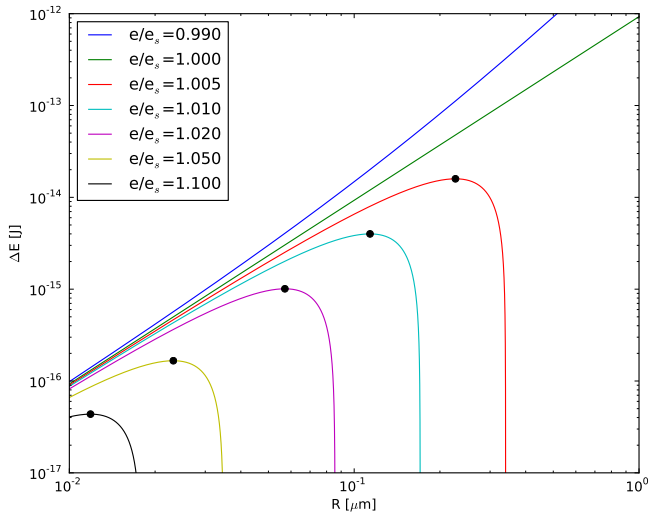


Fig. 6.1 Increase ΔE in the energy of a system due to the formation of a water droplet of radius R from water vapor with pressure e ; e_s is the saturation vapor pressure with respect to a plane surface of water at the temperature of the system.

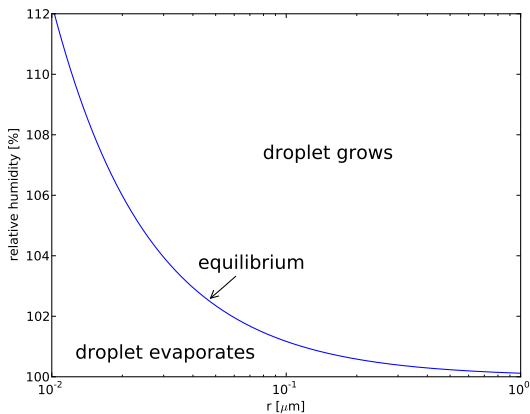
Figure from Wallace and Hobbs

- **blue curve:** subsaturated conditions, formation of droplets not possible
- **red curve:** supersaturated conditions, droplets grow above radius r

Energy difference due to formation of droplet



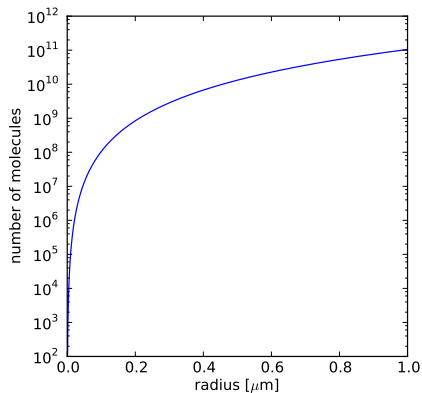
Kelvin equation



Kelvin equation

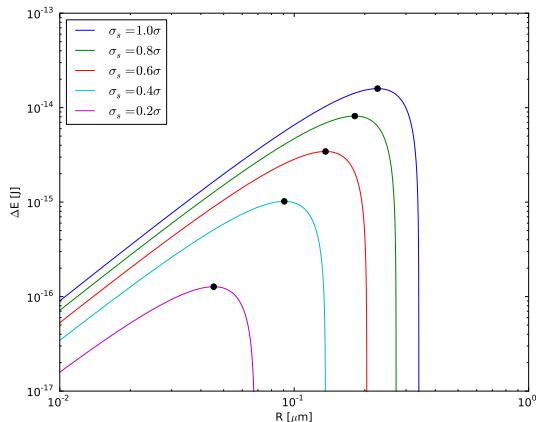
$$r = \frac{2\sigma}{nkT \ln \frac{e}{e_s}}$$

Number of molecules required to form stable embryonic droplet



e/e_s	r (μm)	N
1	∞	∞
1.01	$1.208 \cdot 10^{-1}$	$2.486 \cdot 10^8$
1.1	$1.261 \cdot 10^{-2}$	$2.807 \cdot 10^5$
1.5	$2.964 \cdot 10^{-3}$	$3.645 \cdot 10^3$
5	$7.486 \cdot 10^{-4}$	58

Heterogeneous nucleation



Surface tension is reduced when soluble aerosol is added to droplet.

Calculation for 0.5% supersaturation, $T=293$ K.

Raoult's law

Vapor pressure of an ideal solution depends on mole fraction of the component present in the solution

$$\frac{e'}{e} = f$$

- e' – saturation water vapor pressure adjacent to solution droplet containing a mole fraction f of pure water
- e – saturation water vapor pressure adjacent to pure water droplet
- f – number of moles of pure water divided by total number of moles

⇒ saturation water vapor pressure is reduced when aerosol is solved in droplet

Köhler curves

$$\frac{e}{e_s} = \left(\exp \frac{2\sigma'}{n'kTr} \right) \left(1 + \frac{imM_w}{M_s \left(\frac{4}{3}\pi r^3 \rho' - m \right)} \right)^{-1}$$

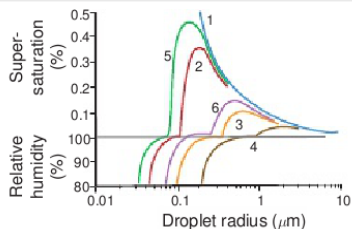


Fig. 6.3 Variations of the relative humidity and supersaturation adjacent to droplets of (1) pure water (blue) and adjacent to solution droplets containing the following fixed masses of salt: (2) 10^{-19} kg of NaCl, (3) 10^{-18} kg of NaCl, (4) 10^{-17} kg of NaCl, (5) 10^{-19} kg of $(\text{NH}_4)_2\text{SO}_4$, and (6) 10^{-18} kg of $(\text{NH}_4)_2\text{SO}_4$. Note the discontinuity in the ordinate at 100% relative humidity. [Adapted from H. R. Pruppacher, “The role

Figure from Wallace and Hobbs

Köhler curves

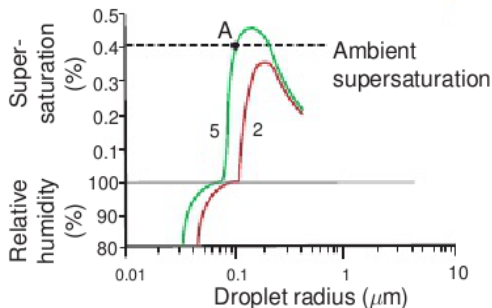


Fig. 6.4 Köhler curves 2 and 5 from Fig. 6.3. Curve 2 is for a solution droplet containing 10^{-19} kg of NaCl, and curve 5 is for a solution droplet containing 10^{-19} kg of $(\text{NH}_4)_2\text{SO}_4$. The dashed line is an assumed ambient supersaturation discussed in the text.

Maximum of Köhler curves

Values of radius at maximum r^* and supersaturation S^* as functions of nucleus mass and radius. Here spherical NaCl particles at 273°K are assumed.

mass of dissolved salt [g]	r_s [μm]	r^* [μm]	(S^*-1) [%]
$10 \cdot 10^{-16}$	0.0223	0.19	0.42
$10 \cdot 10^{-15}$	0.0479	0.61	0.13
$10 \cdot 10^{-14}$	0.103	1.9	0.042
$10 \cdot 10^{-13}$	0.223	6.1	0.013
$10 \cdot 10^{-12}$	0.479	19	0.0042

adapted from R.R. Rodgers

Droplet activation

Droplets grow along Köhler curve

- Case 1: when ambient supersaturation is higher than maximum
⇒ **activated** droplets
- Case 2: when ambient supersaturation is lower than maximum, droplets grow to equilibrium state, where ambient supersaturation equals supersaturation adjacent to droplet
⇒ **unactivated/haze** droplets

Efficiency of cloud condensation nuclei

- small subset of atmospheric aerosols serve as CCN
- CCN are most efficient when droplets can grow at supersaturations as low as possible
 - the larger the size the lower the required supersaturation
 - the greater the solubility the lower the required supersaturation
- completely wettable but insoluble particles
 - ⇒ $r \approx 0.1 \mu\text{m}$ at 1% supersaturation
- soluble particles
 - ⇒ $r \approx 0.01 \mu\text{m}$ at 1% supersaturation

Cloud condensation nuclei in Earth's atmosphere

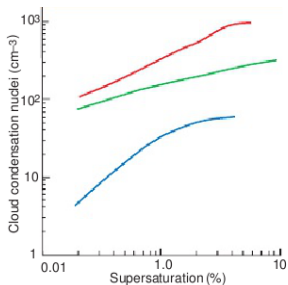


Fig. 6.5 Cloud condensation nucleus spectra in the boundary layer from measurements near the Azores in a polluted continental air mass (brown line), in Florida in a marine air mass (green line), and in clean air in the Arctic (blue line). [Data from J. G. Hudson and S. S. Yun, "Cloud condensation nuclei spectra and polluted and clean clouds over the Indian Ocean," *J. Geophys. Res.* **107**(D19), 8022, doi:10.1029/2001JD000829, 2002. Copyright 2002 American Geophysical Union. Reproduced/modified by permission of American Geophysical Union.]

Figure from Wallace and Hobbs

- no systematic latitudinal or seasonal variations have been found so far
- near Earth's surface: continental air masses contain larger concentrations of CCN than marine air masses
- soil and dust \Rightarrow not the dominant source
- forest fire, engine emission \Rightarrow efficient CCNs
- not yet clear, which are the dominant sources for cloud formation



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Review

Atmospheric organic and bio-aerosols as cloud condensation nuclei (CCN): A review

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Abstract

Organic substances have been recognized as active cloud condensation and ice formation nuclei for several decades. In some regions of the world, these organic compounds (OC) consist predominantly of suspended matter mass, which can have local (e.g. toxicity, health hazards) and global (e.g. climate change) impacts. However, due to the complexity of their chemical nature, the significance of organic molecules in driving physical and chemical atmospheric processes is still very uncertain and poorly understood. The aim of this review paper is to assess the current state of knowledge regarding the role of organic aerosols (including bioaerosols) as cloud condensation nuclei (CCN), as well as to compare the existing theoretical and experimental data. It seems that classical Kohler theory does not adequately describe the hygroscopic behaviour of predominantly identified organic CCN such as pure dicarboxylic acid particles. Factors such as surface tension, impurities, volatility, morphology, contact angle, deliquescence, and the oxidation process should be considered in the theoretical prediction of the CCN ability of OC and the interpretation of experimental results. Major identified constituents of organic CCN, their main sources and their CCN properties will be herein reviewed. We will also discuss areas of uncertainty and expose key issues deserving of future research.

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Organic aerosols as CCN

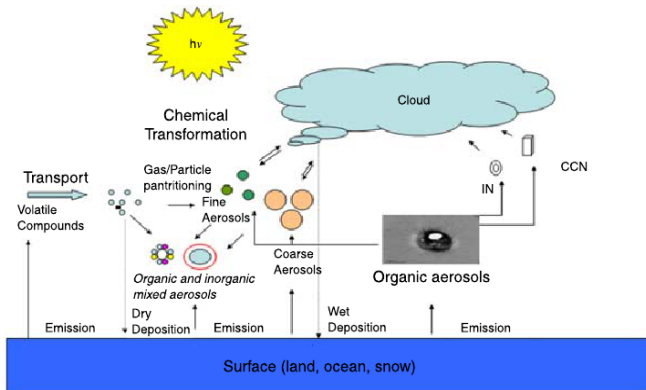
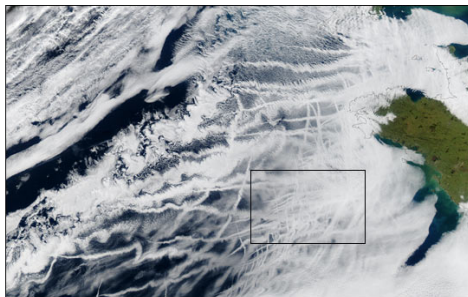


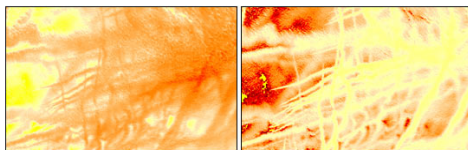
Fig. 1. The simplified schematic of organic aerosol transformation in atmosphere.

from Sun and Ariya 2005

Ship tracks



True Color



Optical Thickness

Effective Particle Radius (μm)

<http://earthobservatory.nasa.gov/IOTD/view.php?id=3275>

Growth of droplets in warm clouds

1. Growth by condensation
2. Growth by collision and coalescence

Droplet growth by condensation

- air parcel rises, expands, cools adiabatically and reaches saturation
- further lifting produces supersaturation
- as supersaturation rises, CCN are activated (most efficient first)
- supersaturation reaches maximum when:

rate of water vapor
in excess of saturation
made available by adiabatic
cooling = rate of water vapor
which condenses on
CCN and droplets

concentration of cloud droplets = concentration of CCN
activated by attained
peak supersaturation

Growth rate and size distribution

- growing droplets consume water vapor faster than it is made available by cooling and supersaturation decreases
- haze droplets evaporate, activated droplets continue to grow by condensation

growth rate of water droplet

$$\frac{dr}{dt} = G_I S \frac{1}{r}$$

- smaller droplets grow faster than larger droplets
- sizes of droplets in cloud become increasingly uniform, approach **monodispersed** distribution

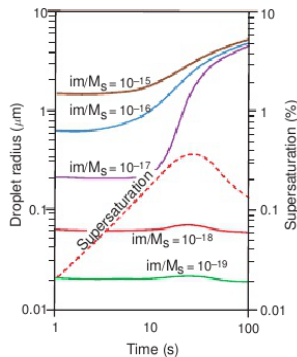


Fig. 6.16 Theoretical computations of the growth of cloud condensation nuclei by condensation in a parcel of air rising with a speed of 60 cm s^{-1} . A total of 500 CCN cm^{-1} was assumed with im/M_s values [see Eq. (6.8)] as indicated. Note how the droplets that have been activated (brown, blue, and purple curves) approach a monodispersed size distribution after just 100 s. The variation with time of the supersaturation of the air parcel is also shown (dashed red line). [Based on data from *J. Meteor.* **6**, 143 (1949).]

Figure from Wallace and Hobbs

Size distribution

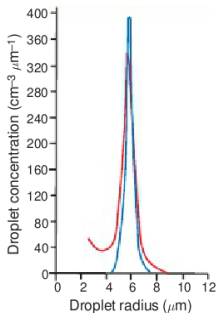


Fig. 6.17 Comparison of the cloud droplet size distribution measured 244 m above the base of a warm cumulus cloud (red line) and the corresponding computed droplet size distribution assuming growth by condensation only (blue line). [Adapted from Tech. Note No. 44, Cloud Physics Lab., Univ. of Chicago.]

Figure from Wallace and Hobbs

- measurement of size distribution shows good agreement to computed droplet size distribution for nonprecipitating warm cumulus cloud
- largest droplets only about $10 \mu\text{m}$

Sizes of cloud droplets

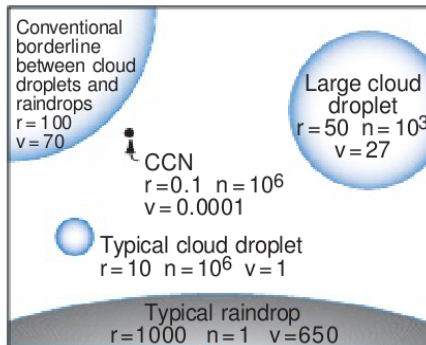


Figure from Wallace and Hobbs

- growth by condensation alone can not produce raindrops with radii of several mm !

Current cloud research

Cloud research at Schneefernerhaus (Zugspitze),
DLR Oberpfaffenhofen, KIT Karlsruhe and MPI Göttingen

Film: “Rätsel am Himmel: Was Forscher aus den Wolken lesen”

BR, 6.11.2011, 23:15, Faszination Wissen

[http://www.br.de/fernsehen/bayerisches-fernsehen/
sendungen/faszination-wissen/fawi-wolken100.html](http://www.br.de/fernsehen/bayerisches-fernsehen/sendungen/faszination-wissen/fawi-wolken100.html)