ICE OBSERVATIONS IN THE ATMOSPHERE
1949 – 2004

Six stories and a status report

June 7, 2004
Ice Initiation Workshop
The stories:

1. The foundation
2. Nucleation plurality
3. The graupel process
4. The multiplication success
5. The cloud seeding question
6. Other facets
Berichte
des
Deutschen Wetterdienstes
in der US-Zone

Nr. 6

Die Eisphase in der Atmosphäre

Von Dr. Helmut Weickmann, Hohenpeißenberg

Bad Kissingen, 1949

Weickmann, 1949
Abb. 16 Mikroskop im Beobachterstand der Nahaufklärermaschine Hs 126

Weickmann, 1949
Ci top, 8900m −47°C

Ci base, 6000m −26°C

Cs, 6000m −30°C

Weickmann, 1949
1. THE FOUNDATION.

• The large variety of cloud forms, and their varying propensity to produce precipitation proved elusive to meaningful scientific analyses until the beginnings of the 20th century.

• Advances in the physical chemistry of colloids proved to be a useful basis for thinking about clouds in the atmosphere, and to address the perplexing question of precipitation formation.
“Snowe is a cloude congeled by greate colde, before it be perfectlye resolued from vapors into water. ...... Other matters of snowe because they are commen with raine, are needles to be spoken of. ...... Snowe causeth thinges growing to be fruitfull, ...”
Alfred Wegener, 1911

Based on observations made during arctic expeditions, he argued that ice formation needed *sublimation nuclei* just as droplet formation needs condensation nuclei.

Thought that "isomorphism" is the essential criterion, and that quartz particles fill that role.

Recongimized that colloidal instability results from vapor pressure difference between ice and water at temperatures <0°C.
Discusses the importance of the difference between saturation with respect to water or ice, the impact this has on ice forms. Cites evidence for ice contrail in clear air (diagnosed by 22º halo), ice fogs at temp. <-40ºC.

Recognizes that the existence of water droplets at temperatures below 0C play a role in the formation of rime-graupel and hail.

Considers pileus (if cold enough) to seed cumulus that rises into it.
Frostübersättigung und Cirren.

Von Alfred Wegener. — (Mit einer Figur.)

Frostübersättigung. Unter den Zuständen des Wasserdampfes in der Atmosphäre gibt es einen interessanten Bereich, in welchem die Luft in bezug auf Eis übersättigt, in bezug auf unterkühltes Wasser aber noch ungesättigt ist. Wir wollen diesen Zustand, um einen kurzen Namen zu haben, als „Frostübersättigung“ bezeichnen. Im Roezeboomischen Zustandsdiagramm (s. Figur), in welchem der Dampfdruck nach oben und die Temperatur nach rechts abgetragen wird, liegt der Bereich der Frostübersättigung in dem schraffierten Raum zwischen der Gleichgewichtskurve der beiden Phasen Dampf und unterkühltes Wasser TW und der davon abweichenden Gleichgewichtskurve der Phasen Dampf und Eis TE. Diese beiden Kurven vereinigen sich einerseits im Tripelpunkt T des Wassers (Koordinaten \( e = 4.5 \)) und liegen in den Bereich der gesenkten Kondensation darstellt, welche das Auftreten wesentlich begünstigt. Die konventionell getroffene Auswahl ist daher erheblich fälliger, wenn die Komponente fehlen, während die Kondensation beider Phasen in dem schraffierten Raum liegen, besonders wenn sie auf unterkühltes Wasser beschränkt bleibt. In anderen Fällen ist die Auswahl der Zustände, bei denen das Wasser unterkühlt ist, wichtig, da sich die Kondensation der Dampfphase ausbreitet.

1) Die Konstanz der Zustände ist essentiell für die Kondensation des Wasserdampfes.
Distribution du stratus observé en février 1922 dans la forêt de sapins de Voksenkollen d'Oslo, 470 m d'alt., à des températures > 0 °C et environ −10 °C respectivement. La partie ombrée représente la couche de stratus.

Bergeron, 1935
ON THE PHYSICS OF CLOUD AND PRECIPITATION
by Dr. T. Bergeron, Oslo.
(Received June 1933.)

Introduction. The evolution of the theories on the physical and meteorological condition of the formation of cloud and their precipitation offers such a typical example of the "zig-zag progress" of science that an introductory retrospect seems worth while.

No means perform.

II. We have thus tried to show that none of the hitherto recognised factors of cloud coagulation represent the universal release of real precipitation (real rain and snow). Either the factor is only active at special times of the day (3), or under abnormal electric conditions (1b), or can only coagulate droplets of fog dimensions or smaller, without causing few great drops (1a), (2), (4), or is ineffective on the whole as a release of coagulation (5).

6. Then there remains only one factor: neighbouring elements of different phase (i.e. some cloud elements liquid, some solid) at temperatures below the freezing point — an effect that seems to have been too little heeded hitherto, but which I hope to show is the chief one, physically and meteorologically.

We will first treat the more physical side of the question. A supercooled mixture of crystals and droplets must be colloidal-thermodynamically unstable, due to the difference of maximal vapour tension over ice and water at frost-temperatures. This difference $\Delta e$ amounts to 0.24 mb at $-10^\circ C$, and is thus much greater than the corresponding differences due to the factors (1) or (2) above ever can get when $d > 5 \mu$. As to the factor (3), a $\Delta e = 0.24$ mb corresponds to a $\Delta t$ of almost 1.0$^\circ C$ at $-10^\circ C$. 
"..... the difference in phase between neighbouring elements ... can in two cases occur without any considerable relative motions:

a) Berson, Wegener, Douglas and others have observed fogs consisting of droplets down to temperatures of $-20^\circ$ or even $-30^\circ$C. ... In the air there will, however, probably be a small amount of such particles, which can gradually get into action as sublimation nuclei, as the temperature falls. ... Thus, to every temperature $< 0^\circ$C will correspond a certain probability of crystallization resp. a certain frequency of crystals within supercooled water cloud ...

b) ... layers above the isotherm of $-10^\circ$ or $-20^\circ$C mostly contain ice crystals ..... ascending water cloud mass, protruding into this region, may then become infected by some crystals by turbulence ..."

" In our mixed or supercooled cloud the three phases ice, vapour and liquid water are in contact with each other through the vapour. ... the process of transporting tha total water quantity of the droplets by molecular diffusion to the crystals would be achieved in 10 – 20 minutes."
the stable layer clouds in the former picture.

Conclusions. — This cloud genetics and classification is only a first attempt. The International Cloud Year, I hope, will give us the necessary data to verify, modify or abandon it. But in any case it might perhaps serve as a base of discussion and help to tell us what we have to look for in the abundant material which has been collected now.

The historical display of the theories of cloud and rain have also shown us that theories, once completely done away with, may rise to great honour again, as for instance the „barometric fog” — revived by Swoboda and me 1924, and now even exaggerated by Giaiö 1931 — and that an entirely one-sided theory is less helpful to our extremely complex science.

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Vali - June 7, 2004
Bergeron, 1935 (paraphrased):

In addition to other processes that can induce precipitation in colloidally stable clouds, a physically and meteorologically important process arises from the special situation at temperatures below 0°C created by the simultaneous presence of supercooled liquid and ice.

As a consequence of this realization, it became imperative to look carefully at how ice particles originate, how many get started at various temperatures, how other factors (cloud type, etc.) might have importance.
Forschungs- und Erfahrungsberichte

des Reichswetterdienstes

Im Auftrage des
Reichsministers der Luftfahrt und Oberbefehlshabers der Luftwaffe
herausgegeben vom
Reichsamt für Wetterdienst
Reihe B, Nr. 1

Unterkühlte Wasser wolken
und Eiswolken

Von Wilhelm Peppler

Berlin 1940 * Gedruckt in der Reichsdruckerei
Fig. 9
Häufigkeit der Unterkühlungstemperaturen

Fig. 12
Vorkommen der Unterkühlungen und der Eiswolken

Peppler, 1940
Forschungs- und Erfahrungsberichte des Reichswetterdienstes
Im Auftrage des Reichsministers der Luftfahrt und Oberbefehlshabers der Luftwaffe herausgegeben vom Reichsamt für Wetterdienst (Luftwaffe)
Reihe B, Nr. 8

Geheim!

Ergebnisse von Wolken- und Niederschlagsbeobachtungen bei Wettererkundungsfügen über See
Von W. Findeisen, Prag

Berlin 1942 * Gedruckt in der Reichsdruckerei
onset of ice formation

absence of ice formation

Zeichenerklärung:

- - - - - Eisteilchenbildung über See.
- - - - - Eisteilchenbildung über Land.

Keine Eisteilchenbildung über See.
Keine Eisteilchenbildung oder Mischwolken über Land.

- - - - = Grenzen für Landgebiete; - - - - = Grenzen für Seegebiete

Quellwolkenstatistik nach Beobachtungen über Seegebieten;
zum Vergleich Werte aus der Wolkenstatistik nach W. Peppler für Landgebiete.

Findeisen, 1942
late 1930s

Findeisen (building on Wegener and Bergeron, and using observations of Peppler and of his own):

Examined frequencies of supercooled clouds from soundings and the occurrence of ice from optical phenomena and found that ice clouds are frequent only at temperatures below $-10^\circ C$ or even $-20^\circ C$.

Recognized that riming further accelerates the growth of ice crystals. Dependence on droplet size is speculated. Discussed the link to aircraft icing (translated by National Advisory Committee for Aeronautics).

Foresaw possibility of deliberately influencing precipitation due to sparcity of sublimation nuclei.

The Wegener–Bergeron–Findeisen theory is now complete and is rapidly accepted. All of cloud physics revolves around two kinds of nuclei – condensation nuclei and sublimation nuclei, and the competition between them under various conditions (e.g. Findeisen, 1938). "Colloid–meteorology" to become a partner to "meteorology" and "aerology".
2. NUCLEATION PRULARITY.

up to late 1940s:

• Concept of germ formation - nucleation - is well known for variety of systems (Volmer, Krastanow, others).

• Freezing of water is studied in the laboratory (Dorsey, Rau)

• Weickmann (1942) reported little and very slow ice formation at $T = -40^\circ\text{C}$ and $S_w < 1$ on particles placed on a chilled mirror. Ice always formed at $S_w > 1$, in amounts depending on material tested.

• Cwilong, Fournier d’Albe found the same as Weickmann, and noted critical temperatures of $-33^\circ\text{C}$ for ice always forming, and $-41^\circ\text{C}$ at which ice formation became abundant.

• Findeisen and Schulz (1944, Prague) used a 2-m$^3$ chamber with slow adiabatic expansion. Found very few sublimation nuclei. Water clouds formed after $S_w > 1$ even at $T < -30^\circ\text{C}$ with just a few crystals observed.
aufm Kampe, Wall, Fournier-d'Albe:

*Ice crystals in contrails and in fogs form only when water saturation is reached or exceeded; 'freezing nuclei' are needed.*

The hope (expectation) arose that nucleus measurements can quantify ice occurrence and can be used to predict ice initiation in clouds.

The possibility of cloud seeding seemed to have an ample and open window.
late 1940s

**Langmuir, Schaefer, Vonnegut (Project Cirrus)**

Bowen:

*Cloud seeding tests show that supercooled clouds can be "glaciated" with the addition of AgI nuclei.*

*Confirmation of Bergeron's thesis. Start of many weather modification projects.*

**Cwilong, Dorsey, Weickmann:**

*Laboratory measurements of ice nuclei support or even explain the trend observed in clouds toward more ice at lower temperatures. Metals and metal halides are effective freezing nuclei.*

**Nakaya, Kumai, Weickmann:**

*Ice crystal form also varies systematically with temperature.*
**Principal ice nucleation modes**

**Deposition**
- $S_i > 1$

**Immersion freezing**
- $T < T_{\text{act}}^{\text{imm}}$

**Condensation freezing**
- $S_w > 1$ and $T < T_{\text{act}}^{\text{cond}}$

**Contact freezing**
- $T < T_{\text{act}}^{ct}$
Some additional modes

Evaporation: \( S_w < 1 \) and \( S_i > 1 \) and \( T < T_{act} \)

Memory effect: \( S < S_i \) and \( S > S_i \)

Haze freezing: \( S_s > 1, S_w < 1 \) and \( T < T_{act} \)
• Only most basic of cloud processes can be simulated in laboratory chambers and samples are not fully representative.

• Prularity of nucleation modes (pathways) presents a complex instrumentation challenge.

• Reverse path: the examination of crystal residues, or the interpretation of crystal form (“droplet centers”), can offer some important distinctions, but never fully overcome the inherent ambiguity of the evidence.

• Shocks, cavitation, collisions, electric fields and discharges, ....
Figure 1. Nucleus concentrations measured by various instruments for natural aerosols in Experiment 12 (750527, 1400-1800). Data points are designated with symbols which are coded according to the instrument from which the measurement originated: A = Arizona low pressure chamber, F = Frankfurt low pressure chamber, C = NCAR continuous flow chamber, G = Gotz centrifuge with "puff" humidification, N = NOAA static diffusion chamber, S = SUNYA static diffusion chamber; W = Wyoming static diffusion chamber, DFC = Wyoming drop freezing counter, SCC = settling cloud chamber. The values on the abscissa are $S_i$ supersaturation with respect to ice, except for the DFC and SCC for which the values are $-T$ ($^\circ$C).
UW KING AIR DATA

DATE: 900427   TIME INTERVAL: 090343-125914

number of accepted points below and above range x: 0 2115
    y: 5024 4

number of points accepted: 9137
number of points rejected: 4994
PEP 1979 - 1981
CLASS: ALL

AVERAGE ICE CONCENTRATION [per liter]

CLOUD TOP TEMPERATURE [°C]
Cooper, 1986

for cases where ice concentration can be attributed to nucleation
Fig. 5. Plot of the phase ratio (the proportion of water to ice) in cloud against temperature from 11 frontal flights. Each point represents the average over a 7-min horizontal leg in cloud. Crosses indicate clouds in continental air masses and squares indicate clouds in maritime air masses. The dotted line is the best fit to the data for continental clouds and the dashed line for maritime clouds. The solid line is the current parameterization in the U.K. Meteorological Organization Atmospheric GCM (courtesy S. Moss and D. Johnson).

Fig. 6. Observations of ice crystal concentrations (no. L^{-1}) as a function of cloud top temperature. The ice nucleus spectrum is from Fletcher (1962), the Australian data from Mossop (1968) and King (1982), the Spanish data from Vali et al. (1988), and the Colorado data from Cooper and Saunders (1980) are shown as linear fits to the data. Observations from Hobbs and Rango (1985) over the Pacific Northwest of the United States are shown as open dots, and observations from the Australian Winter Storms Experiment (J. Jensen 1994, personal communication) are shown as closed dots.
3. THE GRAUPEL STORY.

• The importance of riming has been recognized from Wegener and Findeisen on, but primarily in connection with snow.

• Focus on hail damage reduction led to studies of riming in more detail.
NE Colorado summer Cu con.

- No evidence for coalescence process.
- Particles collected in clouds (sailplane) are graupel. Often observed at the ground as well.
- Evidence for prior vapor growth is infrequent but has been found.
- No large frozen drops centers in hail.
- First echo heights are above 0°C level, or are at melting band. Rough quantitative agreement between measured echo intensity and reflectivity calculated for observed graupel sizes and concentrations.
Observations consistent with the Knight et al. conclusions were obtained by Krauss et al. in S. Africa, and perhaps others.

The main question remaining is the origin of the ice crystals, or frozen cloud droplets, that start the graupel growth.
4. THE MULTIPLICATION SUCCESS.

Prologue:

By the mid-fifties, the importance of ice particles to precipitation formation is well accepted and the search is on to discover how many ice particles get initiated, and how, in different cloud types in relation to other parameters like liquid water content, droplet concentration, etc.
"It has been suggested that the explanation lies in some process of self-multiplication (e.g. splintering) by which a large number of ice crystals could build up from very few parent crystals."

Murgatroyd and Garrod, 1960
Ice pellets captured via a tube leading into the aircraft, when melted and refrozen are found to freeze at temperatures 6–10°C colder than the cloud top temperature. This casts doubt on freezing nucleation as the cause of ice initiation.

Hoffer and Braham, 1962

Koenig (1963) concludes that some chain reaction process, propagated by the formation of satellite ice particles during the solidification of water drops, may be responsible for the observed concentrations of ice particles much in excess of nuclei concentrations.
-6°C cloud top

lead foil impactor
>250 um particles

liquid and ice
liquid only

same sizes

all ice

345 sec

475 sec

Koenig, 1963
Measured ice nucleus concentrations much lower.

Pre-activation of nuclei

Accumulation of crystals in time.

Multiplication: splinters from drop freezing or electric effect associated with riming (both controversial).

Mossop, Ruskin and Heffernan, 1968
In the temperature range $-4 \text{ to } -10 ^\circ\text{C}$, high concentrations of small columns are found in association with graupel.

Ono 1971, 1972
Fig. 2 Production of secondary ice particles by rimeing as a function of temperature at a target velocity of 2.7 m s\(^{-1}\). Different symbols indicate different days. The curve was drawn by averaging the points over narrow temperature intervals.

drops > 23 \(\mu\text{m} \text{ (cm}^{-3}\text{)}\) produced per second plotted against the concentration of drops larger than 23 \(\mu\text{m}\) in diameter in the cloud through which the rimeing rod was moving. The temperature was \(-4.7^\circ\text{C}\) and the rimeing rate was approximately the same in all experiments. The line of best fit by the least-squares method is shown.

Hallett & Mossop, 1974

\(-4.7^\circ\text{C}; \text{ rimeing rate roughly the same}\)
Rime–splintering story wrapped up:

Hallett, Sax, Lamb and Murty (1978) in Florida demonstrate the graupel to needle sequence.

Harris–Hobbs and Cooper (1987) in Montana show quantitative agreement between the rate of generation of secondary particles and predictions based on laboratory findings.
Other secondary ice generation processes:

Hobbs & Farber, 1972 – crystal fragmentation
Vali, 1980 – rime fragmentation
electric effects, shock waves .....  

*There is ample evidence for unexpectedly high ice concentrations in many situations where the Hallett-Mossop mechanism is not active.*
5. CLOUD SEEDING PROSPECTS:

• Creation of numerous ice crystals in supercooled clouds is clearly possible. This confirms the basic Bergeron concept, and is consistent with the paucity of ice nuclei of comparable activity to those of the artificial nuclei.

• Precipitation on the ground is not a sure consequence of the creation of more ice crystals at some point in the cloud. This can be due to insufficient cloud volume getting seeded, the timing of seeding not being optimal, other processes out-competing the ice created by seeding, etc.

• The interplay of cloud seeding with studies of ice initiation in undisturbed clouds is beneficial.

• Industrial, or other well-defined sources, which create cloud glaciation deserve more attention.
6. OTHER ISSUES:

- **Ice frequency** vs. cloud type and other parameters is depicted by a large body of observations, but there are few repeated and generalizable patterns. Even definitions vary a great deal.
  
  Schemenauer and Isaac  
  Nevzorov  
  Rangno and Hobbs - OSCIP

- **Phenomenology** of ice particles is also well advanced.

- **Cloud dynamics** frames all ice initiation studies.

- **Aerosol physics and chemistry**, and perhaps air chemistry, provide useful characterizations of cloud input, but forward links to ice formation are teneous at this point.
CHALLENGES AND RESPONSES (from the observational point of view)

• May be still missing some fundamental process of ice nucleation. Theories are of limited help at this point. ➔ Aerosol physics and chemistry. Laboratory work.

• Time lapse between nucleation and an observable result (measurable ice particle) introduces the ambiguities of growth history through imperfectly known condition and cloud motions. ➔ Cloud physics framework.

• Conditions at cloud/clear air interface are difficult to define on the scale that may be determinant for ice nucleation. ➔ Finescale observations.

• Distinction between primary and secondary generation mechanisms may be hard to draw in some cases. ➔ Clarification of secondary processes.