

Colloidal meteorological processes in the formation of precipitation

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Original citation:

FINDEISEN, W., 1938: Die kolloidmeteorologischen Vorgänge bei der Niederschlagsbildung. – Meteorol. Z. 55, 121–133.

Suggested citation:

FINDEISEN, W., 1938: Die kolloidmeteorologischen Vorgänge bei der Niederschlagsbildung (Colloidal meteorological processes in the formation of precipitation). – Meteorol. Z. **55**, 121–133. (translated and edited by VOLKEN, E., A.M. GIESCHE, S. BRÖNNIMANN. – Meteorol. Z. **24** (2015), DOI:10.1127/metz/2015/0675).

Abstract

This paper is the edited translation of the paper by WALTER FINDEISEN "Die kolloidmeteorologischen Vorgänge bei der Niederschlagsbildung" (Colloidal meteorological processes in the formation of precipitation) that was published 1938 in the Meteorologische Zeitschrift **55**, 121–133.

Abstract [Original, translated abstract]

The microphysical processes of rain formation are discussed based on new insights. All fundamental phenomena of rain formation are explained. Important new insights arise concerning practical meteorology.

Keywords: Findeisen-Bergeron process, cloud microphysics, precipitation formation

Superscript numbers indicate original footnotes (translated at the bottom of the page), E... numbers indicate editorial endnotes (at the end of the article), square brackets [] indicate editorial comments in the text.

After it was found that the solid state of water, the "ice phase", is of major importance for the formation of precipitation and in particular for rain formation, colloidal meteorological research was assigned several new tasks. The processes in water clouds (i.e. clouds consisting solely of liquid water), which used to be the main focus of colloidal meteorological research, do not suffice to explain rain formation. Bergeron¹ was the first to point this out in 1933; he assumed that all substantial precipitation events are caused by the ice phase, where the existence of ice crystals in water clouds plays the decisive role. The colloidal instability of clouds, including both ice crystals and supercooled droplets side by side, has already been discussed by A. WEGENER² quite some time ago. The assumption made by BERGERON has been confirmed in recent years by aerological surveys

by W. PEPPLER³ and by the findings of the author from data collected during several hundred aerological flights. From this work it can be established that every precipitation event of at least moderate intensity, and in particular every event with larger rain droplets, is caused by ice crystals.

The observations made from aircrafts have shown that the presence of different aggregate states of water in clouds at temperatures below 0 °C does not follow simple laws. It could only be shown by observations that, for temperatures only a few degrees below zero, water clouds usually occur slightly more often than ice clouds, and that for temperatures below -10 °C, ice clouds strongly dominate. However, water clouds have been observed even below -20 °C.⁴

A regularity for the seemingly unintelligible pattern of the aggregate state in the formation of clouds can be found when one assumes two entirely different kinds of nuclei, namely [cloud] condensation nuclei and sublimation nuclei [ice condensation nuclei]. A. WEGENER has

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¹T. BERGERON, Mémoire présenté à l'Association de Meteorologie de l'U.G.G.I., Lissabon, 1933.^{E1}

²A. WEGENER, Thermodynamik, Leipzig 1911^{E1}; A. and K. WEGENER, Physik der Atmosphäre, Leipzig 1935.^{E1}

³W. PEPPLER, Beitr. Z. Phys. D. fr. Atm. 23, 275, 1936.^{E1}

⁴Already 50 years ago, CH. RITTER (Annuaire de la Société Mét. de France 33, 261, 1885 and 35, 362, 1887) stated the existence of supercooled water droplets at temperatures as low as that.

already considered such an existence of two nuclei types as probable. Here, we take this assumption as the starting point to gain new insights about the colloidal meteorological processes in clouds. It should be mentioned in advance that these insights have been examined in detail in light of the observations to-date, and that there is in no way any contradiction between theory and phenomena observed in nature. Based on the hypothesis of the existence of two kinds of nuclei, it is possible to easily explain the varied manifestations of ice and water clouds. Above all, it is now possible to understand precipitation formation processes much more clearly than before.

1 Condensation nuclei and sublimation nuclei

Condensation nuclei are almost always particles of hygroscopic substances that absorb some water even when relative humidity is still below 100 %. In other words, they are liquid before cloud formation begins. Some condensation nuclei are even always liquid, for instance those consisting of sulphuric acid. Particles of this kind probably account for the greatest number of atmospheric condensation nuclei. They are generated in large amounts during artificial and natural combustion processes.⁵

Since the condensation nuclei are, in any case, liquid at high relative humidities (they are solutions of hygroscopic substances that strongly reduce the freezing point), the sublimation⁶ of water, i.e. ice formation, cannot occur even with temperatures of -20 °C. Evidently, only solids are capable of sublimation, which means that, in contrast to the condensation nuclei, the atmospheric sublimation nuclei must be solids.

The chemical nature of the sublimation nuclei and their sources are still unknown. One might assume that the sublimation nuclei are of cosmic origin, and are supplied by meteorites that are vaporized when entering the atmosphere. In fact, the amount of stardust would suffice to cover the entire demand for sublimation nuclei. However, it seems that the sublimation nuclei are of terrestrial origin. This is indicated by the decreasing number of sublimation nuclei in the atmosphere with height; it is proven by the fact that the highest ice clouds (i.e. cirrus clouds) only contain very few particles. One can assume that sublimation nuclei partly consist of quartz particles or fine grains of sand that are suspended in the atmosphere. The hexagonal shape of the quartz crystal is particularly suited for the adsorption of ice crystals since their formation occurs hexagonally as well; it reduces vapour pressure similarly to the hygroscopic condensation process. This fact, however, which was already pointed out by A. WEGENER (l.c.), may possibly be of only limited significance for atmospheric sublimation. Just as hygroscopicity only has a minor influence on the critical size of small condensation nuclei,⁷ the effectiveness of small sublimation nuclei may be only slightly increased by a favourable shape. In the polar regions, a considerable amount of sublimation nuclei probably enter the atmosphere from the snow surface. Many snow crystals get mobilised and airborne due to the uplift and suspension of snow, of which the smaller crystals will evaporate and leave a suspension of sublimation nuclei.

By observing the beginning of sublimation in supercooled water clouds, the size of sublimation nuclei can be roughly estimated. According to the records of the author, sublimation often occurred at about -10 °C, and sometimes not until -18 °C. On the reasonable assumption that the air in clouds is water saturated, these temperatures indicate an ice supersaturation of 9 % and 16%, respectively. If, for lack of better knowledge, one assumes that the well-known formula by THOMSON^{E2} for the increase of vapour pressure at the surface of small particles can also be applied to sublimation processes, then the size of sublimation nuclei would be on the order of $r = 10^{-6}$ cm for the cases mentioned. This is the same order of magnitude as the condensation nuclei. The sublimation nuclei are certainly so small that they usually cannot serve as condensation nuclei. In atmospheric condensation processes, only low levels of water vapour supersaturation are reached, which are not sufficient for the condensation of insoluble particles on the magnitude of condensation nuclei. Thus, the sublimation nuclei remain unaffected when water clouds form.

The amount of condensation and sublimation nuclei is very different. After numerous observations by means of the common nuclei counters, the number of nuclei in atmospheric air varies between 1 and 10^6 /cm³. In the natural formation of water clouds, far fewer nuclei are involved than indicated by the nuclei counters, because much lower levels of water vapour supersaturation are reached in the atmosphere, meaning that droplet formation can only take place on larger and hygroscopic nuclei. The number of effective condensation nuclei in the atmosphere probably ranges mostly between 10^2 to at most 10^4 /cm³, and occasionally only 10 or less.

From the observation of ice clouds, which had consistently fewer particles compared to water clouds, it follows that the number of sublimation nuclei is significantly fewer than the number of condensation nuclei. In any case, this is true for the layers in which ice cloud formation can take place, which in winter already occurs at 1000 m altitude. The number of sublimation nuclei could range between about 10^{-2} to $10/\text{cm}^3$, and certainly varies greatly just as the number of condensation nuclei do. Observed weather phenomena suggest that there is often a lack of effective sublimation nuclei in the atmo-

⁵Chr. JUNGE, Gerlands Beitr. z. Geophys. 46, 108, 1935; Meteorol. Zeitschr. 1935, p. 467; W. Findeisen, ibid. 1937, p. 377.^{E1}

⁶The term "sublimation" generally refers to the transition of a substance from the gaseous state to the solid state and vice versa, but is used here only for ice formation from water vapour, in contrast to "condensation."

⁷W. FINDEISEN, Mikrophysik der Wolken (Teil I der "Physik der Wolken", T. BERGERON, W. FINDEISEN and W. PEPPLER, in preparation) Section A., a., 2.^{E1}

sphere, which can significantly alter weather dynamics. This is of utmost importance to weather analysis and forecast.

Not only the two mentioned types of nuclei need to be distinguished. Even nuclei of the same kind can be very different from each other, certainly regarding their size and probably also regarding their chemical nature and shape. It would be difficult to imagine a natural process that results in the suspension of equally sized nuclei in the atmosphere. The unequal size of condensation nuclei has already been demonstrated by JUNGE (l.c.). Accordingly, the particle size distribution of condensation nuclei can be described as a continuous mass spectrum. This is likely also true for the sublimation nuclei.

Currently, there is no instrument to determine the number, let alone the characteristics, of sublimation nuclei. The common condensation nuclei counters by AITKEN and SCHOLZ^{E3} do not only count all condensation nuclei (including nuclei that are ineffective in the atmosphere), but they also count sublimation nuclei, since these act as condensation nuclei in the presence of the high supersaturation values in the measuring chamber (about 300%). Therefore, colloidal meteorological research can draw only little benefit from the nuclei number studies carried out thus far. However, it is possible to design instruments that can investigate condensation and sublimation nuclei separately. Such studies would considerably enhance the understanding of cloud and rain formation and clarify the controversial questions presented by the above line of thought.

2 Formation of water clouds

Condensation processes in the atmosphere always proceed in the following way:

A steady increase of relative humidity is caused by a process such as adiabatic lifting, exchange, or emission. The relative humidity rises at a constant rate up to the value where condensation begins on the largest condensation nuclei. After this point, the relative humidity increases at a slowed rate since water vapour is absorbed by the nuclei.

In hygroscopic nuclei, this already holds true with low relative humidity values, and it is, for example, always the case for sulphuric acid nuclei. However, when relative humidity is below 100 %, the water uptake by nuclei is only minimal and, despite a large number of nuclei, insignificant for the water vapour content of the air. At first, the nuclei remain small because the concentration of the hygroscopic solution is sufficient to maintain the balance between the chemical lowering of vapour pressure (hygroscopicity) and the physically induced increase in vapour pressure (surface tension). This can be derived from the "growth curves" calculated by JUNGE (l.c.) for sulphuric acid nuclei.

For any condensation nuclei, there is a critical relative humidity above which the nucleus can continue to grow without a further increase in relative humidity. When the size of the nucleus increases, the decrease of the physically induced increase in vapour pressure at the nucleus' surface is equal to the decrease of the chemical lowering of the vapour pressure; at any later stage it is larger. This defines the physical transition from nucleus to droplet and from vapour to cloud. After the critical relative humidity has been reached, the condensation nuclei quickly grow into droplets, at which point the content of the hygroscopic substance and the surface tension are only of limited significance.

The critical relative humidity of any condensation nucleus depends on the amount and kind of its hygroscopic substance and can be determined arithmetically. For normally sized condensation nuclei (around 7×10^{-7} to 10^{-5} cm)⁸, it is between 106 and 101 % under the assumption of sulphuric acid being the hygroscopic substance.⁹

As soon as the relative humidity has reached the critical value for the largest and most hygroscopic nuclei and consequently droplet formation begins, a further increase in relative humidity is strongly inhibited. It stops entirely as soon as water vapour adsorption to droplets is equal to the water vapour released into the air, which would otherwise add to the relative humidity. From this point onwards, the condensation nuclei that have not yet been included in the condensation process stop to grow further, and new droplets are no longer formed. This is more likely to occur the greater the surface areas of the droplets are and the less constantly-releasing water vapour is present in the air; in other words, the slower the process of cloud formation takes place.

From this it follows that the number of the largest and most hygroscopic nuclei is of importance. If the number is small, a further increase of the relative humidity is not hindered, allowing for possibly numerous smaller condensation nuclei to participate in the process. On the other hand, if the number of large nuclei is great, then the total number of droplets formed remains relatively small under certain circumstances.

The significance of the rate of the cloud formation process is also easy to understand. If the relative humidity increases quickly, it reaches a high maximum value, leading to the inclusion of quite small nuclei and causing many droplets to form. For this reason, fog and stratiform clouds contain fewer droplets than cumulonimbi. During fog formation, relative humidity hardly rises above 100 %; the number of droplets usually amounts to around 100/cm³ compared with 100'000/cm³ condensation nuclei. However, during the artificial reproduction of cumulonimbi where an adiabatic expansion corresponding to 3.5 m/sec was applied, the maximum relative humidity amounted to between 103 to 112 %, and the number of droplets was around 10'000/cm³.¹⁰

⁸W. FINDEISEN, Mikrophysik der Wolken, l.c.^{E1}

⁹According to CHR. JUNGE, l.c.

¹⁰W. FINDEISEN, Gerlands Beitr. z. Geophys. 35, 295, 1932.^{E1}

Once the further increase in relative humidity in a cloud is halted due to condensation on droplets, relative humidity begins to decrease because the steadily increasing droplet surface area leads to more water uptake by the droplets and allows the surplus water vapour to be used up quickly. The value of the relative humidity that is eventually reached in the cloud depends on the number and size of the droplets as well as the condensation rate. Even in cumulonimbi with high vertical velocity, it corresponds to only little over 100 %. This has been discussed in detail elsewhere.¹¹

Since not all condensation nuclei are included in the condensation process simultaneously, but rather in succession due to their different characteristics, the resulting droplets are of different ages and also sizes. The older droplets were exposed to the condensation process for longer than the younger ones and could grow more strongly. However, even if the condensation nuclei had the same characteristics and the droplets were of the same age, size differences would result because the droplets are distributed in natural randomness throughout the space rather than having an equal distribution. Isolated droplets are favoured in the condensation process whereas the droplets located in clusters quickly absorb the water vapour surplus of the air and grow only slightly.

The unequal size of droplets in clouds exists permanently. The size differences continuously increase since the larger droplets grow at the cost of the smaller ones, whose surfaces have slightly higher vapour pressure. Still, the water transport induced in the process is only small. As a result of the different fall velocities of the differently sized droplets, a separation according to size occurs during the sedimentation process. However, the thickness of natural clouds is usually so great in comparison to the fall distance of the small cloud droplets that the development of layers with similar droplet sizes can hardly occur, particularly since the droplet sizes are not graded in groups, but rather have a continuous size distribution.

In fact, the occurrence of equally-sized droplets in clouds or fog has never been observed, and was only assumed by some researchers due to lack of knowledge of the real conditions. The assumption was not justified and led to wrong conclusions. Later, more detailed examinations showed that fog and clouds are entirely inhomogeneous, containing droplets of a variety of sizes.¹²

The variability in droplet size, as well as the number and size of the droplets are the most important factors for the rain formation process of clouds; their origin has hereby been discussed.

3 Formation of ice clouds

The formation of ice clouds normally only occurs at temperatures of at least several degrees below zero; at higher temperatures only water clouds form. This is known from experience and has been confirmed by aerological observations. Additionally, it is closely related to what has been said about condensation and sublimation nuclei.

The freezing point does not represent a limiting threshold for the formation of water clouds. They can form and continue to exist even at very low temperatures, and the supercooled status of the suspended water droplets does not change this. As has already been mentioned, water clouds have been observed even at temperatures below -20 °C. However, the formation of water clouds does become more and more unlikely with lower temperatures, since sublimation on sublimation nuclei (i.e. ice cloud formation) takes place more easily than condensation on condensation nuclei in these conditions. Generally, sublimation requires a lower relative humidity than condensation. The relative humidity of an even ice surface is 100 % for 0 °C, but only 91 % for -10 °C and only 82 % for -20 °C, so that, for instance, at -10 °C and 100 % relative humidity there is already 9% supersaturation with regard to an ice surface whereas there is only saturation with regard to a water surface. If, in such a case, sublimation nuclei that do not require more than 9% supersaturation are available, then sublimation begins before droplet formation can start. At this point, only an ice cloud can form because the relative humidity normally decreases quickly and remains below 100 % due to ice crystal growth after sublimation begins.

Ice cloud formation at a given temperature is therefore only a question of the sublimation nuclei. As already mentioned above, the content of sublimation nuclei in the air certainly varies considerably. This fact explains the observation that ice clouds frequently form at about -10 °C, but usually do not form at much lower temperatures even when all thermodynamic conditions for cloud formation are fulfilled. The varying number of effective sublimation nuclei in atmospheric processes is more important than the varying number of effective condensation nuclei because 1) the numbers of condensation nuclei are almost always sufficient and 2) the absence of water cloud formation is not as decisive for rain formation as the absence of ice cloud formation.

What has been said about the formation of water clouds can also partly be applied to the formation of ice clouds. The nuclei available for the sublimation process are certainly unequal and, depending on their size and shape, correspondingly suitable for ice crystal formation. Here again, a selection of nuclei takes place in the ways discussed earlier. The transition from nucleus to ice crystal and from mist to cloud is simpler for sublimation than for condensation; the nuclei remain unaffected until the beginning of crystal formation.

Ice crystals grow into different shapes depending on the degree of ice supersaturation. At high supersaturations, skeletons (star-shaped) and single prismatic crystals (sphaerokristalle, also known as reifgraupeln) form,

¹¹W. FINDEISEN, Beitr. z. Phys. D. fr. Atm. 20, 157, 1933.^{E1}

¹²W. FINDEISEN, Gerlands Beitr. z. Geophys. 35, 295, 1932; V. HAGEMANN, ibid. 46, 1936.^{E1}

while at low supersaturation proper crystals form.¹³ Changes in the degree of supersaturation result in corresponding changes of the crystal shapes.

Ice clouds differ greatly from water clouds with regard to particle number and size. As a result of the small number of sublimation nuclei, the number of particles in ice clouds is often very small, probably on the magnitude of 1/cm³. The author has repeatedly observed ice clouds that had a visibility of several kilometres. According to TRABERT's formula^{E4}, this particle size (diameter of 0.3 mm) indicates a particle number of about 1/200 per cm³. However, these clouds were already very thinned out due to the precipitation of particles ("ageing").

The particle sizes found in ice clouds usually considerably exceed those found in water clouds. The elements of ice clouds can grow more quickly because there is a larger amount of surplus water in the air for each particle due to the fact that ice clouds contain fewer particles than water clouds. The fall velocity of ice crystals is usually considerable due to their size, with values commonly 1 m/sec or more. This becomes evident in the diffuse bottom layers of ice clouds, and is particularly striking in cirrus clouds that are characterized by distinct virga. The fact that such large particles can be formed here, despite the minimal amount of water vapour available for sublimation at such low temperatures, shows how extraordinarily few sublimation nuclei exist in the upper troposphere, and proves the decrease of sublimation nuclei with altitude.

4 Condensation in ice clouds

It is understandable that the small number of ice crystals present in older ice clouds will hardly result in the sublimation of similar amounts of water vapour to that released, for instance, during the lifting and adiabatic cooling of air. In such a case, the relative humidity in a cloud increases considerably, leading to the inclusion of untouched nuclei in the renewed process of sublimation or condensation. If most of the sublimation nuclei have been used, droplet formation on condensation nuclei takes place. A "secondary" water cloud forms within the ice cloud. In the beginning, this cloud still contains some ice crystals, but these quickly precipitate out for reasons that will be mentioned later on.

Condensation in ice clouds can actually be observed quite often if convection occurs in an old ice cloud. In the ice stratus cloud, which almost looks like mist due to its low density and mostly can only be recognized as a cloud because of its magnificent halo, several smaller, dense cumuli clouds form that often consist entirely of supercooled water soon after their formation. This is expressed in old, thin altostrati, close to the top boundary where convection results from wave formation or increasing instability. Depletion of crystals occurs particularly near the top of ice clouds due to sedimentation. The developing water clouds, which are called altocumuli, are often no thicker than 100 m. They are clearly visible when observed or photographed in the light of a strong subsun, which seems to be unnaturally located between the altocumuli, but in fact develops entirely in the altostratus. The condensation of ice clouds also explains some optical phenomena, which suggests the simultaneous existence of solid and liquid cloud elements.¹⁴

In the affected areas, ice clouds are turned into water clouds based on the condensation process described above. The process obviously does not have much impact on rain formation because precipitation is enhanced only for the few ice crystals present in the thin ice cloud.

5 Sublimation in water clouds

In contrast, the transformation of supercooled water clouds into ("secondary") ice clouds is of utmost significance for rain formation. It is initiated when sublimation begins on sublimation nuclei, which are usually present in an unaltered state in water clouds. It is indeed true that the sublimation nuclei partly disappear due to adsorption onto droplet surfaces, but this happens only very slowly. This process is probably only significant in old water stratus clouds in which the sublimation nuclei may be almost entirely missing for this reason. Such clouds can no longer change in the way described, and rain formation accordingly proceeds differently than in normal clouds with sufficient sublimation nuclei. The transformation of supercooled water clouds into ice clouds does not happen due to the spontaneous freezing of droplets; this may be safely assumed according to the current state of research.

In most cases, sublimation in supercooled water clouds takes place in rising air streams (e.g. in cumulus clouds) in which air temperature constantly falls. With a constant relative humidity (which usually is quite exactly 100% in dense water clouds, see above), the preconditions for sublimation become increasingly favourable due to the continuous temperature decrease. Supersaturation with regard to the ice surface increases steadily and finally reaches the characteristic threshold for sublimation on sublimation nuclei. At this point, sublimation begins in a similar way as in the formation of new ice clouds, but relative humidity remains around 100 % in the beginning and decreases more slowly due to the presence of droplets. Consequently, a greater number of less suitable sublimation nuclei are used than in the new formation of ice clouds. Furthermore, the ice crystals grow more rapidly due to the continual ice supersaturation. On the other hand, water droplets evaporate to the same degree as relative humidity decreases. It changes from 100 % to the value of ice saturation at the respective temperature, slowly at first, but more and more quickly as the ratio between ice surface and water surface increases. Naturally, the smaller droplets disappear more quickly than the larger ones, but even the large

¹³More detailed in A. and K. WEGENER, l.c. Met. Z. 1938.

¹⁴e.g. E. FRANKENBERGER, Meteorol. Zeitschr. 52, 185, 1935.^{E1}

droplets with a diameter of 10^{-2} cm disappear within about 10 sec as soon as relative humidity has roughly approached the value of ice saturation.¹⁵ Thus, the transformation process takes place very quickly.

The transformation can only take place if the numbers of sublimation nuclei are sufficient for maintaining the entire amount of water, which is first present in the form of droplets, suspended as ice. If only few sublimation nuclei are present and therefore only few ice crystals can form, they grow so quickly within a very short period of time that their fall velocity is soon sufficient to overcome the rising air stream; they fall out and become insignificant to the further development of the cloud. Under such conditions, condensation finally begins. In fact, this does not appear to happen often. Nevertheless, the number of effective sublimation nuclei always seems to be considerably smaller than the number of droplets, so that the ice crystals become larger than the droplets were. This also happens because the water vapour amount, which is released into the air during the transformation from water saturation to ice saturation, is consumed by the ice crystals.

There are also other forms of sublimation that occur in water clouds. If suitable sublimation nuclei enter a strongly supercooled water cloud, which may be entirely motionless, they form single ice crystals that grow quickly and fall out. This phenomenon may occasionally be observed in winter with low stratus during which, for some reason, there are no sublimation nuclei present with sufficiently suitable characteristics for a transformation. In such cases, sublimation nuclei sometimes enter the supercooled water cloud from above. They grow quickly here due to sublimation and coagulation (see below) into snow [Frostgraupeln] and ice pellets [Reifgraupeln], and fall to the ground after reaching a diameter of around 3 mm. The infiltration of nuclei into the cloud most likely predominantly results from their wandering into the air electric field since they are possibly multiply charged, and also because of the atmospheric exchange.

The infiltration of existing ice crystals into supercooled water clouds has the same effect as the infiltration of sublimation nuclei. The ice crystals then act as very suitable sublimation nuclei.

An important process of this kind often occurs when the crown of a cumulus cloud penetrates through an overlying cap cloud. The cap cloud, which develops as a result of passive lifting or as an obstacle cloud on the cumulus (according to MÜGGE^{E5}), often already contains ice crystals, whereas the cumulus cloud only contains supercooled droplets. The ice crystals enter the water cloud as a result of turbulent air movements above the cumulus, and to a lesser extent due to their downward movement. There, they grow quickly due to sublimation and coagulation, and then fall into lower-lying layers. Usually, the number of ice crystals that enter the water cloud in this way are, of course, much smaller than the number of sublimation nuclei in the cloud, and their effect is therefore less significant. A complete transformation of the supercooled water cloud into an ice cloud is hardly possible in this way since the large amount of water contained by the water cloud in an unstable state as droplets or partly as vapour cannot be maintained in suspension by a few ice particles. This requires sublimation on sublimation nuclei. As long as the nuclei are uniform both inside and outside the cumulus, sublimation begins soon after the ice cap has been penetrated.

Ice crystals may also enter the secondary water clouds mentioned in Section 4. The crystals grow quickly in the way described, and precipitate out of the water cloud. Quantitatively, the process may be only of minor significance.

The ongoing transfer of ice crystals from an ice cloud to a lower-lying water cloud is quantitatively very significant. In this process, the ice crystals also grow considerably. The water droplets evaporate or fall out so that the water cloud eventually disappears entirely. In winter, this can occasionally be observed in the typically low-lying stratus cloud (which always consists of water droplets), when ice crystals from an overlying altostratus fall out into the water cloud. The process is important for flight meteorology because it leads to a considerable and rapid improvement of flight weather conditions.

6 Spatial arrangement of water and ice clouds

As has already been shown in the previous section, the spatial arrangement of water and ice clouds is by no means determined exclusively by the vertical atmospheric temperature distribution; it is not always the case that ice clouds are located at higher elevations than water clouds. In fact, the probability for the existence of ice clouds does rise with increasing height and decreasing temperature, but experience has shown that the existence of different aggregate states is much more complex than a simple temperature rule. The condensation and sublimation processes are decisively influenced by the nuclei and cloud elements in the air, which can differ greatly depending on the origin and the history of the air, and does not follow any regularity depending on height or temperature. Since the beginning of condensation and sublimation can at times be decisively influenced by small differences in the colloidal meteorological state of the air, and since such differences occur within short distances in the atmosphere (similarly to other meteorological elements), ice and water clouds can form side by side and on top of each other. The aggregate state of cloud elements changes from cloud to cloud. This has often been observed from aircrafts and can be identified without any instrumental means and can be identified by changes in ice states. This sometimes appears as a rapid change in the form of precipitation, for instance from snowflakes to sleet and vice versa. The coexistence of both aggregate states is significant for precipitation formation because the atmospheric exchange provides the

¹⁵According to a calculation not published yet.

conditions which can lead to sublimation in water clouds and result in precipitation.

7 On the difference between cloud and precipitation elements

Cloud elements fall out from most clouds because the fall velocity of cloud elements is always finite and the vertical movement on the lower boundary of the cloud is usually not uniform and cannot compensate for the downward movement of the cloud elements. However, precipitation falls from only relatively few clouds. Concerning rain formation in a cloud, it is decisive whether cloud elements are formed that are large enough to reach the Earth rather than evaporating on the way. Only such particles may be called precipitation elements.

Small droplets as well as small ice particles evaporate much more quickly than large ones when falling through dry air layers. As a derivation¹⁶ shows, the fall distance that a droplet can cover in unsaturated air grows with the fourth power of the droplet diameter. Whereas droplets with a diameter of $r = 10^{-3}$ cm already evaporate after falling a distance of 0.03 cm in air with 90 % relative humidity, droplets with a diameter of 10^{-2} cm can cover 3 m, droplets with a diameter of 5×10^{-2} cm cover about 2000 m, and larger rain droplets last for even longer distances. This is why only large rain droplets reach the ground when the air is not saturated (e.g. in summer rain), although, apart from the large droplets, many smaller droplets also certainly leave the cloud. Small droplets are only observed near the ground when the clouds are low-lying and humidity is very high. Otherwise, they cannot cover the distance from the cloud to the ground.

The r^4 -law explains why the bases of clouds that only contain very small cloud elements are always sharply delimited even if there are no dynamical causes for this. However, as soon as the cloud elements in stratus clouds are noticeably larger than $r = 10^{-2}$ cm, the cloud base becomes fuzzy and virga develop. The fuzzy cloud base is an external characteristic of ice clouds, whose elements are usually larger than 10^{-2} cm for the reasons discussed.

If the difference between cloud and precipitation elements should be determined based on size, then the limit between the two must be set at around $r = 10^{-2}$ cm. Incidentally, this has already been done for some time without knowledge of the r^4 -law.¹⁷ In individual cases, however, the lower size limit of the precipitation elements always depends on the elevation of the cloud and the humidity conditions below the cloud. As is well-known, the diameter of rain droplets is generally much larger than 10^{-2} cm.

¹⁶Unpublished as of yet.

8 Rain formation in pure water clouds

The droplets suspended in water clouds grow due to condensation and coagulation. Normally, neither of the two processes leads to large droplets.

The number of condensation nuclei is always so great that the amount of water vapour released in the air allows only for the formation of small droplets. For instance, even in summertime cumuli, an uplift of the air in a cloud from 1000 to 6000 m, along with a corresponding cooling of the air from $+15^{\circ}$ to -12° that results in a very high water content of 4.9 g/m³, with an extraordinarily small nuclei number of 100/cm³, only results in a mean droplet diameter of $r = 2.3 \times 10^{-3}$ cm. Droplets of such a small size cannot fall out as precipitation. The growth of droplets due to condensation plays an even more minor role in stratus clouds because of such minimal uplift. Even if the number of nuclei is extraordinarily low, the process is only of moderate significance. With a low nuclei count of only 2/cm³, which was observed once in oceanic air by Wigand¹⁸, the calculation would yield $r = 0.8 \times 10^{-2}$ cm, or a size that is relatively small compared to normal rain droplet sizes.

Coagulation is more important than condensation for the growth of the droplets. It has been experimentally confirmed¹⁹ without a doubt that coagulation, or the collision and coalescence of droplets, does occur. When two droplets collide, they join due to coalescence. Whether or not this can happen only for droplets with an entirely pure surface is yet to be determined. It is possible that droplet surfaces contaminated by absorbed soot and oil cannot coagulate, as is the case for aged fog in metropolises. Generally, however, droplet surfaces are sufficiently clean so that no impediments to coagulation exist.

The only essential cause for coagulation is the difference in fall velocity of single droplets, given by their difference in size. As has already been shown in Section 2, it cannot be doubted that droplets of different sizes coexist in clouds. In "homogeneous" clouds, where all droplets are of the same size, coagulation could only occur due to Brownian motion and turbulence. However, these processes are quantitatively entirely insignificant compared to the much stronger effect of coagulation due to the difference in fall velocity; these processes can therefore be neglected when considering natural, inhomogeneous clouds.

Due to differences in fall velocity, the droplets display movements relative to one another, which now and then result in collision and coalescence. The process can be described by a simple theory, and its validity has been confirmed for small droplets.²⁰ However, in case the proportion between the different droplets exceeds the value $r_1/r_2 = 10$, the theory does not seem to hold anymore. For hydrodynamic reasons, it seems

¹⁷R. SÜRING, Leitf. D. Meteorol., Leipzig 1927, p. 125.^{E1}

¹⁸A. WIGAND, Ann. D. Hydr. U. mar. Met. 58, 212, 1930.^{E1}

¹⁹W. FINDEISEN, Gerlands Beitr. z. Geophys. 35, 295, 1932.^{E1}

²⁰W. FINDEISEN, Gerlands Beitr. z. Geophys. l.c.

that these droplets of very different sizes coalesce much more rarely than would be expected. This is very important since, under such circumstances, the further growth of large droplets is made difficult or even prevented. In fact, experience shows that large droplets do not develop in water clouds (observed, for instance, in low stratus clouds), although layer thickness and the density of the clouds would suffice for coagulation as a result of fall velocity, provided there was no impediment for coagulation. Thus, the existence of such an impediment must be assumed. It is probably of a hydrodynamic rather than of an electric nature.

It seems that electric processes are not as relevant for rain formation as has been assumed in the past. Coagulation is, indeed, constrained by the same electric charge of the droplets, as this partly prevents collisions. However, even with a very high electric charge of the clouds, coagulation is not entirely prevented, and is probably only reduced to half of the extent that it would have with uncharged droplets. This can be drawn from the results of experimental studies.²¹

From this follows that water clouds always generate rain with small droplets. This is known as drizzle, which can only fall from low clouds and with high relative humidity. All water clouds at higher elevations cannot generate precipitation. Thus, water clouds are only of minor significance for rain formation.

Intensive drizzle must be more frequent in regions where ice cloud formation is inhibited by the lack of sublimation nuclei. This appears to be the case in western Scandinavia, for instance, where precipitation predominantly occurs with the supply of purely polar maritime air, indicating that the sublimation nuclei are of continental origin.

9 Precipitation formation in pure ice clouds

As has already been discussed, the small number of sublimation nuclei in sublimation leads to the formation of only few, large ice crystals. Soon after the formation of ice clouds, the ice crystals already reach the size that has been given above as the limit between cloud and precipitation elements. Provided that sublimation continues as a result of constant adiabatic cooling within the cloud (which is infrequent), the ice crystals grow into large sphaerocrystals (ice pellets) with a diameter of up to a few millimetres. Therefore, the sublimation process alone may generate precipitation elements in ice clouds, which can cover considerable fall distances through water-saturated air layers. The propensity for precipitation is generally larger in ice clouds than in water clouds.

The coagulation process is also important for precipitation formation in ice clouds. Ice crystals coagulate into snowflakes. The collision of particles is again caused almost entirely by the differences in fall velocity. For larger particles, hydrodynamic attraction due to the downward motion is probably also relevant. It is still unknown exactly in which cases the collision of particles results in coagulation.

Contact freezing may play an important role during coagulation of ice crystals. One may assume that high pressure is present on the small contact areas when two crystals collide, which may suffice to melt the ice when temperatures are not too low. As soon as the pressure decreases shortly after collision, the melting water freezes again and permanently combines the two crystals. If temperatures are above 0 °C, the crystals remain combined due to the adhesive effect of the melting water, a process that does not entirely result from the contact. Large snowflakes, composed of several single crystals (skeletons), may form as a result of continued coagulation. Experience shows that these occur primarily in thawing snow conditions. This suggests that coagulation conditions are less favourable at low temperatures. According to A. and K. WEGENER (l.c.), crystals only coagulate at temperatures above 0 °C and snow can only fall in the form of single crystals at below-zero temperatures. However, this does not match observations made by the author.

When the falling ice crystals or snowflakes melt, rain is generated. Rain formation is therefore considered to be a special case of snow formation. Regarding rain formation, it is of minor importance to know at what temperature ice crystals coagulate. Before ice particles melt into rain droplets, they always pass through a state in which their coagulation is easily possible and large snowflakes can form. Accordingly, melting then always leads to large rain droplets. Their size characterizes ice cloud precipitation in contrast to the drizzle of water clouds. Every large snowflake certainly results in several rain droplets, as they can only exist in the air up to a certain size and break apart as soon as they exceed the maximum size when falling.

The size of the snowflakes as well as the size of the rain droplets generated by coagulation depends on the coagulation conditions prevailing during the time when the ice crystals fall through the layer of the most favourable temperature. If there is a large number of falling ice crystals, coagulation is frequent and the single particles become particularly large. However, when there are few particles, only few collisions take place and large particles cannot form. Evidently, coagulation is also primarily influenced by the different fall velocities of the particles. The original size of the crystals is also a decisive factor for the eventually resulting size of the precipitation elements generated by coagulation. If these two factors are similar for a large and a small number of particles, then the rain from ice clouds should consist of large droplets in the case of high precipitation intensity and of small droplets in the case of low precipitation intensity. This happens to agree quite well with observations.

²¹W. FINDEISEN, Gerlands Beitr. z. Geophys. l.c.

Ice clouds occur much more frequently in the atmosphere than one used to think. They are often even significantly thicker than water clouds. Regarding precipitation-generating ice clouds, the most important level is the alto level. The typical ice cloud that is ready to precipitate is a stratus cloud at medium elevation, called the "typical alto stratus" according to the International Cloud Atlas. There is no difference between this cloud and the ice-containing nimbus, except for a difference in their vertical extent. Both forms occur on the front of any low pressure area and are decisive for the precipitation conditions there. From this it is clear what an important role ice clouds play in precipitation formation.

Ice clouds generally have a higher precipitation intensity than water clouds because of their greater affinity for rain and their greater thickness. However, extremely high precipitation intensities cannot be provoked by pure ice clouds. Only clouds in which both supercooled water droplets and ice crystals coexist are able to do so.

10 Precipitation formation in mixed-phase clouds

As discussed in Section 5, ice crystals that are suspended next to supercooled water droplets grow quickly at the cost of the droplets. Within a short period of time, all of the water that has been present in the form of water droplets will change to ice by attaching to the ice crystals; in addition, the ice crystals grow by the weight of the water vapour that was released into the air with the change of water saturation to ice saturation. The ice crystals become considerably larger than the droplets were because there are fewer ice crystals than there were droplets. As the tendency for a cloud to rain out increases with the size of its elements, the transformation of a supercooled water cloud into an ice cloud almost always leads to precipitation. Due to the transformation, many large precipitation elements form within a short period of time, whereas in pure ice clouds, the growth of elements is gradual, according to the rate of adiabatic cooling. Therefore, under the same conditions, precipitation from mixed-phase clouds is more intense than from pure ice clouds.

The growth of ice crystals in water clouds does not only occur with sublimation, as mentioned above, but also due to coagulation. The ice crystals often collide with droplets due to their usually relatively high speed and thereby enable coalescence, which takes place when the droplets freeze onto the crystals. It is not known whether coagulation can occur between every particle size. It may be rare or impossible for hydrodynamic reasons for certain size combinations of the particles to coagulate, as seems to be the case for the coagulation of droplets. This would mean that small graupel, for instance, do not grow due to coagulation, but only with sublimation. Indeed, the appearance of such graupel stemming from clouds of both aggregate states

seems to confirm this assumption. However, it is questionable whether the coagulation of crystals with very small droplets, most of which probably have a diameter of less than 5×10^{-4} cm, would leave traces on the surface of the graupel that are visible to the naked eye. Freezing happens very quickly at low temperatures and a distribution of the small amount of water does not take place; furthermore, the surfaces of the ice particles constantly change due to sublimation. The larger ice particles are certainly able to coagulate with the supercooled water droplets. The known structure of ice pellets and hailstones shows that the absorption of supercooled droplets takes place in the same way as the icing of aircraft parts. Furthermore, as shown by A. and K. WE-GENER (l.c.), hailstones still undergo sublimation even when they are below the 0 °C threshold.

Apparently, even larger and relatively rare supercooled water droplets occasionally coagulate with smaller ice crystals. The droplets are thereby stimulated to freeze and change into ice pellets, a development that can probably only be explained in this way.

Precipitation from clouds with coexisting supercooled droplets and ice particles differs depending on their process of formation and on their colloidal meteorological and thermo-dynamical composition. This can best be illustrated by means of examples.

11 Example: Precipitation formation in stratus clouds

Fig. 1 shows different kinds of stratus clouds that generate precipitation. The graph does not represent an aerological cross-section of the front side of a low pressure area, even if the conditions are similar with regard to some aspects.

On the right edge of the graph, two stratus clouds are shown, which may, for example, develop on subsidence levels. Both contain only water droplets, and the upper one is supercooled. The 0 °C line may be assumed at around 500 m elevation. Only the lower stratus may generate drizzle; the droplets falling from the more elevated stratus evaporate in the layer in between. Above the water clouds, a thick nimbostratus moves from left to right, which extends beyond the altostratus level and consists of only ice crystals. Ice crystals already fall from the higher-lying parts, but they are only small at first and evaporate when falling through the drier layers. With increasing thickness of the ice cloud as it expands downwards, the crystals become larger and soon reach the supercooled water cloud. There, the crystals quickly grow due to sublimation and coagulation, and form snow pellets that fall out with greater velocity (indicated by the longer strokes). Below the 0 °C line, the crystals melt and form large rain droplets that reach the ground along with the drizzle. Rain intensity increases considerably, particularly at the cost of the supercooled stratus, which, as a result, quickly dissolves. The lower stratus disappears more slowly because its droplets are less exposed

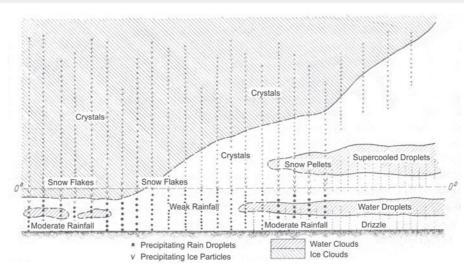


Figure 1: [Aerological cross-section of the front side of a low pressure area showing different kinds of stratus clouds that generate precipitation]^{E6}

to coagulation and evaporation. Rainfall becomes much weaker after the water cloud has dissolved; it intensifies again after some time as a result of the further thickening of the nimbostratus. The rain droplet size is small at first, but grows with increasing intensity of the rain since the falling ice crystals can coagulate to especially large snowflakes in the vicinity of the 0 °C line and become large rain droplets upon melting. The crystals that fall out develop primarily in the upper parts of the nimbostratus. Vertical movements and turbulence play significant roles in this process, perhaps also with contributions from the supercooled water clouds (not indicated in the figure), which are often present in the vicinity of the upper limit of ice clouds. Fractus clouds may form below the lower sections of the nimbostratus due to convection movements. These, however, are irrelevant to rain formation.

12 Example: Precipitation formation in cumuli

Fig. 2 schematically shows, from left to right, the different states of the colloidal meteorological development of a cumulonimbus.

In its first state, the cumulus consists of water droplets, both above and below the 0 °C line, which may be located at about 2000 m a.s.l. As it grows, the cloud eventually reaches a temperature at which sublimation on sublimation nuclei and ice crystal formation suddenly begins. Above this altitude where the critical temperature is reached, only ice crystals can form. The water droplets that are at first still carried above that height due to vertical flow will evaporate quickly to the benefit of the crystals. Ice pellets form as a result of sublimation, and snow pellets may also form due to simultaneous coagulation in the vicinity of the supercooled droplets. The ice particles do not enter the area below the critical temperature threshold as long as their fall velocity is smaller than the velocity of the vertical flow. The vertical updraft varies and, in some locations, is quickly exceeded by the fall velocity of some snow pellets that are exposed to particularly favourable growth conditions. These are then able to grow very quickly and significantly in the supercooled water cloud. The snow pellets turn into hailstones, which, thanks to their size, can fall out in spite of the strongest vertical velocities and even increase in size due to sublimation rather than melting below the 0 °C line. They become larger the longer they stay in the area of the supercooled water cloud. Therefore, they grow with longer distances between the 0 °C line and the height of the critical temperature, and they grow with greater vertical velocity of the air stream.

The thickness of the supercooled water cloud gradually decreases when snow and ice pellets enter in larger numbers and cause the supercooled droplets to disappear. A broad zone forms in which both solid and liquid phases coexist. This "mixed zone" provides the most material for precipitation. This irregularly shaped zone descends depending on the ascending vertical velocity, the fall velocity, and number of ice particles, so that the supercooled water cloud eventually changes into a pure ice cloud. After the thickness of the supercooled water cloud has decreased, the snow pellets falling through grow less vigorously, so that they melt and form rain droplets when they cross below the 0 °C line. Although any snow pellet may turn into several droplets under certain conditions, the droplets are still considerably larger than those stemming from condensation, without the detour of first passing through the solid phase. When the droplets fall they splash in the known manner and are thereby charged due to the Lenard effect; this probably already happens when the snow pellets melt. The origin of electrical phenomena in thunderstorms can be explained in this way, based on the current research. Thus, thunderstorm electricity is predominantly generated in the layer below the 0 °C line because large droplets are present only there. Large droplets occur only occasion-

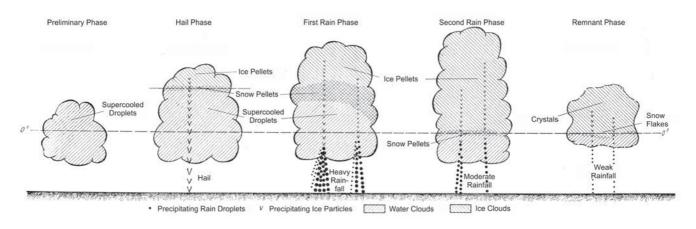


Figure 2: [Different phases (from left to right) of the colloidal meteorological development of a cumulonimbus]^{E6}

ally in the layer above the 0 °C line, taken there by strong updrafts. Strong electrical phenomena in thunderstorms can therefore only occur when the 0 °C line is sufficiently elevated, which is in well-known agreement with observations.

The start of rain depends on the vertical velocity of the flow at the lower limit of the cloud. As long as the velocity is larger than 8 m/sec everywhere, all droplets will remain suspended within the cloud and precipitation can only leave the cloud in the form of ice. The first rain droplets that reach the ground are always large. These often stem from hailstones or snow pellets that melted just before reaching the ground. The large droplets are favoured over the smaller ones because of their greater fall velocity, and especially by their greater stability when falling through dry air layers. Small droplets can only reach the ground when the layers between ground and cloud are already sufficiently moist.

Precipitation formation intensity in the cloud decreases considerably when the mixed zone expands down to the 0 °C line. After all, the mixed zone exists for only as long as water droplets from the layer below the 0 °C line are carried into the supercooled zone by vertical motion. At this point, snow pellet formation has almost stopped. Nevertheless, moderate precipitation continues due to intensive ice pellet formation in the upper part of the cloud. The ice pellets may still be large enough to allow the droplets to splash and continue to cause atmospheric electric phenomena.

When the updraft in the cloud ceases entirely and is replaced by subsidence, then the processes of sublimation and condensation are essentially finished. The ice pellets turn into solid crystals with the outer shrinking of the cloud. To some degree, they also fall out and coagulate into snowflakes in the vicinity of the 0 °C line, and melt with further falling to provide the light later rain.

As is already known, only few cumuli go through all of the five colloidal meteorological stages just discussed. One or several stages may be missing depending on the thermal and thermo-dynamical conditions as well as the kind and number of sublimation nuclei by which the development is decisively influenced.

13 Conclusions. Technical weather control

The discussions in Sections 1 through 12 show that any considerable precipitation formation is triggered by sublimation, and that sublimation nuclei play a decisive role in this process. This statement is more important to practical meteorology than the fact that condensation can only take place on condensation nuclei. While the formation of water clouds is hardly ever hindered because there are almost always enough suitable condensation nuclei present in the air, the character and the number of sublimation nuclei obviously varies, which is expressed in the development of the weather. It is due to differences in the content of sublimation nuclei that some clouds lead to precipitation while others do not under the same thermo-dynamical conditions; that some cyclones lead to heavy rainfall and others only to slight rainfall for seemingly incomprehensible reasons; that sometimes thick supercooled water clouds have severe icing effects and thereby interfere with aviation and on the other hand, with similar weather conditions, no icing at all is observed. The incredibly small mass of sublimation nuclei effectively influences large-scale weather dynamics and large atmospheric energy conversions. This is a wonderful confirmation of the view advanced by A. SCHMAUSS²² almost 20 years ago, which promoted considering the atmosphere as a colloid. A quantitatively insignificant matter, so far not considered in the weather forecast, turns out to be decisive for weather dynamics. A weather forecast that is only based on thermodynamical considerations will always remain uncertain. Sooner or later, it will have to draw on the results of colloidal meteorological research.

²²A. SCHMAUSS, Meteorol. Zeitschr. 1919, p. 16; 1920, p. 1; A. SCHMAUSS and A. WIGAND, Die Atmosphäre als Kolloid, Braunschweig 1929.

The finding that very small, quantitatively insignificant reasons suffice to trigger major weather dynamics confirms that human means may occasionally suffice to artificially influence weather dynamics. It would go beyond the scope of this study to discuss the potential of technical weather control in detail. New insights result from the thoughts presented in this paper. In accordance with these ideas, one may argue that it is possible to artificially trigger rain on occasion with relatively little effort, as well as remove the danger of icing and prevent hail. As a result of the thereby achieved energy transformations, other weather elements (e.g. temperature, wind) would also be affected, which could probably never be affected significantly by technical means in any direct way. The field of colloidal meteorology has been given a broad task simply to consider the potential of technical weather control; this task, of course, may only be solved in collaboration with aerology.

Endnotes

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E2 WILLIAM THOMSON is also known as Lord KELVIN, and the equation generally called Kelvin equation:

$$\ln\left(\frac{p}{p_0}\right) = \frac{2\gamma V_m}{rRT}$$

where p is the actual vapour pressure, p_0 is the saturated vapour pressure, γ is the surface tension, V_m is the molar volume of the liquid, R is the universal gas constant, r is the radius of the droplet, and T is temperature.

- E3 For a historical overview of condensation nucleus counters, including those by AITKEN and SCHOLZ, see: McMURRY, P.H., 2000: The History of Condensation Nucleus Counters. Aerosol Science and Technology, **33**, 297–322.
- E4 TRABERT, W., 1901: Die Extinktion des Lichtes in einem trüben Medium (Sehweite in Wolken). – Meteorol. Z. **18**, 518–524.
- E5 RATJE MÜGGE, German meteorologist. The exact reference could not be located.
- E6 Original figure has no caption.