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The formation of atmospheric ice crystals by the freezing of droplets

By E. K. BIGG
Imperial College, London

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SUMMARY

Using the results of laboratory experiments on the supercooling of purified water, the freezing of cloud and raindrops is examined. It is shown that at temperatures lower than about − 30°C in cold-box or expansion-chamber experiments, the drops freeze in approximately the numbers that would be expected if they were pure water. Those that freeze at temperatures higher than about − 20°C seem to be more numerous than would be the case if the drops were pure. An interpretation of the main features of the Findeisen and Schulz expansion-chamber experiments is found to be possible without appealing to the action of foreign ice-forming nuclei, although there is a discrepancy between calculation and experiment at temperatures higher than about − 20°C. In the atmosphere, formation of cirrus clouds is shown to become possible at temperatures below about − 35°C, glaciation in stratiform clouds to become appreciable at temperatures below about − 20°C and freezing of raindrops in strongly convective clouds to become important below about − 13°C, without the presence of ice nuclei. It is concluded that freezing nuclei may be important at temperatures above about − 20°C, while their presence at lower temperatures will be masked by the freezing of uncontaminated drops.

1. INTRODUCTION

It is well known that condensation of water vapour in the atmosphere takes place on small suspended particles called 'condensation nuclei.' It has also been established that clouds composed almost entirely of water drops can exist at temperatures approaching − 40°C. On the other hand, the ice phase is often observed in clouds at considerably higher temperatures. It is generally assumed that the freezing in these cases is brought about by impurities, which in this context are called 'freezing nuclei.'

In a recent paper (Bigg 1953) the author has described some laboratory experiments on the freezing of water drops which seem, in principle, applicable to such problems of cloud physics. In order that readers may judge the validity of using these results for calculating the probability of freezing of airborne drops, the method and conclusions of that work will be briefly summarized.

A technique used by Dufour (1861) was adopted for studying the freezing of drops in the diameter range 0·1 mm to 2 cm. The drops were suspended at the interface of two insoluble liquids, one heavier and one lighter than water, and were therefore free from the constraints imposed by a solid surface. It was found that temperatures of freezing consistent within the limits of experimental error were obtained with all the combinations of liquids tried. It was also shown that results consistent with these were obtained with drops resting on a hydrophobic film of silicone oil ('drifilm') baked on to a glass surface, and covered with an insoluble liquid; it was concluded that under these conditions the freezing of the drops was uninfluenced by their surroundings.

The numerical results obtained agreed in form with those of Dorsch and Hacker (1950) but differed quantitatively. It was found that the mean freezing temperature of drops depended upon their volume and also the rate at which they were cooled. In Fig. 1 are plotted the mean freezing points of groups of drops having different diameters, as a function of their diameters. Some more recently determined points for very small drops are included. (These small drops, produced by condensation on a drifilm surface and covered with liquid paraffin, were viewed by transmitted light with a microscope and were photographed at short intervals. The points represent a total of 314 drops

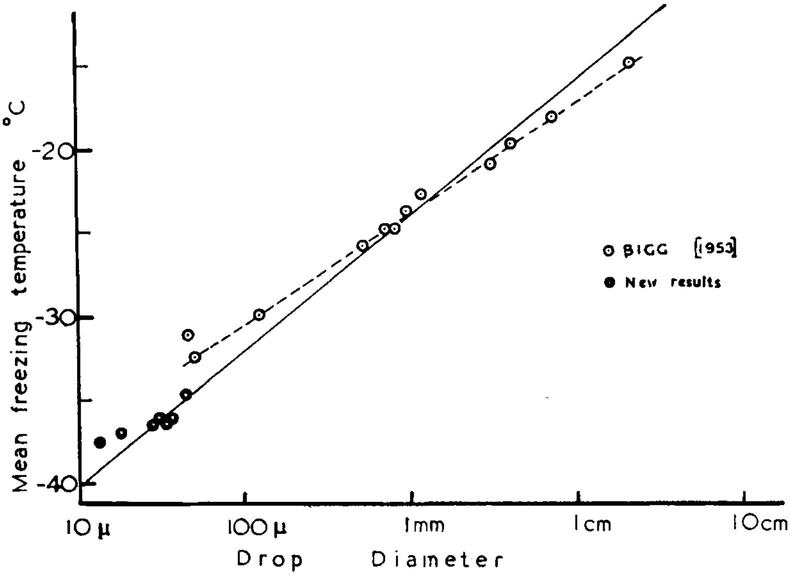


Figure 1. The mean freezing temperature of drops as a function of their diameter, when suspended at the interface of two liquids.

studied.) The full line in Fig. 1 represents a slight amendment to the values published earlier, made necessary by the new experiments; the pecked line is the one previously published. The new experiments confirm the conclusions that the probability of freezing depends upon the volume of the drop and rate of cooling. It was shown that for a drop of volume $V\,\mathrm{cm}^3$ cooled to a temperature T_s below 0°C for t seconds, the probability P that it will freeze is given by

$$\ln(1 - P) = -\,V t\, K(\exp aT_s - 1) \qquad (1)$$

where the factor $K(\exp aT_s - 1)$ has been empirically determined and a and K are constants. From the pecked line of Fig. 1, a was found to be 1·0 and K to be $5{\cdot}2 \times 10^{-10}$. The full line of Fig. 1 requires instead that a should be 0·82 and K to be $2{\cdot}9 \times 10^{-8}$. Although this may seem an excessive adjustment, the effect on mean freezing temperature is seen from the figure to be slight. As the amended results are based on more extended observations, they will be used throughout this paper.

In the case of drops being cooled, or growing, it was shown that Eq. (1) should be modified to read

$$\ln(1 - P) = -\int_0^t V K(\exp aT_s - 1)\,dt \qquad (2)$$

Another conclusion was that distilled water from various sources gave the same mean freezing temperatures when cooled under the same conditions. It was supposed that the impurities present in distilled water were without effect upon freezing.

The statistical nature of freezing and the factors of volume and time have not been fully considered before in discussing the appearance of the ice phase in meteorological experiments. It is therefore proposed to apply the Eqs. (1) and (2) to the freezing of cloud and rain drops in order to assess the role played by freezing nuclei. If the point be granted that the freezing of the drops was not influenced by their environment in these experiments, the equations given above should be equally applicable to airborne drops.

2. An examination of experiments with chilled clouds

When a cloud is suddenly chilled to a known temperature by the passage of a cold body as in the experiments of Schaefer (1946) and Mason (1952), or by rapid expansion followed by a period of constant temperature as in the experiments of Cwilong (1947) and Fournier d'Albe (1949), we can obtain an estimate of the most probable number of ice crystals with the help of Eq. (1). The diameters of drops in these experiments were in the range of 5 μ to 20 μ and the cooling times varied from 0·1 to 1 sec. Table 1 shows the calculated fraction of droplets which will freeze as a function of the temperature and time of cooling.

TABLE 1. The mean number of ice crystals produced per 1,000 drops

Duration of cooling	0·1 sec			1 sec		
Diameter (μ)	5	10	20	5	10	20
Temperature (°C)						
− 38	0·0	0·0	0·0	0·0	0·0	0·4
− 39	0·0	0·0	0·1	0·0	0·1	1
− 40	0·0	0·0	0·2	0·0	0·3	2
− 41	0·0	0·1	0·6	0·1	0·7	6
− 42	0·0	0·2	1·2	0·2	1·6	12

This table shows how it is that, with a consistent method of observation, an apparently critical temperature for the formation of ice crystals can be found, for the drop-size spectrum is narrow in an artificial fog. It shows also that different experimental techniques producing droplets of different sizes and different cooling times must yield rather different critical temperatures, thus explaining why different observers have obtained values ranging from − 39°C to − 42°C. In Cwilong's and Fournier d'Albe's experiments the drops were small and their experimental volume and duration of cooling were small, which explains why their critical temperatures were the lowest reported.

In general, then, agreement between experiment and calculation is good and consequently we are justified in proceeding to estimate the freezing temperature of drops under other conditions.

3. An examination of the expansion-chamber experiments of Findeisen and Schulz

Various experimenters have attempted to determine the properties of freezing nuclei by allowing the air to cool by expansion and counting the number of ice crystals at various stages of the experiment. Findeisen and Schulz (1944) and Schulz (1947) came nearest to achieving atmospheric conditions by expanding rather large volumes of air slowly. Their results are well known (see e.g., Mason and Ludlam 1951) and will only be summarized here.

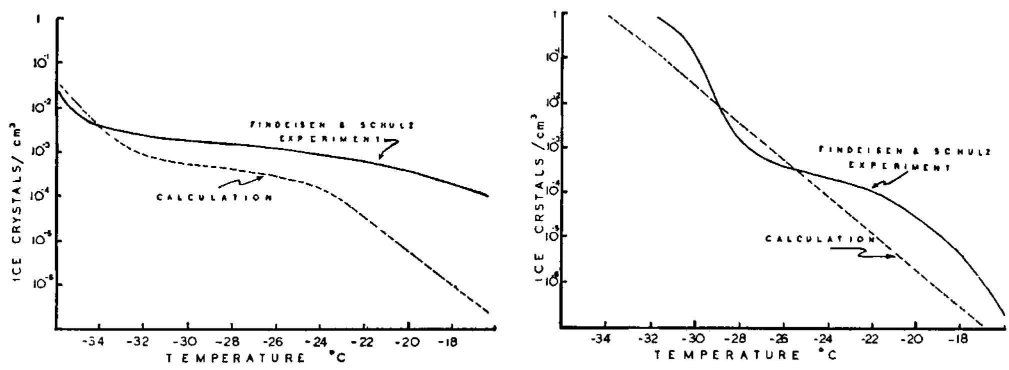
The number of ice crystals formed above − 20°C was very small and decreased with increasing rates of expansion up to a rate equivalent to an ascent speed of 35 m/sec, then increased again. The nuclei presumed responsible for the formation of these crystals were designated ' Type I.' A marked increase in the number of crystals with decreasing temperatures set in at about − 30°C and was attributed to the activation of ' Type II ' nuclei. The critical temperature at which this increase was first noted fell with increasing rates of expansion. The critical temperature of Type I nuclei depended on the expansion rate at the moment of condensation.

The expansion vessel used by Findeisen and Schulz had a depth of 2·5 m and that of Schulz 1·5 m. A little consideration shows that practically all of the original drops must have fallen out during the slowest expansions which last 20 min or so and that therefore a new population of drops must have arisen as the cooling of the air was continued. This must be taken into account in considering freezing, for, according to Eq. (1) both the volume of the drop and the time for which it has been cooled influence its probability of freezing.

The rate of precipitation of drops depends on the degree of mixing of the air in the chamber. If turbulent velocities of the order of a few cm/sec existed, then we can consider the droplets to have a uniform spatial distribution. The walls of the inner chamber were very thin but had a thermal capacity in Schulz's apparatus equivalent to about 60 l of air, and even more in the Findeisen and Schulz equipment. In Schulz's experiments, turbulence was also assured by the presence within the chamber of two 35 w lamps. It seems fairly certain that the temperature inequalities of the larger vessel would also have given rise to a certain amount of turbulence.

The method by which the number of drops present at any moment during the expansion can be calculated, assuming that the air in the chamber is kept mixed, is outlined in the appendix. It requires a knowledge of both the initial dewpoint of the air and the number of condensation nuclei. Neither of these quantities was recorded by the experimenters. At low expansion rates the freezing spectrum is found to depend quite critically on these initial assumptions. We find however that all reasonable values yield curves of a similar form, so that in order to illustrate the method of calculation, we shall arbitrarily choose a dewpoint of 7·5°C and 1,000 activated nuclei/cm^3. One other important assumption has to be made. As the expansion progresses and the original drops fall out, the supersaturation will commence to rise. At what point will a new set of nuclei be activated ? An arbitrary decision must be made here, for in practice the supersaturation will be prevented from rising at the maximum rate by the precipitated ice and water in the chamber. In order to calculate Fig. 2 we have assumed that when the number of drops falls to 1/cm^3, new ones commence to be generated at such a rate that the liquid-water content remains constant.

The calculated curve for 5 m/sec in Fig. 2 (a) suggests that the rather sudden rise in the number of ice particles observed at temperatures below − 30°C may be due merely to the physical limitations of the apparatus rather than the appearance of a new type of ice nucleus. Fig. 2 (b) shows the 20 m/sec curve, again compared with the Findeisen and Schulz results. In this case calculation and experiment are in closer agreement.



(a) Expansion rate equivalent to an ascent of 5 m/sec. (b) expansion rate equivalent to an ascent of 20 m/sec.

Figure 2. Comparison of the number of ice crystals found by Findeisen and Schulz, with the number calculated from the freezing of the droplets.

The initial trend of the experimental observations with increasing expansion rates can be explained. The bend in the curve moves initially upwards and to the left, causing a fall in the temperature at which, say, 100 crystals/litre is observed. This was mentioned by Findeisen and Schulz in passing, but not recorded on their diagrams. At higher expansion rates the bend disappears altogether, so that 100 crystals/litre would be observed at a considerably higher temperature.

The lower end of the curve moves to the left with increasing expansion rates, because the faster rate of cooling reduces the probability of freezing. The reversal of this trend at higher expansion rates found by Schulz cannot be matched by calculation without introducing dubious assumptions.

We conclude from this examination that the main features of the experiments can be explained without invoking ice nuclei at all. The reasonable agreement with the 20 m/sec curves makes it seem likely that the difference between calculation and experiment is largely a function of the experimental design and measurement.

We must not infer from this that freezing nuclei do not exist in the atmosphere but only that they are of minor importance in these rather artificial laboratory conditions.

There have been many cold-box or expansion-chamber experiments in recent years, but in each case there is a lack of knowledge of the conditions which prevents an accurate comparison by calculation. As examples we could mention the experiments of Palmer (1949), expansion chamber; Reynolds (1950), cold box; aufm Kampe and Weickmann (1951), cold room. The number of ice crystals reported in each of these experiments was greater at temperatures higher than $-20°C$ than can reasonably be explained by our method of calculation.

The chief lesson to be learnt from this analysis is that such experiments are subject to grave misinterpretation unless the importance of the history of the drops and the physical dimensions of the apparatus is realized.

4. Ice formation in clouds

Freezing of droplets in clouds can be predicted in the same way as for the laboratory experiments just described. Reliable observational checks are few because of the difficulty of distinguishing from an aircraft between supercooled drops and crystals. Temperature measurements in clouds are also somewhat unreliable. Moreover, the development of a fibrous appearance has often been taken to imply the existence of ice crystals, but, as Ludlam (1952) has pointed out, it may in certain circumstances be due to large water drops.

Again, we must bear in mind the possibility that there is secondary generation of ice particles in the atmosphere. It has often been suggested that splinters could be detached from growing ice crystals and if this should happen the number of ice particles actually occurring may far exceed the number calculated on the basis of the freezing of droplets.

Cirrus

Cirrus clouds consist entirely of ice particles except perhaps during their formation and early growth. If in a growing cloud 1 per cent of the droplets freeze within about a minute of their formation, then the crystals may be expected to grow rapidly and cause the evaporation of the droplets, so that a fibrous-cirrus ice cloud with no ' mother cloud ' develops. The temperature at which this is likely to occur can be estimated from Table 2.

TABLE 2. THE MEAN TIME (SEC) TAKEN FOR 1% OF DROPS TO FREEZE

Diameter (μ)	0·5	5	50
Temperature (°C)			
− 30	$1{\cdot}6 \times 10^{8}$	$1{\cdot}6 \times 10^{5}$	160
− 35	$2{\cdot}8 \times 10^{6}$	$2{\cdot}8 \times 10^{3}$	2·8
− 40	$4{\cdot}8 \times 10^{4}$	48	0·05
− 45	900	0·9	9×10^{-4}

Since at high levels condensation nuclei are scarce, the droplets rapidly acquire diameters exceeding 5 μ, and we therefore see that this kind of cirrus cloud is unlikely to develop unless the temperature is below − 30°C, but could always appear at heights where the temperature approaches − 40°C. This is in broad agreement with the results of Weickmann (1949), who found that a frequency distribution of cirrus tops as a function of temperature showed a maximum in the range − 46°C to − 50°C, with occasional cases occurring at temperatures as high as − 30°C.

Stratus

In a moderately thick layer of stratus cloud drops may have very long lives at approximately constant temperatures. If we assume the drops to be of equal size, then we can write the volume of each drop as $V = w/N$ where w is the liquid-water content and N the number of drops per unit volume. To find the temperature at which there will, on the average, be one ice crystal per unit volume, if the probability of freezing is very much less than unity (so that $\ln(1 - P) \simeq P$), we can write from Eq. (1)

$$1 = NP = 2{\cdot}9 \times 10^{-8}\,(e^{0{\cdot}82\,T_s} - 1)\,wt$$

and obtain Table 3. This table shows that we should not expect ice crystals in concentrations greater than 1/litre unless the temperature falls below about − 20°C. Mason and Howorth (1952) in an analysis of precipitation from stratiform clouds over Northern Ireland state that, 'there was a marked increase in the frequency of rain/snow when the cloud-top temperature fell below about − 12°C, suggesting that only below this temperature does the concentration of ice-forming nuclei, on the average, become significant for precipitation release.' No clouds which had summit temperatures higher than − 13°C gave snow. In fact, in only 3 out of the 10 well-substantiated cases of snow was the summit temperature higher than − 29°C.

TABLE 3. THE TEMPERATURES AT WHICH AN AVERAGE OF ONE ICE CRYSTAL WILL FORM BY DROP FREEZING IN VARIOUS VOLUMES AND TIMES IN CLOUDS OF DIFFERENT LIQUID-WATER CONTENTS

Time	Liquid-water content		Volume	
	g m^{-3}	cm^{3} × 10^{6}	cm^{3} × 10^{3}	cm^{3}
1 sec	0·1	− 24°C	− 32°C	− 41°C
1 sec	1·0	− 21°C	− 29°C	− 38°C
1 min	0·1	− 19°C	− 27°C	− 36°C
1 min	1·0	− 16°C	− 24°C	− 32°C
1 hr	0·1	− 14°C	− 22°C	− 31°C
1 hr	1·0	− 11°C	− 19°C	− 28°C

Isolated cases have been reported of ground fogs and low stratus which contained ice crystals when their minimum temperatures were above $-10°C$. Findeisen and Schulz report one such occasion on which ice crystals were seen at $-6°C$. Dessens (1950) reports that all clouds below $-12°C$ observed at the summit of Puy de Dôme contained ice crystals. This was not, apparently, the temperature of the cloud top and he also remarked that measurements at mountain summits must often be treated with reserve because of the snow-covered ground in the neighbourhood.

Cumulus

Conditions in cumulus are not as steady as in stratus, but the number of cloud droplets freezing in a cumulus will be comparable with those given in Table 3. Probably an important source of ice particles in cumulus clouds (and possibly also in stratus clouds) is the freezing of rain or drizzle drops, which are larger than the representative cloud elements. It is now well established that these larger drops can be sufficiently numerous to provide rain without the intervention of the ice phase; Bowen (1950) and Ludlam (1951) have considered their growth by coalescence in clouds having various updraughts and water contents. As far as freezing is concerned, the most important period is that during which the drop is suspended in the updraught near the cloud summit. Assuming that it spends 100 sec near the summit, we derive Table 4.

TABLE 4. THE TEMPERATURES AT WHICH DROPS OF DIFFERENT DIAMETERS MUST BE KEPT FOR 100 SEC TO HAVE PROBABILITIES OF FREEZING OF 0·1% AND 10%

Diameter of drops mm	Probability of freezing 0·1%	10%
0·1	− 28°C	− 32°C
0·3	− 23°C	− 29°C
1·0	− 19°C	− 25°C
3·0	− 15°C	− 21°C
5·0	− 13°C	− 19°C

The largest drops found in clouds would therefore commence to freeze in significant quantities at about $-13°C$. The copious spicules thrown out by large drops when they freeze could become an important secondary source of ice particles.

Ludlam (1952) summarizes soundings made over the North Atlantic by the British Meteorological Flights during the period Jan. 1944 to Mar. 1946 and also observations on summer cumulus over New Mexico by Braham, Reynolds and Harrell (1951), in a diagram in which the percentage of convective clouds reported as cumulonimbus is plotted as a function of their summit temperature. This reveals a considerable difference between winter and summer clouds, the percentage of cumulonimbus being much smaller, for a given temperature, in winter than in summer. This could be at least partly due to the smaller number of drops freezing in the winter clouds of lower water content. The observations seem to be generally compatible with Table 4 with the exception of the small percentage of cumulonimbus occurring with summit temperatures above $-10°C$.

It must be emphasized again, however, that a convective cloud may contain ice without appearing to be a cumulonimbus, while a cumulonimbus does not necessarily contain the ice phase.

More definite evidence of freezing in relatively warm clouds has been given by Coons, Jones and Gunn (1949) in describing traverses through cumulus of the Gulf Coast of the United States. They state that 'usually when the temperature at the tops of such clouds was below $-6°C$, natural ice crystals were detected during in-cloud traverses.'

Bowen (1952), too, has reported that hail fell from clouds with summits at $-6°C$ and $-7°C$ when sprayed with water. Even allowing copious secondary generation of ice crystals from a frozen drop, these temperatures are far too high to be explained by the freezing of purified water.

5. Discussion

Now that we have discussed quantitatively the effects to be expected from the freezing of distilled water, we must ask the question : are freezing nuclei really important in atmospheric phenomena ?

In the cold-box experiments at about $-40°C$ there seemed to be reasonable agreement of calculation with experiment. We therefore do not need to postulate the presence of nuclei, for homogeneous nucleation for example, as proposed by Mason, would serve equally well to explain freezing.

The experimental observations of Findeisen and Schulz made with expansion rates of 5 m/sec showed a growing departure of calculation from experiment at higher temperatures, amounting to as much as 5°C for corresponding crystal counts. Is this discrepancy genuine ? It cannot be completely explained by making different initial assumptions. Yet, if it is real, it should persist at other expansion rates. Now, we have seen that at 20 m/sec, a rate of ascent for which the physical dimensions of the vessel and the initial conditions are much less important than at 5 m/sec, the difference is much smaller. This casts doubt upon whether freezing nuclei are really responsible for the difference between observation and calculation at slow rates of expansion.

Observations of naturally-occurring clouds have, however, provided us with some examples where ice crystals appear to have been present in far greater quantities than would be expected if they were formed by the freezing of purified water. These cases are those where there is possibility of industrial smoke contaminating the drops (fogs and low stratus), or where strong convection can draw up large supplies of foreign particles from the ground. There is laboratory evidence, of course, that soil particles and combustion products can produce ice crystals in a supercooled cloud.

It seems fair to say that in many well-supercooled clouds the rate of transformation from water to ice may take place as though the water were pure, but can on occasions be speeded up by contamination with impurities. We might perhaps have expected this result, for we know that there are some special substances, such as silver iodide, which can cause cloud droplets to freeze at temperatures as high as $-5°C$.

6. Conclusion

This examination indicates that freezing nuclei may exercise a far less widespread influence than has previously been thought. It shows that expansion-chamber experiments need very careful interpretation and that in them, measurements should be made of dewpoint, droplet size and concentration, as well as of the number of ice crystals.

It shows that ice nuclei are important on occasions in natural clouds. It shows too, the need for statistical data on the frequency of occurrence of ice crystals in such clouds and emphasizes the need for observing the history of the cloud as well as its water content, if deductions are to be made about the efficiency of ice nuclei in promoting freezing.

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Appendix

Outline of calculation for Findeisen and Schulz experiment

First we must calculate the number of drops present and their volume at any time. Then the probability of their freezing can be found from Eq. (2) in Section 2.

For the first step we assume the drops to be uniformly distributed by turbulent mixing, so that the rate of decrease in the population is given by

$$\frac{dN}{dt} = -\frac{Nv}{z} \quad . \quad . \quad . \quad . \quad (3)$$

where N is the number of drops/cm^3, v their fall-velocity relative to the air, and z is the total depth of the vessel. The drops are assumed to be all of equal size.

Then $v \simeq 1{\cdot}3 \times 10^6 r^2$ where r = radius (cm). Their volume $V = \frac{4}{3}\pi r^3 = (w - w_p)/N$ where w is the total liquid-water content/cm^3 if none had been precipitated and w_p the amount precipitated from each cm^3.

Thus, $$v = 1{\cdot}3 \times 10^6 \{3\,(w - w_p)/4\pi N\}^{2/3}$$

Substituting in Eq. (3), $$-\,dN/N^{1/3} = \frac{1{\cdot}3 \times 10^6}{z} (\tfrac{3}{4}\pi)^{2/3} (w - w_p)^{2/3}\, dt$$

Integrating, $$N_0{}^{2/3} - N^{2/3} = \frac{2{\cdot}6}{3} \times \frac{10^6}{z} (\tfrac{3}{4}\pi)^{2/3} \int_0^t (w - w_p)^{2/3}\, dt$$

The integral can be evaluated in small steps, taking w_p at its value for the preceding step and the new w_p found.

Assuming $N = 1{,}000$, $T = 7{\cdot}5°C$ when $t = 0$, $z = 2{\cdot}5$ m, the time at which N has fallen to $1/cm^3$ can be found. It is then assumed that new drops are generated in sufficient numbers to maintain a constant liquid-water content.

Having found the number of drops, their volume and their temperature as a function of time, the probability of their freezing can be calculated from the integral of Eq. (2).