



Nuclea	tion of the ice phase	
	<ul> <li>Homogeneous freezing of a pure liquid drop occurs when statistical fluctuations of the molecule arrangement of water produce a stable, ice-like structure that can serve as an ice purclus (IN)</li> </ul>	
	<ul> <li>Homogeneous nucleation depends on the size of the stable nucleus and the probability of occurrence of embryonic IC by random rearrangement of water molecules.</li> </ul>	
	<ul> <li>These quantities depend on the surface free energy of a crystal/liquid interface (analogous to surface tension at a liquid/vapor surface). The value of the surface free energy is not known accurately (~ 0.02 N/m).</li> </ul>	
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Nuclea	tion of the ice phase
	• IC appear in cloud in appreciable numbers when T < - 15°C, signifying heterogeneous nucleation. Water in contact with most materials freezes at T > -40°C, and deposition occurs on most surfaces at supersaturation and supercooling less than the homogeneous nucleation values $\rightarrow$ the nucleation of ice in supersoled water of a supersaturation environment is aided by the presence of foreign surfaces of suspended particles.
	<ul> <li>The foreign material provides a surface or substrate on which water molecules can impinge, stick, bond together, and form aggregates with an ice-like structure. The larger the aggregate, the more likely it is to be stable and continue to exist.</li> </ul>
Atmosphären- Physik Marc Wüest IAC <b>ETH</b>	<ul> <li>The probability of heterogeneous nucleation of freezing or deposition depends strongly on the properties of the substrate material as well as on the supercooling and supersaturation. The more tightly-bound the water molecules are to the substrate, the grater will be the probability of ice nucleation. The more closely the crystal structure of the substrate resembles that of an IC plane, it will increase the chances of ice nucleation.</li> </ul>

Nucleat	tion of the ice phase
	<ul> <li>Supercooled clouds in the atmosphere develop and exist in the presence of vast numbers of aerosol particles, a small fraction of which serve as IN at temperature considerably warmer than the -40°C threshold for homogeneous freezing:</li> </ul>
	<ul> <li>Ice may form directly from the vapor phase on suitable deposition nuclei.</li> <li>three modes of activation are recognized for freezing nuclei: <ul> <li>a) IN serve first as centers for condensation, then as freezing nuclei</li> <li>b) IN promote freezing at the instant they come into contact with a supercooled droplet</li> <li>c) IN cause freezing after becoming embedded in a CD.</li> </ul> </li> </ul>
Atmosphären- Physik	<ul> <li>A given IN might nucleate ice in different ways, depending on the ambient conditions and its history in the cloud.</li> </ul>
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Nuclea	tion of the ice phase	
Atmosphären- Physik Marc Wüest IAC <b>ETH</b>	<ul> <li>The relative importance of different freezing modes in the atmosphere has not been established. It is also difficult to distinguish between deposition and freezing nucleation when ice nucleates on an insoluble surface in an environment supersaturation with respect to water (a) and (b).</li> <li>Even conditions below water saturation nucleation need not imply deposition, because the nuclei may contain soluble components. The soluble material may nucleate the liquid phase below water saturation and enable the insoluble material to nucleate ice by freezing. Because of the confusion between mechanisms, one often speaks of "ice nucleation" as the phenomenon instead of being more specific. Likewise, the atmospheric particles serving as nucleation centers can most safely be referred to as "ice nuclei".</li> <li>I.e. ~ ice nucleation is very uncertain both theoretically and experimentally</li> </ul>	: 8

Experi nuclea	ments on heterogeneous ice	
	<ul> <li>The nucleating properties of small particles are studied by introducing them into cloud chambers with controlled supercooling and supersaturation. The conditions are noted where the onset of nucleation occurs. (IC are usually discernable even in the presence of liquid by the scintillation of light scattered from a strong beam). It's</li> </ul>	
	not possible to distinguish between a deposition event and a condensation event followed by freezing.	
	<ul> <li>Other experiments consist of adding finely divided material to supercooled, purified water and noting the threshold temperature for freezing. The table below summarizes the threshold ice-forming temperatures of certain pure and natural substances.</li> </ul>	
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### Experiments on heterogeneous ice nucleation

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		TABLE 9.1. 7	"emperatures	at which diff Houghton,	erent substances 1985)	nucleate ice. (From
Silver iodide m	natches ice closest nd also has a		Cry lattice di	stal mension	Temperature	
relatively warr	n nucleating	Substance	a axis (Å)	caxis (Å)	ice (°C)	Comments
temperature.	However, lattice	Pure substances				
dimensions ar determining n several organi have a well-de structure. Son lattice matchir binding, and lo with ice accou nucleating abil Still an open fi	e not the only factor ucleating ability, as c materials do not fined crystal ne combinations of ng, molecular ow interfacial energy nts for the lity of a substance. ield	Ice AgI AgI CoS CoS CoS CoC HgL, Ag,S CdI J J J Minerals Vaterite Kaolinite Volcanice ash Halloysite Vermiculite Vermiculite Cinnabar	4.52 4.58 4.54 3.80 4.65 4.36 4.20 4.24 4.78 4.12 5.16 5.16 5.34 4.14	7.36 7.49 6.86 16.43 5.11 12.34 9.50 6.84 9.77 8.56 7.38 	0 -4 -6 -7 -7 -8 -12 -12 -12 -12 -13 -13 -15 -16	Insoluble Slightly soluble Insoluble Insoluble Insoluble Soluble Soluble Soluble Soluble
		Organic materials				
Atmosphären- Physik Marc Wüest IAC <i>ETH</i>		Testosterone Chloresterol Metaldehyde β-Naphthol Phloroglucinol Bacterium Pseudomonas Syringae	14.73 14.0 8.09	11.01 37.8 17.8 	-2 -2 -5 -8.5 -9.4 -2.6	(Bacteria in leaf mold)

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# Atmospheric ice nuclei .. are studied in cloud chambers and on filter systems, into which samples of air are drawn. ٠ In a cloud chamber the sample is cooled down to a controlled temperature and a cloud is formed by adding sufficient water vapor. Most time, an optical system is used to count the no of ice crystals that form as a function of degree of supercooling. However, no information about nucleation mode and nucleus size can be retrieved that way. ٠

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The second method consists of collecting aerosols by drawing the air sample through filter paper with known pore sizes. The particulates trapped on the filters are then introduced to an environment suitable for IC development and observations are made of the number of crystals that form on a substrate. That way, information about nucleus size can be obtained, but not about nucleation mode.

Atmos	oheric ice nuclei	
	<ul> <li>Reported IC concentration at temperature -20°C range over many orders of magnitu supercooling increases, so does the nucle usually a rapid rate. IN concentration also supersaturation. Furthermore, there is ev nucleation events do not occur immediate long exposure of the nucleus to supercool Accordingly, only a fraction of the IN actu an air sample may be activated during th experiment. IN content of the air is a high quantity.</li> </ul>	es from -15°C to ide. As the us count at o increases with idence that some ely, but require a led conditions. ally present in e time of an hy variable
	<ul> <li>Fletcher (1962), reviewing the data to the a typical concentration 1 nucleus per lit -20°C, increasing by a factor of 10 for ea additional cooling.</li> </ul>	at time, gave as e <mark>r of air at T</mark> = ch 4°C of
Atmosphären- Physik Marc Wüest IAC <b>ETH</b>	<ul> <li>This exponential dependence of IN on sup accepted as typical, but it is recognized th any given location and time can be at leas magnitude above or below.</li> </ul>	percooling is nat the count at st an order of

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# Atmospheric ice nuclei Taking 10<sup>4</sup> cm<sup>-3</sup> as a typical AP concentration, we see that 1 nucleus per liter active at -20°C is only 1 IN in 10<sup>7</sup>. Separating and identifying such rare particles is not an easy task. Much evidence points to clay minerals, especially kaolinite as a major component of atmospheric IN. This is a common material found in many soil types. Snowflakes analysis yields that they contain kaolinite with sizes ranging from 0.1 µm to 4 µm. What is not clear is how kaolinite could explain the occurrence of ice in clouds warmer than -9°C, as sometimes observed. Another source of IN has been revealed by the discovery what the besteries in decaying elect large material case between the source of the second secon

Atmosphären Physik Marc Wüest IAC**ETH**   Another source of IN has been revealed by the discovery that the bacteria in decaying plant leaf material can be effective nucleants at warm temperature. Bacterium pseudomonas syringae serves as IN at -1.3°C, although its nucleating ability is a rare and changeable property. Thus, more work needs to be done here.

Atmosp	oheric ice nuclei	
	<ul> <li>Meteoric material is another source, found, because of apparent correlation between extreme rainfall events and meteor showers and more recently from experiments. However, more evidence points to a terrestrial source at ground, as even at the South Pole, the particulates in snow flakes are found to be clay minerals. Also IN tend to decrease with altitude over continents and at coastal sites measurements indicate more nuclei in air from trajectorie over land.</li> </ul>	s
	<ul> <li>In summary, while supersaturation with respect to water exceeding 1% are extremely rare, supercooling of liquid water to -15°C or colder is not uncommon. Also, probably more than one material serves as IN depending on temperature, humidity and distance from sources.</li> </ul>	
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The ice	phase in clouds				
	<ul> <li>Occurrence of IC in clouds is related to cloud type, temperature and cloud age. Overall observations confirm that the colder cloud top temperature, the greater is the likelihood that some IC are present along with the supercooled CD. Although essentially no clouds with tops as cold as -20°C are ice-free, ice anywhere in a cloud is unlikely unless the top extends to -5°C or colder.</li> </ul>				
	<ul> <li>Ice is more common in decaying Cu than in newly developing clouds, and is probably more common in stratiform clouds than in Cu with the same cloud top temperature.</li> </ul>				
Atmosphären- Physik Marc Wüest IAC <i>ETH</i>	<ul> <li>Measured concentration of IC range from the lower limit of detection (0.01/l) to 100/l. Concentrations are high in Ci, and still higher in ice fogs, which develop under extremely cold arctic conditions. The most perplexing question in ice microphysics is to explain the vast discrepancies, something accounting to a factor of 10<sup>4</sup>, between observed crystal concentration and the measured concentration of IN in ambient air.</li> </ul>				





The ice	phase in clouds			
	<ul> <li>Secondary IC: (a) fracture of IC, and (b) splintering of freezing drops. Often IC fragments are included with falling snow. These are probably produced when dense graupel particle overtake and collide with fragile, slower- falling dendritic crystals. Not much is known about (a).</li> </ul>			
	<ul> <li>Another mechanism of ice multiplication, thought to be very effective in the right conditions, is the production of secondary particles when supercooled drops of the appropriate size and temperature are captured by graupe particles.</li> </ul>			
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# Diffusional growth of IC



This figure shows supersaturation  $S_i$  for water saturation. Water-saturated cloud has high supersaturation with respect to ice and is a favorable environment for rapid growth by diffusion and deposition. The environment will remain favorable for crystal growth as long as CD are available to evaporate and maintain the vapor pressure at equilibrium relative to water. If the CD eventually disappear, by evaporation or freezing, the saturation ratio will decrease to equilibrium relative to ice.

In the













Some r	numbers			
	TABLE 7.1. Values of Dy of Thermal Conductivity K sion D of Water Vapor it			Coefficient tt of Diffu- n, 1985)
	<i>T</i> (°C)	$\mu$ (kg m <sup>-1</sup> s <sup>-1</sup> )	$K(Jm^{-1}s^{-1}K^{-1})$	$D(m^2s^{-1})$
	-40	$1.512 \times 10^{-5}$	$2.07 \times 10^{-2}$	$1.62 \times 10^{-5}$
	-30	$1.564 \times 10^{-5}$	$2.16 \times 10^{-2}$	$1.76 \times 10^{-5}$
	-20	$1.616 \times 10^{-5}$	$2.24 \times 10^{-2}$	$1.91 \times 10^{-5}$
	$-10^{-10}$	$1.667 \times 10^{-5}$	$2.32 \times 10^{-2}$	$2.06 \times 10^{-5}$
	0	$1.717 \times 10^{-5}$	$2.40 \times 10^{-2}$	$2.21 \times 10^{-5}$
	10	$1.766 \times 10^{-5}$	$2.48 \times 10^{-2}$	$2.36 \times 10^{-5}$
	20	$1.815 \times 10^{-5}$	$2.55 \times 10^{-2}$	$2.52 \times 10^{-5}$
	30	$1.862 \times 10^{-5}$	$2.63 \times 10^{-2}$	$2.69 \times 10^{-5}$
Atmosphären- Physik	Note: The D is propy pressure pressure p (100/p).	e tabulated values of $\lambda$ ortional to $\mu/\varrho$ , it fol for a given temper $\rho$ (kPa), the tabulated	D are for a pressure of $10$ lows that D is inversely ature. To obtain D fo d value must therefore b	0 kPa. Because proportional to or an arbitrary be multiplied by
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Diffusio	onal growth of IC
	• Without explaining the shapes of crystals, the classical theory can nevertheless be used for approximate growth calculations if the crystal habit is specified.
	<ul> <li>What is required is an analytical expression for the capacitance of the crystal and empirical relations between the dimensions of the crystal and its mass.</li> </ul>
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## Further growth by accretion



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Furthe	r growth by accretion
	<ul> <li>Difference in fall speed: fastest are graupel particles (which are not really crystals, but aggregates of frozen dropletc). The rimed structures and dry spow flakes fall</li> </ul>
	at about 1 m/s, but all pure crystal types fall slower than 1m/s. Empirical fall speed for graupel:
	$u = 343D^{0.6}$
	<ul> <li>where u is in cm/s and D, the diameter of the sphere which just circumscribes the particle, in cm. Empirical fall speed for snow flakes:</li> </ul>
	$u = kD^n$
Atmosphären-	<ul> <li>where D is the melted diameter. With D in cm and u fall</li> <li>mend is strice baselables found for deadvices (w. 160)</li> </ul>
Physik Marc Wüest IAC <b>ETH</b>	and $n = 0.3$ , and for columns and plates $k = 234$ and $n = 0.3$ .





Furthe	growth by accretion		
	<ul> <li>Mass and size of different i by empirical formulas of the m = aD<sup>b</sup></li> <li>where</li> </ul>	forms of IC are una form:	sually related
	Crystal type	a	b
	Graupel Thin hexagonal plate Stellar crystal Planar dendrite Needle	$\begin{array}{c} 6.5\times10^{-2}\\ 1.9\times10^{-2}\\ 9.4\times10^{-4}\\ 3.8\times10^{-4}\\ 2.9\times10^{-5} \end{array}$	3 3 2 2 1
Atmosphären- Physik	<ul> <li>where D is the major linea</li> </ul>	r dimension of th	e crystal in
Aarc Wuest AC <b>ETH</b>	centimetres and $m$ in g.		



Further	r growth by accretion	
	<ul> <li>Growth equation, leading t drops:</li> </ul>	o graupel, analogous to rain
	$\frac{dm}{dt} = \overline{E}M\pi R^2 u(R)$	where <i>M</i> is cloud liquid water content
	• similar for aggregation:	
	$\frac{dm}{dt} = \overline{E}M\pi R^2 \Delta u$	where <i>M</i> is frozen water density
	<ul> <li>where Δu is the difference and the IC, essentially a co and u(IC) ~ 0.4 - 0.5 m/s)</li> </ul>	in fall speed of the snowflake onstant (u (snowflake) ~ 1 m/s
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Furthe	growth by accretion
	<ul> <li>Those growth equations are only rough approximations for actual growth process, but fit observations of graupel and snowflakes well.</li> </ul>
	<ul> <li>Fundamentally snowflakes develop, because a few crystals, which formed and grew by diffusion, become larger than their neighbors, either by enhanced diffusional growth or by chance collisions with other IC or supercooled CD. Thereafter they're in a favorable position to grow by the sweep-out process.</li> </ul>
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IC proc	ess versus coalescence	
	<ul> <li>Condensation-diffusion alone cannot explain raindrop formation in realistic time. This process is more effective for IC than for CD, because the vapor in the cloud is often at equilibrium relative to water and hence supersaturated with respect to ice. Therefore light precipitation can occur as individual crystals, indicating that aggregation or accretion never took place (clear sky precipitation). Thus, some drizzle or light rain has its origin in unaggregated crystals, which melted before reaching the ground.</li> </ul>	
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