CLOUDS

Cornelius Schiller
Forschungszentrum Jülich, Germany

c.schiller@fz-juelich.de
Outline

Introduction
• Role of clouds in the climate system
• Cloud types
• Cloud life cycle

Cloud formation and precipitation
• Cooling
• Warm Clouds
• Cold clouds
• Precipitation

Special aspects from recent studies
• IWC of cirrus clouds
• Super-Supersaturation
• Contrails
Literature

• Seinfeld, J. H. and S. H. Pandis, Atmospheric Chemistry and Physics, Wiley Interscience, 1997
• Peter, T. et al., When dry air is too humid, Science, 2005
• + many individual publications
• + Meteorology standard textbooks
Role of clouds in the climate system

- Clouds are a major factor in the Earth’s radiation budget
- Clouds are a key step in the hydrological cycle
- Clouds provide a medium for (heterogeneous) chemical reactions
- Clouds affect significantly vertical transport and redistribution of species in the atmosphere
## Radiative Forcing Components

<table>
<thead>
<tr>
<th>RF Terms</th>
<th>RF values (W m$^{-2}$)</th>
<th>Spatial scale</th>
<th>LOSU</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Anthropogenic</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Long-lived greenhouse gases</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ozone</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Stratospheric water vapour from CH$_4$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Surface albedo</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Aerosol</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Solar irradiance</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Natural</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total net anthropogenic</td>
<td>1.6 [0.6 to 2.4]</td>
<td>Global</td>
<td>Low</td>
</tr>
</tbody>
</table>

### Key Components:
- **CO$_2$**: 1.66 [1.49 to 1.83] W m$^{-2}$, Global, High
- **N$_2$O**: 0.48 [0.43 to 0.53] W m$^{-2}$, Global, High
- **CH$_4$**: 0.16 [0.14 to 0.18] W m$^{-2}$, Global, High
- **Halocarbons**: 0.34 [0.31 to 0.37] W m$^{-2}$, Global, High
- **Stratospheric Ozone**: -0.05 [-0.15 to 0.05] W m$^{-2}$, Continetal to global, Med
- **Tropospheric Ozone**: 0.35 [0.25 to 0.65] W m$^{-2}$, Continetal to global, Med
- **Surface Albedo Change**:
  - Land use: -0.2 [-0.4 to 0.0] W m$^{-2}$, Local to continental, Med
  - Black carbon on snow: 0.1 [0.0 to 0.2] W m$^{-2}$, Local to continental, Med
- **Total Aerosol**:
  - Direct effect: -0.5 [-0.9 to -0.1] W m$^{-2}$, Continetal to global, Med
  - Cloud albedo effect: -0.7 [-1.6 to -0.3] W m$^{-2}$, Continetal to global, Low
- **Linear contrails**: 0.01 [0.003 to 0.03] W m$^{-2}$, Continental, Low
- **Solar irradiance**: 0.12 [0.06 to 0.30] W m$^{-2}$, Global, Low
More CCN
→ more but smaller drops (cloud albedo/Twomey effect)
→ higher reflectivity & longer lifetime
→ less sun on Earth’s surface
→ cooling
Aerosols – Clouds – Climate

Scattering & absorption of radiation
Unperturbed cloud
Increased CDNC (constant LWC) (Twomey, 1974)
Drizzle suppression. Increased LWC
Increased cloud height (Pincus & Baker, 1994)
Increased cloud lifetime (Albrecht, 1989)
Heating causes cloud burn-off (Ackerman et al., 2000)

Direct effects
Cloud albedo effect/ 1st indirect effect/ Twomey effect
Cloud lifetime effect/ 2nd indirect effect/ Albrecht effect
Semi-direct effect

Top of the atmosphere
Surface

Indirect effect on ice clouds and contrails

considered in IPCC RF
Hydrological cycle

total water on Earth: $1.4 \cdot 10^9$ km$^3$

- oceans 97.4%
- polar ice 1.9%
- ground water 0.5%
- soil 0.01%
- biosphere 0.003%
- atmosphere 0.001%

atmospheric $\text{H}_2\text{O}$ 4% - 1 ppmv

total atmospheric $\text{H}_2\text{O}$ 25 mm

annual precipitation 800 mm

$\text{H}_2\text{O}$ exchange rate 10-11 days
Chemical reactions in clouds: washout

Cloud Condensation Nuclei
and water vapor supersaturations
relative humidity larger 100%

Washout of particles and gases
Acid rain
Chemical reactions in clouds: surface reactions

stratospheric ozone

Winter

T < -80°C

Cl₂, Cl₂

ClINO₂ + HCl → Cl₂ + HNO₃

Cl₂, Cl₂

Spring

Cl₂

Cl₂ + photon → Cl + Cl

Cl₂, Cl₂, Cl, ClO, (ClO)₂

source gas
CFC

reservoirs
HCl, ClONO₂

reactive
Cl₂, Cl, ClO, (ClO)₂
Cloud impact on vertical transport

Corti et al.
Cloud types
Low Clouds

Stratus (St)

Cumulus Congestus

Cumulus (Cu)
Medium-high Clouds

Altostratus (As)

Altocumulus (Ac)
**High Clouds**

- Cirrostratus (Cs)
- Subvisible cirrus (SVC/UTTC)
- Cirrocumulus (Cc)
- Cirrus (Ci)
- Contrails
Cumulonimbus (Cb)
Polar Stratospheric Clouds (mother-of-pearl; nacreous)

Noctilucent Clouds
Precipitation staircase

Prerequisites for cloud formation:
- water
- low T
- supersaturation
- Cloud Condensation Nuclei (CCN) or Ice Nuclei (IN)
Cloud formation and precipitation

- Cooling
- Warm Clouds
- Cold clouds
- Precipitation
Cooling

- Isobaric cooling
- Adiabatic cooling
Fig. 14 Cloud and front formation—causes of rising air.
Frontal cloud formation

Development of Stratus Clouds by slow lifting at a Warm Front

Cirrostratus

Warm Air Sector

Altostratus

Warm front

Nimbostratus

Cold Air Sector

Precipitation Zone

Several hundred kilometers
Convective cloud formation

(1.)
Temperature | Ground Level | Air Pressure

(2.)
Temperature | Ground Level | Air Pressure

(3.)
Temperature | Ground Level | Air Pressure

(4.)
Temperature | Ground Level | Air Pressure

(5.)
Temperature | Ground Level | Air Pressure

(6.)
Temperature | Ground Level | Air Pressure

Condensation Level
Orographic cloud formation

also up to cirrus / PSC altitudes
Adiabatic cooling

latent heat release $\Delta H_v$ by condensation

moist adiabatic lapse rate:

$$\Gamma_s = \frac{g + \Delta H_v (d \omega_{vs}/dz)}{\hat{c}_p} < \Gamma$$

lifting condensation level

$$h_{l,cli.} = \frac{T_0 - T_L}{\Gamma}$$

$$\frac{dT}{dz} = -\Gamma \quad \text{or} \quad \frac{dT}{dt} = -\Gamma \cdot w$$

$$\Gamma = g/\hat{c}_p \quad \text{dry adiabatic lapse rate}$$
Cumulus formation depends on H₂O content and stability of atmosphere.

1: stable atmosphere
   updraft stopped early cumulus humilis

2: unstable atmosphere
   updraft stopped high up cumulus congestus

ΔT causes updraft
Stability in the atmosphere

unstable
strong T-gradient

stable
weak T-gradient or inversion
Warm Clouds
Equilibrium between phases: Clausius Clapeyron equation

\[
\frac{dp^o}{dT} = \frac{\Delta H_v(T) M_w}{T(v_v - v_w)}
\]

\(\Delta H_v(T)\) specific heat (water evap.)
\(M_w\) molecular weight

\(v_v \gg v_w\)

\(p^o v_v = RT\)

\[
\frac{dp^o}{dT} \sim \frac{\Delta H_v(T) p^o M_w}{RT^2}
\]

Water Vapor Concentration, g m\(^{-3}\)

Supercooled water

Temperature, °C
Sättigungsflechte der Luft
(g/kg feuchter Luft bei 1013 mbar Luftdruck)
Equilibrium of water droplet vs flat surface

Kelvin equation

\[ \frac{p_w(D_p)}{p^o} = \exp \left( \frac{4M_w \sigma_{w0}}{RT \rho_w D_p} \right) \]

\[ p_w > p^o \rightarrow \text{for equilibrium of droplet, air needs to be supersaturated} \]
Supersaturation of several 100% required in particle-free air → cloud condensation nuclei (CCN) required

Higher critical supersaturation is needed for
- less particle solubility (bad water uptake)
- smaller particles

\[
\ln \left(\frac{p_w(D_p)}{p^o}\right) = \frac{A}{D_p} - \frac{B}{D_p^3}
\]

curvature term  solute effect
Activation of aerosol particles to drops

Good CCN: large, high water soluble fraction, i.e. salts
Bad CCN: small, high insoluble fraction, i.e. soot, dust or high organic fraction
Aerosol Composition

Water soluble inorganic
Water soluble organic
Insoluble

McFiggins et al. (2006), ACP
Cold (ice) clouds
Supercooled water, mixed phase, ice clouds

![Graph showing the frequency of appearance of liquid, mixed, and ice phases as a function of temperature.](Image)
Clausius Clapeyron equation

**vapour/ice**

\[
\frac{dp_{\text{sat},i}}{dT} = \frac{\Delta H_s}{T(v_u - v_i)}
\]

\[
\frac{d \ln p_{\text{sat},i}}{dT} \approx \frac{\Delta H_s}{RT^2}
\]

\(\Delta H_s\) molar enthalpy for ice sublimation

Integration \(\ln p_{\text{H}_2\text{O}} = -A/T + C\)

[Marti & Mauersberger, GRL 1993]

\(\Delta [\text{H}_2\text{O}] = 1 \text{ ppmv} \Rightarrow \Delta T_{\text{frost}} \approx 1 \text{ K}\)

**water/ice**

\[
\frac{dp_m}{dT} = \frac{\Delta H_m}{T(v_w - v_i)}
\]

\(\Delta H_m\) enthalpy for melting
p-T phase diagram for water

- Triplet Point: $T=0^\circ C, p=6.1$ hPa
- Metastable equilibrium supercooled water/vapour

Water Partial Pressure, Pa

Temperature, $^\circ C$
Bergeron-Findeisen process

\[ T < 0^\circ \text{C}, \ p_{\text{sat},w} > p_{\text{sat},i} \]
supercooled droplets cannot coexist in equilibrium with ice crystals
Ice Nuclei (IN)
Homogeneous / heterogeneous ice nucleation
determined by IN composition and supersaturation

Homogeneous ice nucleation
T: < 235 K
RH(ice): 140 – 170 %

Heterogeneous ice nucleation
T: < 273 K
RH(ice): 100 – 150 %

bad IN: soluble solutions
organics

good IN: soot
mineral dust
A hom. freez. of solution droplets
B deliquescence + hom. freez.
C hom/het freez. + secondary phase cryst. (immersion freezing)
D het. freez. of solution droplets
E deposition nucl. on insoluble/anhydrous particle
F contact freezing nucleation
Ice saturation at low T: Homogeneous nucleation

Koop et al., 2000 AIDA experiments
Ice saturation at low T: Heterogeneous nucleation

The graph illustrates the relationship between relative humidity (RH) and temperature for both homogenous and heterogeneous freezing processes. Different markers and line types represent various types of nuclei, such as Sulphuric acid (SA), Ammonium sulphate (AS), Soot, Soot+SA coating, Soot+AS coating, Soot coating, Ariz. Test dust – ATD, and Saharan dust – SD. The data points show a decreasing trend in RH with increasing temperature, reflecting the freezing process.
Supersaturation in the atmosphere

inside clouds

outside clouds

Ovarlez et al.
Precipitation

(some) drops need to grow to precipitable size

mechanisms:
• water vapour condensation
• droplet coalescence
• ice processes
Diffusional growth of drops

\[ m_g = 10^{-10} \text{ g} \]

\[ 10^{-13} \text{ g} \]

\[ 10^{-16} \text{ g} \]
Droplet coalescence
alling (large) drops collect smaller drops in fall path  (Mt 25,29)

For everyone who has will be given more, and he will have an abundance. Whoever does not have, even what he has will be taken from him.

Denn wer hat, dem wird gegeben, und er wird im Überfluss haben; wer aber nicht hat, dem wird auch noch weggenommen, was er hat.

Car à celui qui a, on donnera, et il aura encore davantage; mais à celui qui n'a pas, on ôtera même ce qu'il a.
Ice processes

- Aerosolteilchen
  - dienen als Kondensationskeime
  - heterogene Kondensation
  - dienen als Eiskeime

- Wasserdumpf
  - homogene Kondensation
  - sublimiert
  - dienen als Eiskeime

- Wolkentröpfchen
  - wachsen
  - verdunsten

- Nieseltröpfchen
  - Verdunsten

- Kondensation
  - Impaktion
  - Verdunsten

- Wolkentröpfchen
  - Frieren aneinander fest
  - Nieder verbringen andere Hydrometeore

- Eiskristalle
  - haften aneinander, wachsen

- Graupelbildung
- Riming

- Graupel
  - diert als Embryo

- Schneeflocken
  - Schmelzen

- Eiskörner
  - dienen als Kristallkeime

- Hagel
  - dienen als Embryonen

- Regentropfen
  - zerplatzen: Break up

- Temperatur
  - 0 °C
Example: Microphysics in a Cb cloud
Clouds 3
Special aspects from recent studies

- IWC of cirrus clouds
- Super-supersaturation (part 1, part 2 by TP)
- Contrails and contrail cirrus
Ice Water Content (IWC) of cirrus

Measurement: total water – gas phase water

Schiller et al., 2008

- tropical cirrus
- midlatitude cirrus
- polar cirrus

> 5 aircraft campaigns
Detection limits

1st method
total water – measured gas phase

2nd method
total water – saturation (from pT measurement)
FISH/FLASH vs FSSP IWC

- CIP size range contributes more than 50% to IWC
- FSSP size range contributes more than 50% to IWC

IWC from FISH - FLASH (g/m³)

IWC from CIP + FSSP (g/m³)
Mean IWC and frequency distribution

Temperature

IWC_sat (ppmv)

- 40-100%
- 30-40
- 20-30
- 15-20
- 10-15
- 5-10
- 0-5

Logarithmic scale
IWC and cooling / convection

\[ \text{IWC}_{\text{sat}} = \text{H}_2\text{O}_{\text{sat, ice}}(T+\Delta T) - \text{H}_2\text{O}_{\text{sat, ice}}(T) \]

- \( \Delta T = 10 \text{ K} \)
- \( \Delta T = 5 \)
- \( \Delta T = 1 \)
- \( \Delta T = 0.1 \)
- \( \Delta T = 0.01 \)

- Mean IWC, noconvective flights
- Median IWC
- Noconvective flights
- Convective
Ultrathin tropical cirrus (UTTC)

final step of dehydration of stratospheric air

Peter et al., 2002
Stabilisation of UTTC

Vertical motion of a particle

\[
\frac{dz}{dt} = v_{air}(z) - \frac{2g \rho r^2}{9\eta},
\]

Growth/evaporation of particles

\[
\frac{dr^2}{dt} = \frac{m}{\rho} D_{H_2O} n_{H_2O}^{vap}[S_{ice}(z) - 1].
\]
The High Supersaturation Puzzle

Peter et al., 2007
FLASH/OJSTER: supersaturation climatology

Inside Cirrus

Outside Cirrus

Aircraft
Geophysica
Falcon
Lear Jet

green: mid latitudes
blue: high latitudes
red/yellow: tropics

Krämer et al., 2009
Supersaturation: frequency distribution and T-dependence

Reprocessed data (28 flights)
- only few data > homogeneous f.t.
- no data > water saturation
- inside cloud RHi peaks at 100%
- broader distribution at $T < 205$ K

Krämer et al., 2009
Saturation ratio with respect to ice:

\[ S = \frac{p_{\text{H}_2\text{O}}}{p_{\text{vap}}(T)} = \frac{n_{\text{H}_2\text{O}}}{n_{\text{vap}}(T)} \]

- \( p_{\text{H}_2\text{O}} \) = partial pressure of water
- \( p_{\text{vap}}(T) \) = vapor pressure of ice

- \( S > 1 \) \( \rightarrow \) ice particles grow
- \( S = 1 \) \( \rightarrow \) ice particles are in equilibrium with the gas phase
- \( S < 1 \) \( \rightarrow \) ice particles evaporate
How does water vapor condense on ice particles?

$$\frac{dn_{H_2O}}{dt} = -4\pi D^* r n_{ice} (n_{H_2O} - n_{vap})$$

$$\tau_{cond} = \frac{1}{4\pi D^* r n_{ice}}$$

How is supersaturation maintained?

$$\frac{dn_{vap}}{dt} = \frac{dT}{dt} \frac{dn_{vap}}{dT} = -\frac{\Gamma w B}{T^2} \left(1 - \frac{T}{B}\right)n_{vap}$$

$$\tau_{cool} = \frac{1}{\Gamma' w (B/T^2) (1 - T'/B)}$$

$$P_{vap}(T) = A e^{-B/T}$$
\[ \frac{d n_{\text{H}_2\text{O}}}{dt} = \cdots \]

\[ \frac{d n_{\text{vap}}}{dt} = \cdots \]

\[ \frac{dS}{dt} = \frac{d}{dt} \frac{n_{\text{H}_2\text{O}}}{n_{\text{vap}}} \]

\[ = - \frac{S - 1}{\tau_{\text{cond}}} + \frac{S}{\tau_{\text{cool}}} \]

\[ \tau_{\text{cond}} = \frac{1}{4\pi D^* r n_{\text{ice}}} \]

\[ \tau_{\text{cool}} = \frac{1}{\Gamma w(B/T^2) (1-T/B)} \]
Steady-state $S$ in an upwelling air parcel ($w$)

\[
\frac{dS}{dt} = -\frac{S - 1}{\tau_{\text{cond}}} + \frac{S}{\tau_{\text{cool}}} = 0
\]

\[
S - 1 \approx \frac{S \hat{w}}{\hat{r} \hat{n}_{\text{ice}}}
\]

\[
\hat{w} = \frac{w}{(1 \text{ m/s})}
\]
\[
\hat{r} = \frac{r}{(1 \mu\text{m})}
\]
\[
\hat{n}_{\text{ice}} = \frac{n_{\text{ice}}}{(1 \text{ cm}^{-3})}
\]
Ice crystal number densities
- vertical velocity \( w \)
- frost point at which air starts
- aerosol properties varied

Mean ice crystal radii
Ice water content

Kärcher & Lohmann, 2002
Growth time of ice particles

(up to 80% of equilibrium radius)
Supersaturation and $N_{\text{ice}}$

(persistent) supersaturation consistent with low $N_{\text{ice}}$

supersaturation puzzle $\rightarrow$ freezing suppression puzzle

homogen. freezing and low $u_z$?

heterogen. freezing?

freezing suppression by organics?

FSSP measurements from 20 flights by S. Borrmann and M. deReus
Contrails / contrail cirrus
Contrails and RHi

RHi > 100%
persistent contrails

RHi < 100%
lifetime of minutes

embedded in (sub-)visible cirrus?
generating new cirrus?
Cirrus

Contrails

Krämer, CONCERT 2008
During CONCERT large ice crystals (100µm) observed in short-lived contrail → entrainment from surrounding cirrus