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R. R. VIERHOUT
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IN CLOUDS AND FOG

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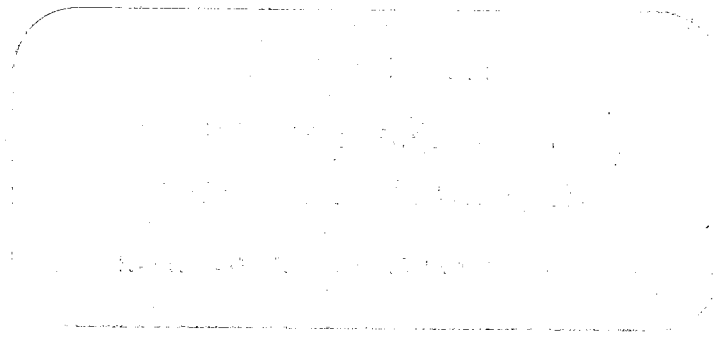
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DEPARTMENT OF CHEMISTRY

LABORATORY

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ON THE FORMATION OF DROPS IN CLOUDS AND FOG

by R. R. Vierhout

Summary

Applying the formula for the diffusion of vapour in a gas and for the rate of evaporation of water above a surface of water at a vapour-pressure differing from the one of saturated vapour and a formula, deduced from theory, for a flow of gas, the evaporation-velocity is determined for a drop of water in a cloud of drops. For an actual case the values of this velocity and also the times required for the complete vanishing of the drops are collected in a table (table 2). It appears from this table that among the drops those with a radius of 10^{-3} cm occupy a special position. Further, the rate of accretion of drops is computed in the case of a slight oversaturation; the results, also given in table 2, make it clear that drops in a drizzle can hardly originate by this process even if the over-saturation is measurable. Further the conclusion is drawn that ice-needles in a surrounding of undercooled waterdrops have a fair chance of increasing their weight to that of raindrops. It appears once again that in normal cases of very slight over-saturation the drops will increase till their radius is of the order of magnitude of 10^{-3} cm, from which it is concluded that drops of this size will be present to a predominating extent in cu-clouds. Larger drops are shown to require a very much longer time for developing. The coalescence effect is then computed and it is made clear that drops of the order of 10^{-3} cm radius and larger have a reasonable probability of vanishing from the fog, from which it is concluded that a radiational fog, at any rate in its bottom layers, cannot contain drops of this size and that the relative humidity must be less than 100%. It is further shown that in its top-layers, on the contrary, drops of this critical size can always originate and that they, after falling down, evaporate, in this way setting up a downward cold transport, until the relative humidity has attained 100%, where upon the fog will become „wet”. Finally it is proved that very strong attractions and repulsions between the drops result from evaporation or condensation from or at the drops.

§ 1. The rate of evaporation or condensation at the boundary surface in the liquid-vapour system of water can be expressed in grammes per second per cm^2 of that surface or in moles per second per cm^2 . This rate is proportional to $(E-e)$, E denoting the pressure of saturated vapour and e the actually prevailing vapour pressure. $E-e$ is positive for evaporation and negative for condensation. In order to determine the proportionality-factor by which $(E-e)$ must be multiplied, we introduce a rectangular system of co-ordinates with the x -axis normal to the boundary surface. For the number da of molecules in 1 cm^3 of the vapour, with velocities between \dot{x} and $\dot{x} + d\dot{x}$, we have:

$$da = n \sqrt{\frac{M}{2\pi RT}} \cdot \varepsilon^{-\frac{M\dot{x}^2}{2RT}} \cdot d\dot{x}, \quad [4]^1$$

where \dot{x} denotes the velocity of a molecule in the direction of the x -axis, n the number of molecules per cm^3 , M the molecular weight, R the gas-constant, T the absolute temperature and ε the base of the natural logarithms. In order to compute the number of vapour-molecules impinging on 1 cm^2 of the boundary-surface in 1 second we shall first consider the molecules having velocities between \dot{x} and $\dot{x} + d\dot{x}$. In 1 sec. they cover the distance \dot{x} , so that their number is

$$da \cdot \dot{x} = n \sqrt{\frac{M}{2\pi RT}} \cdot \varepsilon^{-\frac{M\dot{x}^2}{2RT}} \cdot \dot{x} \cdot d\dot{x}.$$

The total number, striking the boundary-surface per cm^2 per second, is therefore obtained by integrating the above expression over all possible velocities \dot{x} in one direction, say all positive values \dot{x} . This gives for the total number

¹⁾ References are indicated by [].

$$A = n \sqrt{\frac{M}{2\pi RT}} \int_0^{\infty} e^{-\frac{M\dot{x}^2}{2RT}} \cdot \dot{x} \cdot d\dot{x} = n \sqrt{\frac{RT}{2\pi M}}$$

According to M. Volmer and J. Estermann [5] these molecules are, all of them, absorbed by the liquid. Part of them are reflected by the solid phase (ice). Writing $\frac{nRT}{N}$ for e (N =Loschmidt's number), we obtain

$$A = \frac{eN}{RT} \sqrt{\frac{RT}{2\pi M}} = eN \sqrt{\frac{1}{2\pi MRT}} \text{ molecules per cm}^2 \text{ per second.} \quad (1)$$

In the case of thermodynamic equilibrium e is equal to E , so that the number of molecules per cm^2 per sec. moving from the vapour into the water is then

$$A_v = EN \sqrt{\frac{1}{2\pi MRT}} \quad (2)$$

As equilibrium prevails the same number of molecules must per cm^2 per second pass from the water into the vapour; (2) represents therefore also the number of molecules per cm^2 per second, which at the temperature T and at the pressure E of saturated vapour is shot by the water through the boundary-layer. If there is no equilibrium, we can for simplicity still suppose the temperature of the water and of the vapour to be the same and the excess of molecules passing in one direction will then be

$$A_v - A = N \sqrt{\frac{1}{2\pi MRT}} (E - e) \text{ molecules per cm}^2 \text{ per second,}$$

which therefore represents the velocity of evaporation or of condensation according as $E - e$ is positive or negative.

Expressed in grammes this becomes:

$$\frac{M}{N} (A_v - A) = \frac{M}{N} \cdot N \sqrt{\frac{1}{2\pi MRT}} (E - e) = \sqrt{\frac{M}{2\pi RT}} (E - e) \text{ gr/cm}^2 \cdot \text{sec.}$$

For a boundary layer of which the radius of curvature is r , we have Thomson's formula:

$$\frac{\rho RT}{M} \ln \frac{E_r}{E} = E_r - E + \frac{2\gamma}{r}$$

Here ρ denotes the density of water, γ the surface-energy per $\text{cm}^2 \approx 75 \text{ erg/cm}^2$, E_r and E the pressure of saturated vapour belonging to the curved and to the flat water-surface respectively. By expanding into a series, breaking off after the first term and neglecting: $\frac{M}{\rho RT} \cdot E$ against 1, this can be written:

$$E_r = E \left(1 + \frac{2M}{\rho RT} \cdot \frac{\gamma}{r} \right) = E \left(1 + 3,25 \cdot 10^{-5} \cdot \frac{1}{rT} \right) \quad (3)$$

This expression is accurate to within 0.1% for drop-radii down to 10^{-6} cm .

A drop of radius r produces or absorbs an amount of water vapour C , for which we have

$$C = 4\pi r^2 \sqrt{\frac{M}{2\pi RT}} (E_r - e_g) \text{ gr/sec.} \quad (4)$$

Here e_g denotes the vapour-pressure at a distance λ' from the surface of the drop, λ' representing the average distance from which the vapour-molecules arrive. We have therefore left room for the possibility that the value of the vapour-pressure at a certain point depends on the distance of this point from the surface of the drop. In deriving (4) from (1) and (2), one must take into account that the average distance λ' , from which the molecules, passing across the boundary-layer, arrive, is of the order of magnitude of the free path λ of a vapour-molecule. In the conditions prevailing in the case of fog or clouds, λ is of the order of 10^{-5} cm .

Drops in fog or clouds contain, apart from water, small quantities of other substances, which by their presence cause a lowering of the pressure of saturated vapour. Now it may happen that the rise in the saturation-pressure due to the curvature of the surface of the drop is just compensated by the lowering, due to the presence of alien substances. In that case such a drop is in thermodynamic equilibrium with an atmosphere of a relative humidity of 100 %. The radius of such drops produced by the actual processes in nature is of the order of 10^{-5} cm. If by condensation this radius becomes 10 times larger, the lowering of the saturated vapour-pressure will become 1000 times smaller and the raising 10 times smaller, so that the resulting rise will be 99 % of the one computed by means of Thomson's formula. The effect of the dissolved substances present in the drops can therefore be neglected for radii $> 10^{-4}$ cm. For smaller drops the pressure of saturated vapour will be slightly less than would follow from (3).

Let us suppose a drop to be at rest in an infinitely extended quiet atmosphere and let C gram/sec or C/M moles/sec be its strength as a source.

This strength will depend on its own pressure of saturated vapour and on that of its surroundings. This will cause a change of the radius; moreover latent heat will have to be supplied or to be withdrawn for evaporation or condensation. We suppose now, provisionally, that the radius of the drop is kept constant by some suitable device, for example, by supplying or withdrawing water through a diminutive tube, introduced into the drop, and that by a similar contrivance the regulation of the latent heat is properly seen to. Under these conditions a steady flow of vapour will be set up. The pressure of the vapour in this flow at a distance λ' from the surface of the drop will be called $e_{\lambda'}$, at infinity e_{∞} and at a distance a from the centre of the drop e . Owing to the existing radial symmetry the pressure-gradient will be radially directed and through any imaginary spherical surface round the drop the same amount of C gram/sec or C/M moles/sec of water-vapour will be transferred.

Denoting the concentration of the vapour in moles/cm³ by m and that of the air by m' , their sum $m + m' = G$ will be constant as the total atmospheric pressure p is constant. A certain amount of water-vapour will diffuse through an imaginary sphere with radius a , this amount being equal to

$$W = -4\pi a^2 D \frac{\partial m}{\partial a}$$

(D = constant of diffusion). As, however, there is also an air-concentration gradient of which the direction is opposed to that of the vapour-density gradient, there will, at the same time, also diffuse per second an amount of air in the opposite direction; this amount is equal to

$$L = -4\pi a^2 D' \frac{\partial m'}{\partial a} = -4\pi a^2 D' \frac{\partial (G-m)}{\partial a} = 4\pi a^2 D' \frac{\partial m}{\partial a}$$

(D' = constant of diffusion of air). At the surface of the drop the density of the air remains constant; a radially directed atmospheric flow of air and of vapour must, therefore, exist at the same time, neutralizing this diffusion-flow of air. [2]

Writing v for the velocity of this radial flow, we have

$$4\pi a^2 \left(D' \frac{\partial m}{\partial a} + m'v \right) = 0$$

or

$$v = -\frac{D'}{m'} \frac{\partial m}{\partial a}$$

As this radial flow carries also vapour with it, the total vapour-flow will be

$$W + 4\pi a^2 m v = 4\pi a^2 \left(-D \frac{\partial m}{\partial a} + m v \right) = -4\pi a^2 D \frac{\partial m}{\partial a} \left(1 + \frac{m}{m'} \cdot \frac{D'}{D} \right)$$

and this value is also equal to the source-strength of the drop, namely C/M , so that

$$W + 4\pi a^2 m v = \frac{C}{M} = -4\pi a^2 D \frac{\partial m}{\partial a} \left(1 + \frac{m}{m'} \cdot \frac{D'}{D} \right). \quad (5)$$

D and D' are of the same order of magnitude. We shall presently compute in this publication the diffusion-constant by means of a theoretically deduced formula. As such a deduction can of course only give a more or less accurate approximation to reality, the error introduced by applying such a formula is of the same order as that made by putting D equal to D' . It is therefore consistent to introduce this simplification here. We can then write for (5):

$$\frac{C}{M} = -4\pi a^2 D \frac{\partial m}{\partial a} \left(1 + \frac{m}{m'} \right) = -4\pi a^2 D \frac{\partial m}{\partial a} \cdot \frac{G}{(G-m)} \quad (6)$$

Integration gives

$$\frac{C}{4\pi D M a} = -G \cdot \ln \frac{(G-m)}{(G-m_\infty)} = \frac{m-m_\infty}{1-\frac{m_\infty}{G}} - \left(\frac{m-m_\infty}{1-\frac{m_\infty}{G}} \right)^2 \frac{1}{2G} + \dots \quad (7)$$

(m_∞ is the vapour-density at infinity).

In practice the last term in the right-hand member is so small, that we can safely omit it. Introducing mRT for e and GRT for p , we obtain

$$\frac{e-e_\infty}{1-\frac{e_\infty}{p}} = \frac{CRT}{4\pi D M a}. \quad (8)$$

The formula for the atmospheric radial flow \vec{v} becomes:

$$\vec{v} = \frac{D'}{m'} \frac{\partial m'}{\partial a} (\vec{a}) = D \frac{\partial \ln(G-m)}{\partial a} (\vec{a}) = \frac{C}{4\pi M G a^2} (\vec{a}) = -\text{grad} \frac{C}{4\pi M G a}.$$

(\vec{a} is the unit-vector directed radially outwards), while

$$\text{div} \vec{v} = -\text{div} \text{grad} \frac{C}{4\pi M G a} = 0,$$

so that the radial flow is divergence-free.

The vapour-pressure e_g at the distance λ' from the surface of the drop, as determined by (8) is:

$$e_g = e_\infty + \left(1 - \frac{e_\infty}{p} \right) \frac{CRT}{4\pi D M (r + \lambda')},$$

but also by (4):

$$e_g = E_r - \frac{C}{4\pi r^2} \sqrt{\frac{2\pi RT}{M}}.$$

Equalizing these two expressions for e_g gives:

$$E_r - \frac{C}{4\pi r^2} \sqrt{\frac{2\pi RT}{M}} = e_\infty + \left(1 - \frac{e_\infty}{p} \right) \frac{CRT}{4\pi D M (r + \lambda')},$$

from which C can be solved. The result is:

$$C = \frac{(E_r - e_\infty) 4\pi r^2}{\sqrt{\frac{2\pi RT}{M}} + \frac{RT r (1 - \frac{e_\infty}{p})}{DM (1 + \frac{\lambda'}{r})}}. \quad (9)$$

It follows from (9) that the source-strength of the drop is determined by the vapour-pressure at infinity, the radius and the temperature of the drop and its surroundings. The value of $\frac{\lambda'}{r}$ for drops with a radius larger than 10^{-4} cm is very much smaller than 1, so that

for these drops this term can be omitted in (9). For smaller drops the effect of $\frac{\lambda'}{r}$ is an apparent increase of D , amounting to a few %. This apparent increase of D also takes place when a drop falls under the influence of gravity which causes some ventilation to occur, although so long as the drops still obey Stokes' law, this ventilation is slight [2] and the apparent increase of D amounts to only a few %. For drops with a radius smaller than 1.7×10^{-2} cm the falling-velocity is computed from Stokes' law.

Fig. 1 shows the lines of flow of the vapour in the case of two drops at a certain distance apart.

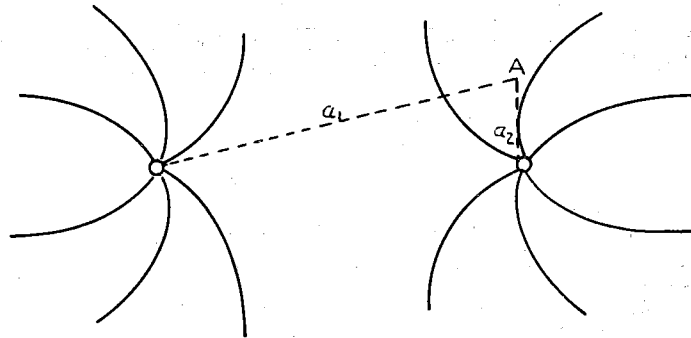


Fig. 1

The vapour-pressure at A depends then on the properties of the drops and on the distances of A from these drops. If many drops are present, a_i denoting the distance of the drop i from A and C_i its source-strength, it may be proved that the vapour-density in A is given by

$$m = G - (G - m_\infty) \cdot \varepsilon^{-\Sigma \frac{C_i}{4\pi DMGa_i}}, \quad (10)$$

or the vapour-pressure e in A by

$$e = p - (p - e_\infty) \cdot \varepsilon^{-\Sigma \frac{C_i RT}{4\pi DMpa_i}}, \quad (11)$$

while the atmospheric radial flow \vec{v} in A is determined by the vector-sum

$$\vec{v} = -\text{grad} \sum \frac{C_i}{4\pi MGa_i} = \sum \frac{C_i}{4\pi MGa_i^2} (\vec{a}_i). \quad (12)$$

The proof that the description of the flow by (10), (11) and (12) is correct, is as follows: For the total transfer of vapour I across an arbitrary surface we have: (denoting by do an element of superficial extent and by $d\tau$ a volume-element inside this surface)

$$\begin{aligned} I &= \iint [-D \text{grad} m + m\vec{v}] \cdot \vec{do} = \iint \left[-D(G-m) \text{grad} \sum \frac{C_i}{4\pi DMGa_i} - m \text{grad} \sum \frac{C_i}{4\pi MGa_i} \right] \cdot \vec{do} = \\ &= -\iint \text{grad} \sum \frac{C_i}{4\pi Ma_i} \cdot \vec{do} = -\iiint \text{div grad} \sum \frac{C_i}{4\pi Ma_i} d\tau. \end{aligned}$$

The last integral is 0 if none of the drops is surrounded by the surface, as then $\text{div grad} \frac{1}{a_i} = 0$. If however, the drop with suffix k is surrounded, the integral can be transformed into

$$\begin{aligned} I_k &= -\iint \text{grad} \sum' \frac{C_i}{4\pi Ma_i} \cdot \vec{do} - \iint \text{grad} \frac{C_k}{4\pi Ma_k} \cdot \vec{do} = -\iiint \text{div grad} \sum' \frac{C_i}{4\pi Ma_i} d\tau - \iint \text{grad} \frac{C_k}{4\pi Ma_k} \cdot \vec{do} = 0 - \\ &= -\iint \text{grad} \frac{C_k}{4\pi Ma_k} \cdot \vec{do} \left(\sum' \text{ means a summation over all drops except the one with suffix } k \right). \end{aligned}$$

The last integral is the sum of two integrals, one over the surface σ of a sphere with radius a enclosing the drop K and lying within the surface o and the other over the volume τ' between these two surfaces. The latter integral is 0. We have therefore,

$$I_k = - \iint \text{grad} \frac{C_k}{4\pi M a_k} \cdot \vec{d\sigma} = - \iiint \text{div grad} \frac{C_k}{4\pi M a_k} d\tau' - \iint \text{grad} \frac{C_k}{4\pi M a_k} \cdot \vec{d\sigma} = 0 + \iint \frac{C_k}{4\pi M a_k^2} \cdot a^2 \cdot d\omega = \frac{C_k}{M},$$

so that it appears that the flow of vapour I_k is equal to C/M moles/sec. The conditions that the total flow must be C/M moles/sec if the drop K is surrounded by the surface and 0 if no drop is surrounded are thus satisfied, the representation by means of (10), (11) and (12) is correct and $\text{div } \vec{v} = 0$ as it should be. By expanding the exponential term in (11) and omitting the terms after the second, this formula can be simplified and becomes

$$e = e_\infty + \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \sum \frac{C_i}{a_i}. \quad (13)$$

We shall make use of (13) to calculate the source-strength of a drop (without suffix) surrounded by other drops (with suffix i). For simplicity we put the distance a_i of a point on the surface of a drop to the centre of an other drop equal to the distance between the two centres of the drops. From (13) we have for $e = e_g$:

$$e_g = e_\infty + \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \frac{C}{r + \lambda'} + \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \sum' \frac{C_i}{a_i}.$$

Another expression for e_g follows from (4), namely: $e_g = E_r - \frac{C}{4\pi r^2} \sqrt{\frac{2\pi RT}{M}}$.

From these two equations C can be determined as follows:

$$E_r - e_\infty = C \left[\frac{1}{4\pi r^2} \sqrt{\frac{2\pi RT}{M}} + \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \frac{1}{(r + \lambda')} \right] + \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \sum' \frac{C_i}{a_i},$$

or, writing $E_r = E \left(1 + 3,25 \cdot 10^{-5} \frac{1}{rT}\right)$,

$$C = \frac{E \left(1 + 3,25 \cdot 10^{-5} \frac{1}{rT}\right) - e_\infty - \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \sum' \frac{C_i}{a_i}}{\sqrt{\frac{2\pi RT}{M}} + \frac{RT}{DM} \left(1 - \frac{e_\infty}{p}\right) \cdot \frac{1}{r}}. \quad (14)$$

When there are n drops we compute the n C_i 's from n equations analogous to (14). These quantities are therefore known, if the mutual distances of the drops, their temperature and size and the vapour-pressure at infinity are known.

For the computation of D , we use the following expression, which is based on theoretical considerations [4]:

$$D = \frac{2\lambda v}{3\pi} = \frac{2}{3\pi} \cdot \lambda \sqrt{\frac{3RT}{M}},$$

where v denotes the mean velocity of a molecule of water. Now the diameter of such a molecule does not differ much from those of the gas-molecules of the air, so that with sufficient accuracy we can write for λ :

$$\lambda = \frac{1/2 \sqrt{2}}{4\pi r^2 \cdot n} = \frac{1/2 \sqrt{2} RT}{4\pi r^2 \cdot Np},$$

where r means the radius of a molecule ($= 1,6 \times 10^{-8}$ cm) and n the number of gas molecules per cm^3 . Giving r , R , N and M their numerical values, we obtain for D :

$$D = \frac{2 \cdot 1/2 \cdot \sqrt{2} \cdot 8,3 \cdot 10^7 \cdot T \cdot \sqrt{\frac{3 \cdot 8,3 \cdot 10^7 \cdot T}{18}}}{3\pi \cdot 4\pi \cdot 1,6^2 \cdot 10^{-16} \cdot 6,03 \cdot 10^{23} \cdot p} = 23,57 \frac{T^{3/2}}{p},$$

and by this (14) becomes:

$$C = r^2 \frac{\left[E \left(1 + 3,25 \cdot 10^{-5} \frac{1}{rT} \right) - e_\infty \right] \sqrt{T} - 1,544 \cdot 10^4 (p - e_\infty) \sum \frac{C_i}{a_i}}{1,544 \cdot 10^4 (p - e_\infty) \cdot r + 429,07 \cdot T} \quad (15)$$

Putting $T = 280^\circ \text{K}$, $p = 800 \text{ mb} = 8 \times 10^5 \text{ dyn/cm}^2$, we have $E = 10 \text{ mb} = 10^4 \text{ dyn/cm}^2$.

By putting $e_\infty = E$, which is equivalent to putting the mean relative humidity in the cloud 100 %, we can write for (15):

$$C = \frac{1,94 \cdot 10^{-5} \cdot r - 12,198 \cdot 10^6 \cdot r^2 \sum \frac{C_i}{a_i}}{12,198 \cdot 10^6 \cdot r + 120,148} \text{ gram/sec.} \quad (16)$$

If we put the second term in the numerator of (16) equal to zero, C represents the source-strength of an isolated drop. This strength is given in column 2 of table 1.

We proceed next to compute the influence of the other drops on the source-strength of the drop K . Let us imagine a spherical cloud of drops, with a concentration of l drops per cm^3 and let us divide this cloud into thin spherical layers with radii a_i and thickness da . The drops present in such a layer have then a total source-strength of

$$dC' = 4\pi a^2 l \cdot C_a \cdot da,$$

where C_a is the mean source-strength of a drop in the layer with radius a .

Owing to the radial symmetry in the cloud, we may assume C_a to depend only on a . The term $\sum \frac{C_i}{a_i}$ relative to a point K is analogous to the expression for the electric potential in a point K , due to charged particles, so that we can apply the results of the theory of the electric field directly to the present computation for $\sum \frac{C_i}{a_i}$. In a point K , outside the cloud at a distance S from its centre, we can therefore write:

$$\sum \frac{C_i}{a_i} = \frac{1}{S} \int_0^A 4\pi a^2 \cdot l \cdot C_a \cdot da = \frac{4/3 \pi A^3 \cdot l \cdot \bar{C}}{S} \quad (17)$$

\bar{C} denoting the mean source-strength of a drop, computed over the whole cloud. For K inside the cloud at a distance S from its centre we have:

$$\sum \frac{C_i}{a_i} = \int_S^A \frac{4\pi a^2 \cdot l C_a \cdot da}{a} + \frac{1}{S} \int_0^S 4\pi a^2 l C_a \cdot da = \int_S^A l C_a d(2\pi a^2) + \frac{1}{S} \int_0^S l C_a \cdot d(4/3 \pi a^3) \quad (18)$$

TABLE 1

r cm	C gram/sec	evaporation-time t
10^{-1}	$1,6 \cdot 10^{-12}$	3.10 ⁴ days
10^{-2}	$1,6 \cdot 10^{-12}$	31 days
10^{-3}	$1,5 \cdot 10^{-12}$	45,7 minutes
10^{-4}	$1,4 \cdot 10^{-12}$	3,1 sec
10^{-5}	$0,8 \cdot 10^{-12}$	$0,7 \cdot 10^{-2}$ sec

Combining (13) and (17) we obtain:

$$e = e_{\infty} + \frac{RT}{4\pi DM} \left(1 - \frac{e_{\infty}}{p}\right) \sum \frac{C_i}{a_i} = e_{\infty} + \frac{RT}{4\pi DM} \left(1 - \frac{e_{\infty}}{p}\right) \frac{4/3 \pi A^3 \cdot l \cdot \bar{C}}{S}.$$

In this expression the whole spherical cloud acts, so to speak, as one large drop with radius A and a source-strength

$$C_w = 4/3 \pi \cdot A^3 \cdot l \cdot \bar{C}.$$

In order to estimate this strength we assume $e_{\infty} = 0$ and $e = E = 10$ mb, while for the other quantities we use the values already previously assigned to them.

Then:

$$C_w = 4/3 \pi A^3 \cdot l \bar{C} = \frac{10^4 \cdot S}{12,2 \times 10^4} = 8,2 \times 10^{-7} \cdot S.$$

For a cloud with radius S of the order of a few hundred meters this becomes:

$$C_w = 10^{-2} \text{ gram/sec} = 36 \text{ gr/hour}.$$

The mean source-strength \bar{C} is then of the order of magnitude $10^{-15}/l$, so that, if l is of the order of 10–10.000 drops per cm^3 , we have

$$\bar{C} \approx 10^{-16} - 10^{-19},$$

from which it appears that the source-strength of a drop in a cloud can be many times smaller than the strength of an isolated drop.

In an inward direction C decreases, so that \bar{C} is chiefly determined by the value in the outer layer of the cloud.

An isolated drop will evaporate slowly; we obtain the time necessary for this process by putting C in (16) equal to $4\pi r^2 dr/dt$ and by then solving the resulting differential equation. To this end we suppose the flow to be quasi-stationary, so that it will adjust itself instantaneously to the changing radius of the drop; then (16) is transformed into

$$4\pi r^2 \frac{dr}{dt} = 10^{-12} \cdot r^2 \cdot \frac{1,94}{1,22 r^2 + 1,2 \cdot 10^{-5} \cdot r},$$

from which we obtain

$$t = 4\pi \cdot 10^{12} \int_0^r \frac{1,22 r^2 + 1,2 \cdot 10^{-5} \cdot r}{1,94} dr = 0,4 \cdot 10^8 \cdot r^2 + 2,7 \cdot 10^{12} \cdot r^3 \text{ sec.}$$

Values of this evaporation-time are given in table 1, column 3. From these values one gathers that drops with a radius $> 10^{-3}$ cm., once formed, can exist for a fairly long time in an atmosphere of 100 % relative humidity, while smaller drops will disappear quickly. In a cloud it will take a still longer time before the drops are evaporated. These times may differ by a factor 10^4 to 10^7 , so that there even drops with a radius of 10^{-5} cm. can exist for hours, when all drops are of the same size. When drops of different size are mixed, the smaller drops will disappear in the time of column 3, tab. 1.

In the above deductions three conditions have been introduced:

1. the atmosphere is at rest and the drop does not move;
2. the equilibrium of the flow of vapour adjusts itself instantaneously;
3. latent heat is locally supplied or withdrawn.

As regards 1: If the drop should move and its motion should obey Stokes' law, the above formulas would be influenced in such a way, that D would apparently increase a few %. This would but slightly alter the results in table 1, as, moreover, D has not been computed very accurately.

As regards 2: We shall here prove that for the drops to which the data in Table I refer, the time necessary for the flow of vapour to become stationary, is very much shorter than the evaporation-time.

If, namely, a drop of radius r is suddenly placed in an atmosphere, the concentration of the vapour in moles per cm^3 , m , is determined by

$$m = m_{\infty} + \frac{2(m_g - m_{\infty})r}{a\sqrt{\pi}} \int_{\frac{a-r}{2\sqrt{Dt}}}^{\infty} e^{-x^2} \cdot dx.$$

which satisfies the conditions $m = m_{\infty}$ for $t = 0$ and $a > r$, and $m = m_g$ for $a = r$.

The drop is here supposed to create instantaneously the vapour-pressure e_g at its surface. The rate of evaporation $\frac{C}{M}$ is determined by the gradient of the vapour-concentration at the surface of the drop (6).

$$\begin{aligned} \frac{C}{M} &= \left(-4\pi a^2 D \frac{\partial m}{\partial r} \right)_r = \left[4\pi D (m - m_{\infty}) a \right]_r + \left[\frac{2(m_g - m_{\infty})r}{a\sqrt{\pi}} e^{-\frac{(a-r)^2}{4Dt}} \frac{1}{2\sqrt{Dt}} \cdot 4\pi D a^2 \right]_r = \\ &= 4\pi D (m_g - m_{\infty}) r + 4\pi D (m_g - m_{\infty}) \frac{r^2}{\sqrt{\pi D t}} = 4\pi D (m_g - m_{\infty}) r \left(1 + \frac{r}{\sqrt{\pi D t}} \right). \end{aligned}$$

For $t = \infty$, this becomes

$$\frac{C_{\infty}}{M} = 4\pi D (m_g - m_{\infty}) r,$$

in agreement with formulas, already deduced.

We have then:

$$C = C_{\infty} \left(1 + \frac{r}{\sqrt{\pi D t}} \right). \quad (18a)$$

In our case $\sqrt{\pi D}$ is equal to 0,67, so that even for large drops of 1 mm radius the correction will be only 1% already after 3 or 4 minutes, for drops of $\frac{1}{10}$ mm radius already after 2,2 seconds. The evaporation-times given in Table 1 are very much longer than the times, here deduced for the flow to become stationary, so that it was perfectly legitimate in computing the former to consider the latter to be infinitely small.

As regards 3: A temperature-gradient will be created, which will cause the required latent heat to be supplied (or withdrawn). The necessary supply is 590 C cal through a spherical surface of radius a , so that

$$590 C \text{ cal} = j \frac{dT}{da} \cdot 4\pi a^2 \quad (19)$$

Here j denotes the heat conduction coefficient, for which we have:

$$j = \frac{n\lambda}{3} \sqrt{\frac{3RT}{M}} \cdot C_v = 4,56 \cdot 10^{-5}.$$

Integration of (19) gives

$$\int dT = \frac{-590 C}{4\pi j} \int d \frac{1}{a},$$

$$T_g - T_{\infty} = \frac{-590 C}{4\pi j} \frac{1}{r} = \frac{-590 C}{4\pi \cdot 4,56 \cdot 10^{-5}} \cdot \frac{1}{r} = -1,64 \cdot 10^6 \cdot \frac{C}{r} = -1,56 \cdot 10^{-6} \cdot \frac{1}{r}.$$

In the last term the value $1,6 \times 10^{-12}$ has been used for C . It is clear from the above, that the temperature-difference between the drop and points at infinite distance is very small, so that it can be neglected in our formulas.

We shall now consider the question whether the drops, present in a drizzle, can be formed by the coalescence of smaller drops. We start from the extreme case of a few % over-saturation, say, for example, $e_\infty - E = 0,28 \text{ mb} = 280 \text{ dyn/cm}^2$.

Relative to this value, the difference between E_r and E is very much smaller, so that we can replace E_r by E . For an isolated drop we have then: (14), (15),

$$C = \frac{RT}{DM} \cdot r \left(1 - \frac{e_\infty}{p} \right) + \sqrt{\frac{2\pi RT}{M}} = r^2 \frac{-280 \cdot \sqrt{265}}{r \cdot 1,544 \cdot 10^9 \cdot 7 + 265 \cdot 429,07},$$

from which we compute the time of accretion by writing $4\pi r^2 dr/dt = C$, so that

$$C = 4\pi r^2 \cdot \frac{dr}{dt} = r^2 \frac{-280 \sqrt{265}}{r \cdot 1,544 \cdot 10^9 \cdot 7 + 265 \cdot 429,07} = \frac{r^2}{2,37 \cdot 10^6 \cdot r + 25},$$

$$dt = 4\pi dr (2,37 \cdot 10^6 r + 25) \therefore t = \left[1,48 \cdot 10^7 r^2 + 314 r \right]_{r_2}^{r_1} \text{ sec} = \left[4125 r^2 + 0,088 r \right]_{r_2}^{r_1} \text{ hours}.$$

The second column of Table 2 gives the times of accretion, computed in this way, starting from a drop of 10^{-5} cm radius.

TABLE 2

r cm	t	2,8%	t	0,1%
10^{-1}	41,3 hours		48 days	
10^{-2}	24,5 min		11,5 hours	
10^{-3}	15 sec		7 min	
10^{-4}	0,18 sec		5 sec	

It is clear from these values, that the formation of drizzle drops (radius 10^{-1} cm) would take an exceedingly long time under the circumstances prevailing in practice. To shorten this time a much higher degree of oversaturation would have to be assumed, the former being inversely proportional to the latter. Indeed, for the formation of a drizzle drop in half an hour an over-saturation of a few hundred % would be necessary, a case never noticed in practice. On the other hand an ice crystal of the same weight as a raindrop can be formed when an excess of vapour pressure of 0,28 mb exists, this being the difference between the pressures of saturated vapour of ice and of under-cooled water at -12°C . The area of the ice crystal surface can, namely, be very much larger than that of the drop and this would shorten the accretion-time very considerably as, instead of $4\pi r^2$, the area of the crystal surface must be used in the formula. It is true that the vapour-molecules will partly rebound from the ice-surface, by which the time of formation will be lengthened, but this effect will be very small compared with the effect of the larger surface. If the ice-crystal is rod-shaped, the times of formation can obtain plausible values. During the formation of clouds over-saturations of several 0,1 % do indeed occur in practice.

Table 2 gives in column 3 the accretion-times of drops for 0,1 % over-saturation. As appears from these data, drops of 10^{-3} cm radius can be formed in about 10 minutes, while larger drops require a very much longer time. For that reason, when cu-clouds are formed, mainly drops with radii of the order of 10^{-3} cm will be present, as smaller drops can quickly attain that size and larger drops have not sufficient time to form, since this is a matter of hours.

It can be gathered from Table 1, that when the formation of cu-clouds is over and the relative humidity of the clouds is 100 %, as could be the case for a cloud with drops of

different sizes, the drops with a radius smaller than 10^{-3} cm are bound to have disappeared after a few minutes, while those with a radius of 10^{-3} cm will still be present. It follows from this that in cu-clouds with a formation-time of the order of 10 minutes drops with a radius of about 10^{-3} cm or 10μ will largely predominate. Fog, however, as, for example, radiational fog requires a longer time for formation and it is questionable whether the relative humidity of the latter attains an amount of 100 %.

Advective fog will result from an over-saturation of, say, 0.1 %, maintaining itself during a few hours. A great many drops with a radius of $\pm 50 \mu$ will be formed under these circumstances.

Generally speaking, a layer of fog has a longer duration than cu-clouds and the question arises, therefore, whether droplets with a radius of 10^{-3} cm can hold their own in a fog. The falling velocity of such drops is 50 meter per hour, so that a low-hanging fog, lasting for 4 hours cannot contain these drops unless violent turbulence carries them upwards or unless they are continually formed anew. In the case of radiational fog, however, the turbulence is mostly rather weak. There is, moreover, still another effect, namely, coalescence, which causes these drops to vanish fairly quickly, as will be shown below. The question whether two drops will coalesce when they contact has not yet been answered satisfactorily. As we shall deduce below, very strong attractive forces can exist between drops and these may influence the coalescence favourably. On the other hand the drops can be prevented from coalescing by a layer of air clinging very closely to their surfaces, covering them, so to speak, by a thin skin. For the time being, we shall suppose the droplets to coalesce on contact.

§ 2. The falling velocity of a drop is governed by Stokes' law, so long as the Reynold number, belonging to this motion is less than 50. For a Reynold number higher than 160.000 Newton's law is valid and for intermediate values an empirically deduced law exists, so that the resistivity is proportional to the velocity, raised to a power between 1 and 2. The Reynold number in our case is $\rho v r / \eta = R_e$, where ρ represents the density of the air and η the viscosity-coefficient of the air. According to Stokes' law we have:

$$\frac{4}{3} \pi r^3 \cdot g = 6 \pi \eta r \cdot v,$$

or

$$\frac{2g\rho}{9\eta^2} r^3 = \frac{\rho \cdot v \cdot r}{\eta},$$

so that

$$R_e = \frac{2g\rho}{9\eta^2} r^3 = 0,98 \cdot 10^7 \cdot r^3 \leq 50,$$

from which we obtain

$$r \leq 1,7 \cdot 10^{-2} \text{ cm.}$$

Drops with a smaller radius will obey Stokes' law, those with a larger radius will fall slower than would follow from that law. The value $R_e \leq 50$ has been taken from experiments with spheres, which naturally, did not consist of water. As, however, the limiting values for R_e depend largely on the degree of turbulence in the boundary-layer, that is, on the degree of roughness of the surface, it may very well be that also larger drops, than those mentioned above, still obey Stokes' law, as, indeed, a drop of water is presumably "smoother" than a wooden ball. Let us suppose Stokes' law to remain valid for drops with a radius up to 10^{-1} cm; by this the computed coalescence-effect will be slightly raised, so that the real effect will be smaller than our computed result. It will appear, from our computations that the coalescence of droplets into one drop with a radius of 10^{-2} cm demands the greater part of the time while a further accretion to drops with a larger radius is a much quicker process. For that reason it will suffice to restrict ourselves to Stokes' law only in computing the coalescence-effect.

The probability of selecting from among the drops in 1 cm³ of an atmosphere a drop with a radius between r and $r + dr$ will depend, apart from r , on x , the height in the cloud, and on t , the age of the cloud, so that we shall write for it $W(r, x, t) dr$. The statistical distribution will change as to place and time, by a slow sedimentation and also by coalescence.

We suppose the atmosphere to be free from accelerations.

Consider a droplet A with radius R . In the time dt it will encounter dB droplets with radius R' , for which we have:

$$dB = W(R', x, t) dR' \cdot 1,3 \cdot 10^6 (R^2 - R'^2) \pi (R + R')^2 \cdot dt. \quad (20)$$

Indeed, the number N_B of droplets B in unit-volume is:

$$N_B = W(R', x, t) dR'.$$

The velocity of A relative to these droplets is $v - v'$, where

$$v - v' = \frac{4/3 \pi R^3 \cdot g}{6\pi\eta \cdot R} - \frac{4/3 \pi R'^3 \cdot g}{6\pi\eta R'} = 1,3 \cdot 10^6 (R^2 - R'^2).$$

The combined cross-section O of A and of the drops grazed by A is given by

$$O = \pi (R + R')^2,$$

and these expressions together lead to (20).²⁾

All drops with $R' < R$ are overtaken by A so that A coalesces in the time dt with an amount of water, which is obtained by multiplying (20) by $4/3 \pi R'^3$ and integrating:

$$\int_0^R dB \cdot 4/3 \pi \cdot R'^3 = dt \int_0^R W(R', x, t) dR' \cdot 4/3 \pi R'^3 \cdot 1,3 \cdot 10^6 (R^2 - R'^2) \pi (R + R')^2,$$

which will cause an increase of volume of A equal to $4\pi R^2 dR$

We have then:

$$\begin{aligned} dt \int_{R'=0}^{R'=R} W(R', x, t) dR' \cdot 4/3 \pi R'^3 \cdot 1,3 \cdot 10^6 (R^2 - R'^2) \pi (R + R')^2 &= 4\pi R^2 \cdot dR, \\ dt \cdot \frac{1,3 \cdot 10^6}{4} \int_{R'=0}^{R'=R} W(R', x, t) dR' \cdot 4/3 \pi R'^3 \left\{ 1 - \left(\frac{R'}{R}\right)^2 \right\} \left(1 + \frac{R'}{R} \right)^2 &= \frac{dR}{R^2} = -d \frac{1}{R}. \end{aligned}$$

The integrand contains the product

$$\left\{ 1 - \left(\frac{R'}{R}\right)^2 \right\} \left(1 + \frac{R'}{R} \right)^2,$$

with $R' < R$. This product will have a maximum value for $\frac{R'}{R} = 1/2$, so that this value becomes

$$(1 - 1/4) (3/2)^2 = 1,7.$$

On substituting this maximum value in the integral, its computation will yield a too high coalescence-effect. By substituting $\frac{R'}{R} = 0$, the product becomes 1. Putting $\left(1 + \frac{R'}{R} \right)^2 \left\{ 1 - \left(\frac{R'}{R}\right)^2 \right\} = 1,5$ in the average, we find:

$$1,5 \int_{R'=0}^{R'=R} W(R', x, t) \cdot 4/3 \pi R'^3 dR' = 1,5 m',$$

where m' stands for the total amount of water in 1 cm³, contained in all drops for which $R' \leq R$. The number of drops, however, overtaking other drops and not being themselves

²⁾ During corrections of the proof, the writer read the article of I. LANGMUIR: The production of rain by a chain reaction in cumulus clouds at temperatures above freezing; J. of Met. 1948, 5, 5, 175—192. In this article coalescence is calculated in quite a different way, considering a collection efficiency $E < 1$, which should be admitted as a factor at the right side of (20).

overtaken by larger drops is not considerable, so that we assume that m' is practically equal to the water content of all drops. We call the water-content of the atmosphere $m \cdot 10^{-6}$ if m gram of water is present in 1 m^3 of air. By replacing m' by $m \cdot 10^{-6}$ the coalescence-effect will once more come out too high. The final result is:

$$-d \frac{1}{R} = dt \cdot \frac{1,3 \cdot 10^6}{4} \cdot 1,5 m \cdot 10^{-6} \left[-\frac{1}{R} \right]_{R=R_0}^{R=R} = \frac{1,3 \cdot 1,5 \cdot m}{4} \left[t \right]_{t=0}^{t=t},$$

R_0 denoting the initial and R the final radius. The time, necessary for the coalescence, is

$$t = \frac{2}{m} \left(\frac{1}{R_0} - \frac{1}{R} \right),$$

and we have also

$$-\frac{dx}{dt} = V = 1,3 \cdot 10^6 \cdot R^2,$$

from which it follows, that

$$dx = 1,3 \cdot 10^6 \cdot R^2 \cdot dt = 1,3 \cdot 10^6 \cdot R^2 \cdot \frac{4}{1,3 \cdot 1,5 \cdot m} d \left(-\frac{1}{R} \right) = \frac{4 \cdot 10^6}{1,5 m} dR, \quad |\Delta x| = \frac{2,67 \cdot 10^6}{m} (R - R_0) \text{ cm},$$

where $|\Delta x|$ represents the vertical distance covered in the cloud. Expressed in hectometers, this becomes:

$$|\Delta x| = \frac{267}{m} (R - R_0) \text{ hectometers},$$

and the expression for t in minutes is

$$t = \frac{1}{30 m} \left(\frac{1}{R_0} - \frac{1}{R} \right) = \frac{1}{30 \cdot m \cdot R_0} \left(1 - \frac{R_0}{R} \right) \text{ minutes.} \quad (22)$$

From (21) and (22) we obtain

$$|\Delta x| \cdot t = \frac{267}{30 \cdot m^2} \cdot \frac{R}{R_0} \left(1 - \frac{R_0}{R} \right)^2 \text{ Hectometers} \cdot \text{minutes}.$$

Table 3 gives the number of hectometers and minutes, necessary to increase the radius of a drop by coalescence from a value in the first column to a value in the first row.

Fig. 2 is a graphical representation of this table.

TABLE 3 ($m = 2$)

r cm	10^{-4}	10^{-3}	10^{-2}	10^{-1}
10^{-4}	0 hm/0 min	0,13 hm/2,7 hr	1,3 hm/3 hr	13,2 hm/3 hr
10^{-3}		0 hm/0 min	1,2 hm/15 min	13 hm/16 $\frac{1}{2}$ min
10^{-2}			0 hm/0 min	12 hm/1 $\frac{1}{2}$ min
10^{-1}				0 hm/0 min

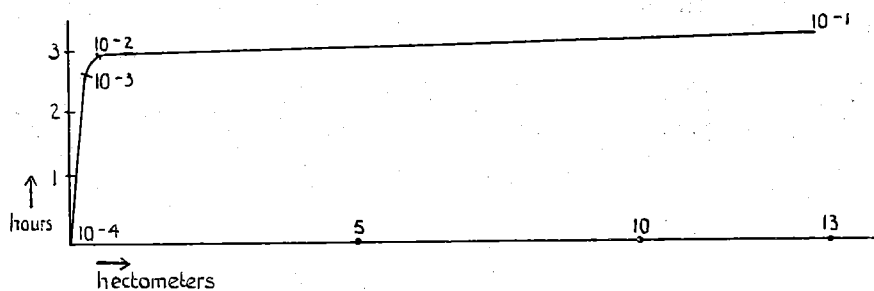


Fig. 2 ($m = 2$)

Table 3 gives extreme values on the low side, as our computation has made the effect come out too favourable. The result shows that the formation of a drop in a drizzle requires the falling of the increasing drop through a fairly large distance. If stratus-layers are of this thickness, small drops might fall out of them and reach the earth, provided they do not evaporate completely on their way. A thickness of 13 hectometer is, however, enormous. If this thickness is not available the drop will certainly have left the cloud in a quarter of an hour, for, either the drop will have attained a high velocity by accretion, or the thickness of the cloud is not sufficient, in which case the drop will already have traversed the cloud before then. This refers to drops with a radius $> 10^{-3}$ cm. For drops with smaller radii conditions are more favourable. Such drops require hours for their radius to reach the value 10^{-3} cm. If coalescence is sure to occur on contact of two drops, one must draw the conclusion that in radiational fog of small height (< 100 meter) drops with a radius of 10^{-3} cm cannot be present if this fog is to last for a few hours. It is true that a turbulence may carry these drops upwards in parcels of air, but relatively to these parcels they will continue to fall and to coalesce with other drops and will have reached the earth in a quarter of an hour, as in that time they will have grown to be drizzle-drops, if they have not reached the earth already before then. The turbulence is then only effective in producing the drops the opportunity to fall over a sufficient number of hectometers.

If no drops of this size can exist in radiational fog but only smaller drops ($R=10^{-3}$ to 10^{-4} cm) the relative humidity in this fog can never be 100%; indeed, a slight lowering of the temperature causes already an oversaturation of say 0,1%, so that, according to table 2, the critical drops of 10^{-3} cm radius would then be formed. Radiational fog will, therefore, contain for the greater part drops with a radius smaller than 10^{-3} cm and its relative humidity will be less than 100%. These drops would thus be of the order of magnitude of the nucleus of growth. It is also possible that the relative humidity is less than 100% at the bottom side of the radiational fog and 100% at the top. In that case the following process will take place. At the top-layer, drops with the critical radius of 10^{-3} cm are formed by cooling; these drops come down (50 meter in an hour) and while doing so, coalesce, so that their falling velocity increases and they would have fallen on the earth within a quarter of an hour, but for the fact, that in falling they penetrate into regions with a relative humidity of less than 100%. That means, however, a rapid decrease of the critical drop (as is shown in table 2 for 99,9% relative humidity) and at the bottom of the layer the drop would decrease still further. In this way cold is transported from the top downwards to the bottom. By this transport of water and by this evaporation the relative humidity at the bottom can, at a certain moment, have reached 100%. The critical drop will then not be dissolved so quickly (table 1) and will rain out. This may explain how the radiational fog becomes wet and the hanging of drops on the branches of trees. This suggests further the idea to dispel radiational fog by spraying drops of the critical size on top of the layer. It must be borne in mind that the whole of the above deductions is based on the supposition that two contacting drops will invariably coalesce, a fact which has not yet been ascertained satisfactorily by experiment.

§ 3. Before proceeding to calculate the forces, which can arise from evaporation and condensation, we shall first consider any possible influence of the Brownian movement on the evaporation-equilibrium. The displacement Δx of a droplet in t seconds is given by Einstein-Perrin's formula

$$\Delta x = \sqrt{\frac{RT \cdot t}{3\pi\eta \cdot r \cdot N}} = \sqrt{\frac{8,3 \cdot 10^7 \cdot 300 \cdot t}{3 \cdot 3,14 \cdot 1,7 \cdot 10^{-4} r \cdot 6,03 \cdot 10^{23}}} = 5 \cdot 10^{-6} \sqrt{\frac{t}{r}}$$

If the final steady evaporation may differ from the actual one by an amount of 1% we must have: (18a)

$$\frac{r}{0,67 \sqrt{t}} = \frac{1}{100}$$

(see above), so that

$$\Delta x = 5 \cdot 10^{-6} \sqrt{r} \times \frac{100}{0,67} = 7,5 \times 10^{-4} \sqrt{r},$$

where Δx denotes the distance over which the particle has moved after a time t , during which the flow of vapour has deviated less than 1% from the ideal one. It gives us an idea of the extension of the region round the drop, inside which the vapour-pressure can differ more than 1% from the one computed from the steady flow. Table 4 gives these values in the second row. As appears from this table this region is for the drops in question very much smaller than the drops themselves, so that the Brownian movement does not have any influence on the flow and the vapour-pressure.

TABLE 4

r cm	10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}
Δx cm	$2,3 \cdot 10^{-6}$	$7,5 \cdot 10^{-6}$	$2,3 \cdot 10^{-5}$	$7,5 \cdot 10^{-5}$	$2,3 \cdot 10^{-4}$

§ 4. Throughout the whole region round the drop the pressure of the air is constant. Where molecules of water are numerous, only a small number of molecules of air will be present and the other way round. Let us write p for the total atmospheric pressure and p' for the partial one, then $p' + e = p$ is the pressure at the surface of the drop. The pressure of the air on a surface-element do of the drop is p' , that of the vapour-molecules is $\frac{1}{2} e_g$, as these molecules are absorbed by the liquid. If they were reflected a pressure of reaction would be added so that their pressure would then be e_g . From the liquid, however, molecules pass into the vapour, thereby exerting a pressure of reaction $\frac{1}{2} E_r$, if E_r represents the pressure of saturated vapour of the liquid underneath its surface. The total inward pressure on do is, in this way, $p' + \frac{1}{2} (e_g + E_r) = p' + e_g + \frac{1}{2} (E_r - e_g) = p + \frac{1}{2} (E_r - e_g)$. The pressures p and E_r are constant over the whole surface and the forces arising on this account, neutralize each other. For the resulting force \vec{R} , acting on the surface we find, therefore,

$$\vec{R} = -\frac{1}{2} \iint e_g |\vec{n}| do = \frac{1}{2} \iint e_g \vec{n} do,$$

\vec{n} denotes the unit vector drawn normally outwards.

The drop experiences also a force \vec{K} exerted by the flow of convection. This Stokes' force is given by:

$$\vec{K} = 6\pi\eta r \vec{v} = -6\pi\eta r \text{grad} \sum' \frac{C_i}{4\pi M G a_i}$$

According to (13) we have

$$e_g = e_\infty + \frac{RT}{4\pi DM} \left(1 - \frac{e}{p}\right) \sum \frac{C_i}{a_i},$$

so that

$$\begin{aligned} \vec{R} &= \frac{1}{2} \iint e_g \vec{n} do = \frac{1}{2} \iint e_\infty \vec{n} do + \frac{1}{2} \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \iint \sum \frac{C_i}{a_i} \vec{n} do = \\ &= 0 + \frac{1}{2} \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \left[\iint \frac{C}{A} \vec{n} do + \iint \sum' \frac{C_i}{a_i} \vec{n} do \right]. \end{aligned}$$

Here $\sum' \frac{C_i}{a_i}$ means a summation over all drops except the one, on which the acting force is being computed. By imagining all functions to be mathematically continued into the interior of the drop, the surface integral can be transformed into a volume-integral so far as the second integral is concerned. We transform the first integral into an integral over the volume

contained between the surface of the drop and that of the inscribed sphere and a surface-integral over this sphere. The latter integral vanishes. Denoting an element of the volume just mentioned by $d\tau'$ and the radius of the sphere by S , we have: for the first integral:

$$\iint \frac{C}{a} \vec{n} do' = \iint \frac{C}{S} \vec{s} do' + \iiint \text{grad} \frac{C}{a} d\tau' = 0 + \iiint \frac{C}{a^2} |-\vec{a}| d\tau'.$$

Writing $d\tau''$ for a volume-element inside the sphere, the second integral becomes:

$$\iint \sum' \frac{C_i}{a_i} \vec{n} do = \iiint \text{grad} \sum' \frac{C_i}{a_i} d\tau'' + \iiint \text{grad} \sum' \frac{C_i}{a_i} d\tau' = \sum' \iiint \frac{C_i}{a_i} |-\vec{a}_i| d\tau'' + \sum' \iiint \frac{C_i}{a_i^2} |-\vec{a}_i| d\tau'.$$

The expression for the resulting force is then:

$$\vec{R} = \frac{1}{2} \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \left[\sum' \iiint \frac{C_i}{a_i^2} |-\vec{a}_i| d\tau'' + \sum' \iiint \frac{C_i}{a_i^2} |-\vec{a}_i| d\tau' + \iiint \frac{C}{a^2} |-\vec{a}| d\tau' \right].$$

If we imagine the quantity $\frac{1}{2} \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right)$ to be uniformly distributed over the interior of the drop as if it were a space charge and to be such as to experience from other drops an attractive force $\frac{1}{2} \frac{RT}{4\pi MD} \left(1 - \frac{e_\infty}{p}\right) \frac{C_i}{a_i^2} |-\vec{a}_i|$, then the resultant of these forces is equal to the one calculated by us in the first and second integral. The third integral represents the force by which the bulging part of the drop is drawn towards the centre by the evaporation forces of the drop itself.

If the drop is spherical, the resultant force exerted on it by the other drops acts from the centre outwards as can be easily proved. This expression for the attractive force is analogous the one for electric forces; the resultant of the latter will also pass through the centre in the case of spherical symmetry of the charges. For a spherical drop the third integral vanishes and the total amount of the above "force experiencing quantity" becomes

$$\frac{1}{2} \frac{RT}{4\pi DM} \left(1 - \frac{e_\infty}{p}\right) \frac{4}{3} \pi r^3 = \frac{1}{6} \frac{RT}{DM} \left(1 - \frac{e_\infty}{p}\right) r^3.$$

The total force on the drop is then

$$\vec{R} + \vec{K} = \frac{1}{6} \frac{RT}{DM} \left(1 - \frac{e_\infty}{p}\right) r^3 \sum' \frac{C_i}{a_i^2} |-\vec{a}_i| + 6\pi\eta r \sum' \frac{C_i}{a_i^2} |\vec{a}_i|,$$

and the force with which one drop of source-strength C acts on an other drop with radius r

$$\vec{R} + \vec{K} = \left[\frac{RT}{6DM} \left(1 - \frac{e_\infty}{p}\right) r^3 - 6\pi\eta r \right] \frac{C}{a^2} |-\vec{a}| \text{ dynes.} \quad (23)$$

For $T = 289^\circ K$, $b = 10^6$ dynes/cm², $C = +1,5 \cdot 10^{-12}$ and $D = 23,57 \cdot \frac{T^{3/2}}{p}$ this becomes

$$\vec{R} + \vec{K} = \left[\frac{8,3 \cdot 10^7 \cdot 10^6 \left(1 - \frac{1}{100}\right)}{6 \cdot 23,57 \cdot \sqrt{289 \cdot 18}} r^3 - 6 \cdot 3,14 \cdot 1,7 \cdot 10^{-4} \cdot r \right] \frac{1,5 \cdot 10^{-12}}{a^2} =$$

$$[2,87 \cdot 10^{-3} r^3 - 4,8 \cdot 10^{-15} \cdot r] \frac{1}{a^2} |-\vec{a}| \text{ dynes.}$$

Table 5 gives this force, multiplied by the square of the distance, in column 2. Column 4 shows the factor by which these quantities must be divided in order to obtain the Coulomb force (also multiplied by the square of the distance) with which two singly charged ions act on each other. As appears from this table the forces due to evaporation and condensation are many times greater than any possible electric forces. Only for very small droplets ($< 10^{-5}$ cm)

attractive forces change into repulsive ones and vice versa. It appears that an evaporating drop attracts and a condensing drop repulses an other drop. Two drops might therefore run after each other if one is an evaporating and the other a condensing drop. The reaction-force must then be ascribed to the vapour-flow, moving in the opposite direction.

TABLE 5

r cm	$(R + K) \times a^2$	Coulomb force $\cdot a^2$	ratio
10^{-1}	$2,9 \times 10^{-6}$	$2,2 \times 10^{-19}$	$1,3 \times 10^{13}$
10^{-2}	$2,9 \times 10^{-9}$	„	$1,3 \times 10^{10}$
10^{-3}	$2,9 \times 10^{-12}$	„	$1,3 \times 10^7$
10^{-4}	$2,9 \times 10^{-15}$	„	$1,3 \times 10^4$
10^{-5}	$2,9 \times 10^{-18}$	„	13

The expression for the attractive or repulsive force contains C in the numerator and D in the denominator of the first term. For D we have here used the value of the molecular diffusion constant. The diffusion can be intensified by turbulence; in that case the same formula holds, provided we use for D the turbulence diffusion coefficient. With increasing D , the quantity C will also increase, though less than D . C is namely determined by the value of D in the immediate neighbourhood of the attracting drop, and there the value of D is only slightly higher than that of the molecular D . The D in the formula, on the other hand, refers to the surroundings of the attracted drop, where the turbulent diffusion prevails to its full extent. If the attracting force is caused by a large drop C/D will change but little by turbulence. If it is caused by a small drop C will remain practically constant, while D may increase suppose a 100 or 1000 times. Even then, however, the attractive and repulsive forces are many times stronger than the Coulomb forces.

The first-mentioned forces may play a prominent part in the coalescence process. In (23) we have used for C the value $1,5 \times 10^{-12}$ gr/sec. In the interior of a cloud extending over a few hectometers C will not have this value but a much smaller one. The attractive and repulsive forces will then be also very much weaker. The cloud as a whole, however, can exert a fairly strong force on a drop at its edge. Assigning to C the order of magnitude 10^{-2} gr/sec., (23) becomes

$$R + K = (1,91 \cdot 10^7 \cdot r^3 - 3,2 \cdot 10^{-5} \cdot r) \frac{1}{A^2} \text{ dynes.}$$

Here A denotes the radius of the spherical cloud and $\vec{R} + \vec{K}$ is the force, by which the drop is drawn towards the evaporating cloud. Putting A in the order of magnitude of 1 hectometer, we have

$$R + K = 0,191 r^3 - 3,2 \cdot 10^{-13} r \text{ dynes.}$$

The force of gravity acting on the drop is

$$K' = \frac{4}{3} \pi r^3 \cdot g = 4,1 \cdot 10^3 \cdot r^3 \text{ dynes.}$$

The ratio $\frac{R+K}{K'}$ is, therefore:

$$\frac{R + K}{K'} \approx \frac{0,191}{4,1} \times 10^{-3} = 5 \cdot 10^{-5}.$$

The sharp boundary of the cu-cloud is explained by other considerations, and it is clear from the low value of $\frac{R+K}{K'}$ that any attraction-effect will not have the least contributive influence on the sharpness of this boundary.

Experiments

Measurements have already been carried out concerning the size of drops in clouds and in fog [1], [3], but they were for the greater part selective as regards this size and mostly restricted to drops with a radius $>10^{-3}$ cm.

In a few cases, however, measurements have been made practically without any effect of selection on the result, and it was then stated that the great majority of the drops in a radiational fog have radii of less than 10^{-3} cm and that a few coalesced drops with a radius $\geq 10^{-3}$ cm moved along the observing apparatus. [1]

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