Atmospheric Diffusion Shown on a Distance-Neighbour Graph

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Atmospheric Diffusion shown on a Distance-Neighbour Graph.

By Lewis F. Richardson.

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§ 1. The Need for a New Method.

§ 1.1. *Introduction.*

If the diffusivity K of a substance whose mass per volume of atmosphere is χ be defined by an equation of Fick's type

$$\bar{u}\frac{\partial\chi}{\partial x}+\bar{v}\frac{\partial\chi}{\partial y}+\bar{w}\frac{\partial\chi}{\partial z}+\frac{\partial\chi}{\partial t}=\frac{\partial}{\partial x}\left(\mathrm{K}\frac{\partial\chi}{\partial x}\right)+\frac{\partial}{\partial y}\left(\mathrm{K}\frac{\partial\chi}{\partial y}\right)+\frac{\partial}{\partial z}\left(\mathrm{K}\frac{\partial\chi}{\partial z}\right), \qquad (1)$$

x, y, z, t being Cartesian co-ordinates and time, $\bar{u}$, $\bar{v}$, $\bar{w}$ being the components of mean velocity, then the measured values* of K have been found to be 0·2 cm.^{2} sec.$^{-1}$ in capillary tubes (Kaye and Laby's Tables), 10^5 cm.^{2} sec.$^{-1}$ when gusts are smoothed out of the mean wind (Akerblom, G. I. Taylor, Hesselberg, etc.), 10^8 cm.^{2} sec.$^{-1}$ when the means extend over a time comparable with 4 hours (L. F. Richardson and D. Proctor), 10^{11} cm.^{2} sec.$^{-1}$ when the mean wind is taken to be the general circulation characteristic of the latitude (Defant). Thus the so-called constant K varies in a ratio of 2 to a billion. The present paper records an attempt to comprehend all this range of diffusivity in one coherent scheme.

Lest the method which I shall adopt should strike the reader as queer and roundabout, I wish to justify it by showing first why some known methods are in difficulties.

§ 1.2. *Does the Wind possess a Velocity ?*

This question, at first sight foolish, improves on acquaintance. A velocity is defined, for example, in Lamb's 'Dynamics' to this effect: Let Δx be the distance in the x direction passed over in a time Δt, then the x-component of velocity is the limit of $\Delta x/\Delta t$ as $\Delta t \to 0$. But for an air particle it is not obvious that $\Delta x/\Delta t$ attains a limit as $\Delta t \to 0$.

We may really have to describe the position x of an air particle by something rather like Weierstrass's function, of which F. Klein gives an entertaining description ('Anwendung der Differential und Integralrechnung auf Geometrie,' Leipzig, B. G. Teubner, 1902), say,

$$x = kt + \sum_n (\tfrac{1}{2})^n \cos(5^n \pi t),$$

* For references to publications, see the table in § 5.2.

where k is independent of t and n is a positive integer. This gives a definite position x to the air particle, because the series of co-efficients $\frac{1}{2} + \frac{1}{4} + \frac{1}{8} + \frac{1}{16} + \ldots$ converges, and makes x a continuous function of t; but it gives no limit to $\Delta x/\Delta t$, because, as the series proceeds, the elementary waves, while becoming shorter, become also steeper. It is not suggested that these particular periods and amplitudes, which Klein chose for illustration, have anything to do with the wind.

A general and beautiful theory of "Diffusion by Continuous Movements" has been given by G. I. Taylor.* It is expressed in terms of velocity.

Although this theory of Taylor's is available, yet I think it will be a useful adventure to try now to make a theory of diffusion without assuming that $\Delta x/\Delta t$ has a limit.

§ 1.3. *The Lagrangian Specification adopted. Notation for Means. Time Rate of a Mean.*

In view of the foregoing considerations, let us not think of velocity, but only of various hyphenated velocities, such as the one-minute-velocity, or the six-hours-velocity, the words attached by the hyphen indicating the value of Δt.

The position of a particle is, however, a continuous function of time. The Lagrangian specification of fluid motion is applicable. A particle at the point (a, b, c) at time zero, is at (x, y, z) at time t.

Following Taylor, a square bracket [ ] will be used to denote a mean value, so that [A] is the mean of any quantity A. The portion of space-time over which the mean is taken will be specified as occasion arises.

Even if $(x - a)$ has no derivative with respect to t, yet $[(x - a)]$ may have such a derivative. For instance, this happens with the Weierstrassian function mentioned above, if the mean is taken over a time. Let us *assume* that $[(x - a)]$ has a derivative when taken over either a space or a time; for there is no evidence to the contrary.

§ 1.4. *A Search for Natural Mean Values.*

At first sight a good way of specifying diffusion would be to take the displacements $x - a$, $y - b$, $z - c$ of an air particle, and to form means of their powers and products such as $[x - a]$, $[(x - a)^2]$, $[(x - a)\,(y - b)]$, and the like.

But observation shows that the numerical values would depend entirely upon how large a volume was included in the mean. To see this, imagine that

* 'Proc. Lond. Math. Soc.,' Ser. 2, vol 20, Part 3 (1920).

we could introduce just two molecules of acetylene and trace their wanderings. If initially they are 10^{-5} cm. apart it seems likely, judging by what is known about molecular diffusion and by what one sees of the motion of smoke, that after travelling for one second they would still be within 10 cm. of one another. If, however, the two molecules are initially 10^{+5} cm. apart they may be caught in two gusts moving in rather different azimuths, so that after one second their separation may have altered by several metres.

Thus it appears that if y is a co-ordinate directed horizontally at right angles to the mean-wind so that $[(y - b)] = 0$, then the value obtained after a fixed time for $[(y - b)^2]$ would increase with the range of distance normal to the wind over which the mean was taken.

Is there any type of mean that forms a natural standard ? We might try—

(i) A mean over a volume so large that its exact size did not matter, a limit to $[(y - b)^2]$ being attained. This will not do, because Defant's researches show that no limit is attained within the volume of the atmosphere.

(ii) A mean taken over a definite set of molecules. Suppose that we were to let loose a sphere 0·01 cm. in diameter of acetylene, which has much the same density as air. The sphere contains about 10^{13} molecules. For the first few hundredths of a second its rate of diffusion will be the molecular one $K = 0{\cdot}2$; then micro-turbulence will spread it less slowly ; then, after a few seconds, part may get caught in one of the gusts such as are shown by a pressure-tube anemometer, while another part may remain in a lull, so that it is torn asunder and gusts scatter it, K being 10^4. Next squalls of several minutes' duration separate it more rapidly. Its rate of diffusion is now measured by $K = 10^8$. Then one part gets into a cyclone and another remains behind in an anti-cyclone, and its rate of diffusion is measured by Defant's value $K = 10^{11}$. Finally, it is fairly uniformly spread throughout the earth's atmosphere at the rate of about one molecule of acetylene for every cube of surface air 70 metres in the edge.

This diffusing dot is in a sense a natural standard. In the theory of the diffusion of heat (see, for example, E. W. Hobson,' Encyk. Math. Wiss.,' vol. 4, p. 187) something rather like this is found to be useful. A small dot of heat is imagined to spread out as time proceeds into an unbounded medium. This distribution of heat in space-time is taken as an element, like the point-charge in electrostatics, from which more complicated distributions can be built up. Can we do likewise for diffusion in the atmosphere ? Imagine at an instant a gradient of concentration of acetylene in the atmosphere over an area measuring 100 km. × 100 km. Let each cube of 10^{-2} cm. edge begin separately to spread

out in the manner described above, and let us superpose the separate spreadings in the hope of finding the flux of mass down the gradient of concentration. To do this we would naturally consider the change in a *short* time Δt of, say, 1/100 second. But in this short time each dot spreads with molecular diffusivity 0·2 cm.^{2} sec.$^{-1}$. So any results deduced from the superposition of the effects of the dots will correspond to molecular diffusivity and will ignore the effects of eddies. This picture is false to Nature. So we must conclude that *in the atmosphere a spreading dot will not serve as an element from which general distributions can be built up.*

§ 2. A Type of Mean which avoids the foregoing Difficulties and which might with Advantage be Observed.

The fundamental idea of this paper is that the rate of diffusion increases with the distance apart. To state this carefully let us revert to the two molecules of acetylene, let loose at $t = 0$ at the points (a_1, b_1, c_1), (a_2, b_2, c_2). At time t let their positions be (x_1, y_1, z_1), (x_2, y_2, z_2). The x component of their separation is initially $a_2 - a_1$ and becomes $x_2 - x_1$ at time t. Now let the release of a pair of particles at the same points be repeated many times in succession, and let [ ] denote a mean taken over these successive pairs. Consider the mean square of the deviation of $(x_1 - x_2)$ from its mean at time t, that is,

$$[(x_1 - x_2 - [x_1 - x_2])^2] = [j] \text{ say.}$$

Suppose, for example, that $[x_1 - x_2]$ is a kilometre. Then gusts, which may be seen on a lake or on a cornfield as patches of ruffled surface a fraction of a kilometre long, would affect individual members of the pair x_1, x_2 separately and so would tend to increase $[j]$. We should get the average effect of such gusts if we prolonged the time of averaging indefinitely. The time of averaging must not be confused with t, the time of flight. It is an advantage to have a *pair* of marked molecules. For if instead we considered molecules released one at a time, then their mean square deviation from their mean position at t, namely, $[(x - [x])^2]$, would depend on larger and larger eddies as the time included in the average [ ] was increased, so that no limit to the average would be attained until cyclonic changes were included. But when molecules are released in pairs this is not so. For if a cyclone passes over the district so that the wind changes its direction through one or two right angles, these changes will occur nearly simultaneously at the two stations one kilometre apart, so that they will not have much effect on individual values of $(x_1 - x_2)$ nor on $[j]$. We have at last found in $[j]$ a mean which attains a limit, as the time of averaging is prolonged

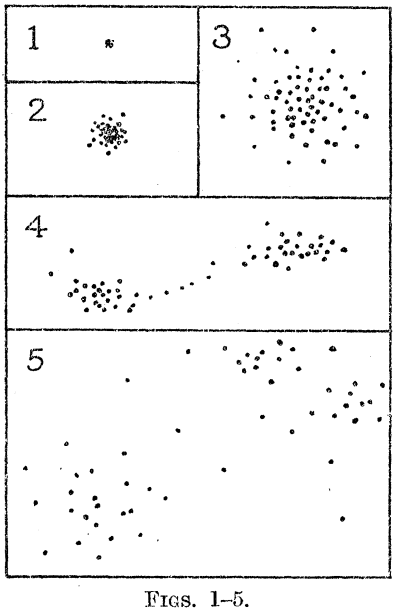
indefinitely, and yet only brings in the effects of eddies comparable in diameter with $[x_1 - x_2]$ or less.

Direct observations of $[j]$ have not been made so far as I know, but there is a mass of published evidence about turbulence which strongly suggests that $[j]/t$ is independent of t, if t is neither too small nor too large, and that $[j]/t$ increases with the distance between the starting points.

§ 3. A Statistic for Clusters.

§ 3.1. *Introduction.*

The failure of the dispersal of a point-charge to serve as a mathematical element, from which the dispersal of an extended system may be built up, appears to be intimately connected with the fact that in the atmosphere the dispersal goes on in patches. That is to say, a small dense cluster of marked molecules, represented by the dot in fig. 1 which, by molecular diffusion alone, would spread through the successive spherical clusters shown in figs. 2 and 3, actually seldom passes through the large spherical stage 3, because it is first sheared into two detached clusters as suggested in fig. 4. These are carried far from one another, and are likely to be again torn into smaller pieces as in fig. 5. Meanwhile each of the torn parts is gradually spreading by molecular diffusion. These diagrams are, of course, merely illustrative fictions.



Figs. 1–5.

As a preliminary to describing the succession of changes, we must find out how to describe the distribution at a single instant. The problem is rather like that of finding some simple specification of the extent to which the population is divided between cities, towns, villages and isolated houses, without making a map; for in the atmosphere a map of all the eddies would be too bewilderingly complicated. We want something that gives us a general measure of the spread of the molecules, thus serving the purpose of the standard deviation of the whole aggregate of marked molecules from its mean, and yet at the same time informs us about the internal details of the cluster. Both purposes will be served, as will be shown, by the following method, which is not intended as a practical observation, but as a mathematical specification.

§ 3.2. *Definition of the Statistic* Q, *the Mean Number of Neighbours per Length.*

The air is supposed to contain a large number of "marked molecules." They might, for example, be acetylene. For simplicity, let us confine attention to distribution of points on a straight line. Take any marked molecule, which for reference we will call A. With A as origin, divide the line by sections at positive and negative integral multiples of a unit h, thus forming "cells" each of length h. Count the number of marked molecules in each of these cells. When a molecule is exactly on the partition between a pair of cells, half of it is attributed to each cell. Let $h \,.\, A_{n,\, n+1}$ denote the number in the cell between $l = nh$ and $l = (n + 1)\, h$ where l is the distance from A measured in the positive sense, and n is an integer. Let there be N marked molecules altogether. A molecule might conceivably be considered to be its own neighbour at zero distance, but we do not make this convention, and therefore the sum of the numbers in the cells is $N - 1$. Next repeat the performance with the origin at each one of the other marked molecules B, C, D, in turn. Then form the mean

$$Q_{n,\, n+1} = \frac{1}{N} (A_{n,\, n+1} + B_{n,\, n+1} + C_{n,\, n+1} + \text{etc., to N terms}).$$

In this way we obtain a set of quantities $Q_{0,\, h}$, $Q_{h,\, 2h}$, $Q_{2h,\, 3h} \ldots$, which are numbers of marked molecules per length, classified according to their distances l from other molecules; these distances lying in the ranges 0 to h, h to $2h$, $2h$ to $3h$, and so on. Next, it will be well to draw a diagram in which the ordinate is $Q_{n,\, n+1}$ for the range of abscissæ extending from $l = nh$ to $l = (n + 1)\, h$. We may now drop the suffixes and regard Q as a function of l. This (l, Q) diagram has many interesting properties.

To illustrate the definition the lower part of fig. 6 was obtained by making the prescribed counts on the linear cluster of 7 molecules marked above. The

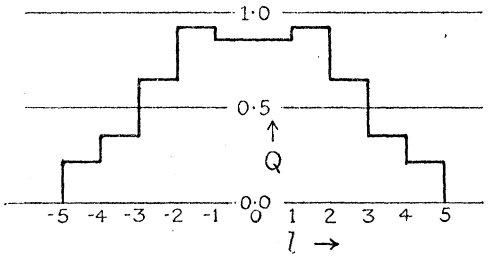


FIG. 6.

successive distances between the molecules are, in tenths, 2, 4, 6, 8, 10, 12, and h is unity.

§ 3.3. *Note on the Step h.*

This element of length should be chosen so that in the average $Q_{n,\,n+1}$ the element shall contain a considerable number, say, at least 100, marked molecules over the values of l where they are most crowded. Otherwise, random errors of sampling might become apparent. Even if this is done for most of the diagram, there may be other ranges of l where marked molecules are so scarce that sampling errors might become noticeable. On the other hand, if h were made too large the steps in the diagram might become too wide; whereas we want the stairs to look like a curve. These compromises are perfectly familiar in statistical work, and are inevitable. Although mentioned here for completeness, they are really of no importance, as we may easily have a billion molecules in the cluster. So that in future we shall replace the stairs by a curve drawn through the centre of each step. In other words, the mean number of neighbours per length, like the density, or the concentration, attains a " quasi-limit " when the element of space has a magnitude lying in a certain range.

§ 3.4. *The* (l, Q) *Diagram is Symmetrical about the* Q *Axis.*

For the distance between every pair of molecules is counted twice, as negative from one end, as positive from the other. Therefore, if Q be expanded in powers of l only even powers can occur.

§ 3.5. *As Diffusion proceeds, the Area enclosed between the l Axis and the (l, Q) Curve remains constant.*

For this area when expressed in units of l and Q is simply one less than the whole number N of marked molecules. This is easily proved from the definition of Q. And by hypothesis N remains constant.

This property of the graph suggests that Q must satisfy a differential equation of the type

$$\frac{\partial Q}{\partial t} = \frac{\partial}{\partial l} \left\{ \begin{array}{l} \text{some function of } l \text{ which attains} \\ \text{a limit as } l \to \infty \text{ equal to that} \\ \text{attained as } l \to -\infty \end{array} \right\},$$

for, if so

$$\int_{-\infty}^{\infty} \frac{\partial Q}{\partial t} dl = 0.$$

§ 3.6. *The (l, Q) Diagram exhibits the Size of a Linear Cluster.*

For if the distance between the extreme molecules at the opposite ends of the cluster be L, then Q is zero for all values of $|l|$ greater than $|L|$; and Q is finite when $l = \pm L$. Thus, the extreme width of the l, Q curve is twice the extreme diameter of the cluster. The relation between the standard deviations will be discussed in § 6.8.

§ 3.7. *The Changing Form of the (l, Q) Diagram as Diffusion proceeds.*

It is evident from the foregoing that if there is only one linear cluster, and it spreads along its line, the (l, Q) graph must spread along the l axis. And as the area under it must be constant, its mean height in the Q-direction must decrease.

Let us consider another very simple case (analogous to the melting of a crystal).* Suppose that initially-marked molecules are equally spaced at intervals of one centimetre all along the line without bound in either direction. What will the (l, Q) graph look like? No molecule will have a neighbour nearer than a centimetre, so Q is zero for $0 < l < 1$.

At $l = 1$ cm. neighbours are indefinitely common and Q is infinite. Again, there are no neighbours in the range $1 < l < 2$, and so on. The graph consists of a series of infinities of Q at $l = \pm$ (1, 2, 3, 4, ... cm.) with $Q = 0$ everywhere else.

Now suppose that diffusion takes place. Can we find the changes from Fick's equation? Not from it alone, for Fick's equation is a statement about the gradients of a continuous function of position, whereas we have only particles

* Note added December 7.

widely separated. If Fick's equation is to be applied to this special example, it would have to be by the aid of an additional hypothesis derived from the theory of probability. Instead, I get the following from memories of snow-flakes falling, of the Brownian motion under a microscope, or of foam circulating on a millpond. With these in mind, it is evident that the perfect regularity of the arrangement of marked molecules will soon be a little disturbed. Molecules will acquire neighbours a little nearer and a little farther away than the exact 1, 2, 3, 4, ... cm., and neighbours at these distances will no longer be infinitely common. That is to say, the infinities of Q will be softened down into peaks with spreading bases. In an early stage the bases will not join; there will still be no neighbours at distances, such as $\frac{1}{2}$, $1\frac{1}{2}$, $2\frac{1}{2}$, ... cms. In this stage the area enclosed between each peak and the l axis must remain constant. The form of the intermediate curve shown in fig. 7 is intended merely to suggest that flattening proceeds more rapidly as l is greater; otherwise, the curve is a guess. Later (§ 5.4, § 6.7) some other cases will be discussed quantitatively.

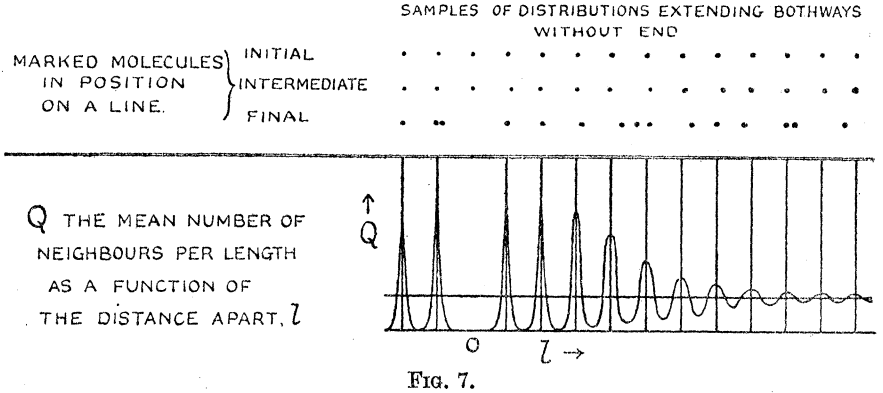


FIG. 7.

§ 3.8. *The Final State after Thorough Diffusion.*

From the foregoing we should expect the final state to be represented by a straight line parallel to the l axis. This expectation is confirmed by considerations of probability. For there seems no reason why the number Q of marked molecules per length should have any dependence upon the distance to any molecule, when the distribution is purely random.

§ 3.9. *A Failure of Concentration as a Descriptive Idea.*

In the preceding special example, if we are to speak of concentration at all, we must take a long element of length, say 1,000 cms., in order to have a good

many marked molecules in it. Then all that can be said about the concentration is that it was initially independent of position and remained so always. According to this view nothing happened. How different from the lively process shown on the (l, Q) diagram!

§4. Fick's Equation and the Distance-Neighbour Diagram.

§4.1. *Introduction.*

The new theory which is intended to apply to both eddy- and molecular diffusion ought to be consistent with Fick's equation in the special case of no eddies. Let us now explore this connection.

We have seen that the idea of the "concentration" of marked molecules, given as a function of position, has in one instance signally failed to describe that which we wish to discuss. Fick's equation, being based on the idea of concentration as a function of position, has failed there also. Elsewhere we shall find both very useful.

§4.2. *Continuous Concentration.*

In order to bring Fick's equation into our theory we must suppose that the concentration ν, defined to be the number of marked molecules per length, is a continuous function of x possessing derivatives $\partial\nu/\partial x$, $\partial^2\nu/\partial x^2$. This supposition is a little artificial. But it seems likely that $\Delta\nu/\Delta x$ really attains with sufficient accuracy a quasi-limit when Δx is neither too large nor too small. Fick's equation is then

$$\frac{\partial \nu}{\partial t} = \mathrm{K}\frac{\partial^2 \nu}{\partial x^2}.$$

Next Q must be redefined in terms of ν. The definition in terms of continuous concentration can be made to agree with that in terms of particles, except as regards neighbours as close as or closer than the closest pairs of particles. As a reminder of this, often unimportant, discrepancy, the new function will be denoted by small q. For instance, if each of the particles in fig. 7 were replaced by a small dot of continuous substance, then q would have an infinity at $l = 0$ where Q is zero.

§4.3. *The Definition of q applicable when the Diffusing Substance is Continuous, not Molecular.*

We take any point x on the line at which the concentration is ν a function of x and time only. It will sometimes be written $\nu\,(x, t)$. We proceed from x a further distance l to $x + l$. Let the concentration at $x + l$ be ν_1. Here

ν_1, unlike ν, must be regarded as a function of three independent variables x, l and t, and may accordingly be written $\nu_1(x, l, t)$. The analogue of $A_{n,\ n+1}$ in the previous definition is here simply ν_1 regarded as a function of l while x is fixed. Then, because in taking the mean, each particle comes in once as origin, we must here form a weighted mean, the weight being ν. We thus arrive at

$$q = \underset{\substack{\lambda \to \infty \\ \theta \to \infty}}{\text{Limit}} \frac{\int_{-\theta}^{\lambda} \nu(x, t) \cdot \nu_1(x, l, t)\, dx}{\int_{-\theta}^{\lambda} \nu(x, t)\, dx} \cdot \tag{1}$$

This leaves q a function of l and t only.

If there is a limit there can only be one, so that to a given function $\nu(x)$ only one function $q(l)$ as defined in this way can correspond. As we shall see later, the converse is not true.

It generally happens that the integrals in the numerator and denominator attain their limits separately, and when this occurs we can write the definition in the simpler form

$$q = \frac{1}{\mathrm{N}} \int_{-\infty}^{+\infty} \nu(x, t) \cdot \nu_1(x, l, t)\, dx. \tag{2}$$

where

$$\mathrm{N} = \int_{-\infty}^{+\infty} \nu\, dx, \tag{3}$$

so that N is the whole number of marked particles as in the definition of Q.

§4.4. *Correspondence of Areas on the* (x, ν) *and* (l, q) *Graphs when the Areas are Finite.*

$$\int_{-\infty}^{+\infty} q\, dl = \frac{1}{\mathrm{N}} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \nu(x),\ \nu_1(x, l)\, dx\, dl.$$

Since the termini are independent of one another, changing the sequence of integrations makes no difference to the result. Integrate first with respect to l. Then as the range is infinite, the inner integral transforms thus :—

$$\int_{-\infty}^{+\infty} \nu_1\, dl = \int_{-\infty}^{+\infty} \nu_1\, dx = \int_{-\infty}^{+\infty} \nu\, dx = \mathrm{N}.$$

Therefore, on inserting this value of the inner integral

$$\int_{-\infty}^{+\infty} q\, dl = \frac{\mathrm{N}}{\mathrm{N}} \int_{-\infty}^{+\infty} \nu\, dx = \mathrm{N}.$$

So

$$\int_{-\infty}^{+\infty} q\, dl = \int_{-\infty}^{+\infty} \nu\, dx,$$

a result which we shall often require.

§ 4.5. *The Differential Equation for the* (l, q) *Graph when the Diffusion is Fickian.*

It will now be proved that—

When there is no mean motion or eddies then q, the mean number of neighbours per length, is related to the time t and the separation l by a differential equation like Fick's, which relates the concentration ν to the time t and the position x; but for q the diffusivity is double that for ν. It is assumed that $\nu d\nu/dx$ vanishes at infinity.

If $\Pi = \nu\nu_1$, then because $\partial\nu_1/\partial x = \partial\nu_1/\partial l$ and $\partial^2\nu_1/\partial x^2 = \partial^2\nu_1/\partial l^2$, it may be proved without any assumption about vanishing at infinity, or about Fick's equation, that

$$\frac{\partial^2\Pi}{\partial x^2} - 2\frac{\partial^2\Pi}{\partial x\,\partial l} + 2\frac{\partial^2\Pi}{\partial l^2} = \nu\frac{\partial^2\nu_1}{\partial x^2} + \nu_1\frac{\partial^2\nu}{\partial x^2}. \tag{1}$$

In view of Fick's equation, namely,

$$\frac{\partial\nu}{\partial t} = \mathrm{K}\frac{\partial^2\nu}{\partial x^2} \quad \text{and} \quad \frac{\partial^2\nu_1}{\partial t} = \mathrm{K}\frac{\partial^2\nu_1}{\partial x^2}, \tag{2}$$

the second member of (1) transforms, into

$$\frac{1}{\mathrm{K}}\left(\nu\frac{\partial\nu_1}{\partial t} + \nu_1\frac{\partial\nu}{\partial t}\right) = \frac{1}{\mathrm{K}}\frac{\partial\Pi}{\partial t}. \tag{3}$$

Thus ν, ν_1 no longer appear separately, and their product Π satisfies the linear equation

$$\frac{\partial\Pi}{\partial t} = \mathrm{K}\left\{\frac{\partial^2\Pi}{\partial x^2} - 2\frac{\partial^2\Pi}{\partial x\,\partial l} + 2\frac{\partial^2\Pi}{\partial l^2}\right\}. \tag{4}$$

Now

$$\int_{-\infty}^{\infty}\Pi\,.\,dx = \mathrm{N}q, \tag{5}$$

and on integrating the Π-equation with the proviso that $\partial\Pi/\partial x$ and $\partial\Pi/\partial l$ both vanish as $x \to +\infty$ and $-\infty$ we obtain

$$\frac{\partial q}{\partial t} = 2\mathrm{K}\frac{\partial^2 q}{\partial l^2}. \tag{6}$$

Compare this with (2) and the theorem is proved.

* § 4.6. *Theorem: The Effect of a Mean Motion independent of* x *disappears when the number of Neighbours per Length is related to Separation and Time.*

For, by the definition, q is independent of the choice of the origin of x, and so, if the mean velocity $\bar{u}$ is independent of x, we can get rid of $\bar{u}$ by giving a

* § 4.6 was added, and the corresponding alterations were made, in § 5.1, § 5.2, § 8, on December 7.

suitable velocity to the origin of x. This can be done even if $\bar{u}$ is an arbitrary function of time.

For simplicity in § 4.5 it was assumed that there was no mean motion. Actually when K has been derived from observations of smoke or of balloons, the mean velocity has customarily been taken into account by using as the definition of K some equation more or less equivalent to §1.1 (1), in which $\bar{u}$, $\bar{v}$, $\bar{w}$ appear.

We see now that

$$\frac{\partial \nu}{\partial t} + \bar{u}\frac{\partial \nu}{\partial x} = \mathrm{K}\frac{\partial^2 \nu}{\partial x^2} \tag{1}$$

leads to

$$\frac{\partial q}{\partial t} = 2\mathrm{K}\frac{\partial^2 q}{\partial l^2}, \tag{2}$$

in which $\bar{u}$ does not appear.

Incidentally it is of interest to take the form (1) in place of § 4.5 (2) in forming the Π-equation. We thus obtain

$$\frac{\partial \Pi}{\partial t} + \bar{u}\frac{\partial \Pi}{\partial x} = \mathrm{K}\left\{\frac{\partial^2 \Pi}{\partial x^2} - 2\frac{\partial^2 \Pi}{\partial x\,\partial l} + 2\frac{\partial^2 \Pi}{\partial l^2}\right\}, \tag{3}$$

and, on integration with respect to x, the term in $\bar{u}$ vanishes if Π has the same value for all indefinitely great values of $|x|$. Thus we arrive again at equation (2) of the present section.

§ 4.7. * *The Present Theory, written for Diffusion on a Straight Line, is applicable also to the Projection of Three-dimensional Diffusion on to this Straight Line.*

For let ρ be the number of marked molecules per volume, and let them be diffusing according to

$$\frac{\partial \rho}{\partial l} = \frac{\partial}{\partial x}\left(\mathrm{K}\frac{\partial \rho}{\partial x}\right) + \frac{\partial}{\partial y}\left(\mathrm{K}\frac{\partial \rho}{\partial y}\right) + \frac{\partial}{\partial z}\left(\mathrm{K}\frac{\partial \rho}{\partial z}\right). \tag{1}$$

Now project each molecule normally on to the x-axis, and let ν' be the number of projections per length. Then

$$\nu' = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} \rho\, dy\, dz. \tag{2}$$

Let us assume that ρ vanishes at infinity in such a way as to make ν' finite. On integrating (1) with respect to y and z so as to produce an equation in ν', the term $\frac{\partial}{\partial y}\left(\mathrm{K}\frac{\partial \rho}{\partial y}\right)$ yields

$$\int_{-\infty}^{+\infty}\left[\mathrm{K}\frac{\partial \rho}{\partial y}\right]_{y=-\infty}^{y=+\infty} dz,$$

* § 4.7 and consequent explanations elsewhere were added on December 7.

and we may usually safely assume that the integrand vanishes. The term $\frac{\partial}{\partial z}\left(\mathrm{K}\frac{\partial \rho}{\partial z}\right)$ behaves similarly. Thus there results

$$\frac{\partial \nu'}{\partial z} = \mathrm{K}\frac{\partial^2 \nu'}{\partial x^2}. \tag{3}$$

This is the interpretation that must always be given to the present theory before it can be applied to observations.

§ 5. Non-Fickian Diffusion.

§ 5.1. *Generalisation for Atmospheric Eddies.*

When the diffusion is molecular both equations § 4.6 (1) and § 4.6 (2) correctly describe it. When the eddies of the free atmosphere come into action neither of these two equations describe the phenomena correctly; but whereas in Fick's equation the defect appears to be incurable, it is very easily remedied in the new equation now presented. That is to say, the chief advantage of the variables l and q, to which all the foregoing is merely preparatory, appears when we consider the effects of eddies. For, as already stated, observation shows that the rate of diffusion increases with the separation l of neighbours. We can represent this by writing

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial l}\left(\mathrm{F}(l) \cdot \frac{\partial q}{\partial l}\right), \tag{1}$$

where $\mathrm{F}(l)$ is an increasing function of l. In passing it should be noted that it would not do to write the second member $\mathrm{F}(l) \cdot \partial^2 q/\partial l^2$, for then $\int_{-\infty}^{+\infty} \frac{\partial q}{\partial t}\, dl$ would not necessarily vanish and the total number of particles would not be fixed.

If we were to modify Fick's equation by writing

$$\frac{\partial \nu}{\partial t} + \bar{u}\frac{\partial \nu}{\partial x} = \frac{\partial}{\partial x}\left(f(x) \cdot \frac{\partial \nu}{\partial x}\right)$$

that would mean that the diffusivity depended on position, an effect altogether different from the one represented by $\mathrm{F}(l)$, and one which will not be studied in this paper. Instead, the paper discusses an atmosphere in which the diffusivity is independent of position, but depends on separation.

§ 5.2. *Reduction of Existing Observations.*

Let us next consider some observations which show how $\mathrm{F}(l)$ depends on l. The observations have been made for other purposes, and are not quite what

is desirable here. The quantity usually measured has been K in the equation § 1.1 (1) or something equivalent.

In accordance with § 4.5 (6), F (l), if it were merely a constant, would be equal to 2K. Actually, many values of l co-operate in the diffusion, but those which are largest not only produce most diffusion, but also have most weight in determining K, when K is calculated from the mean *squared* deviation from the mean. Thus, it seems likely that we shall get the right order of magnitude both for F (l) and l, if we put F (l) equal to 2K and l equal to the standard deviation of the particles from their mean. As l varies in the ratio $1 : 10^9$, even very crude estimates of F (l) show its relation to l quite clearly.

When K has been obtained from the variation of wind with height, 2K is still assumed to be roughly equal to F (l) and the corresponding l is taken to be the mean vertical separation of the anemometers that were used in finding the second derivatives of the wind-components with respect to height. When the observations were obtained from pilot balloons, we know that l cannot be less than the vertical displacement of a balloon between two sightings, so that l will probably not be less than 100 metres. On the other side l cannot be greater than the height of the observation above the ground. The mean of these two distances has been taken to be l.

The distance at which molecular motion is the chief cause of diffusion in free air may be roughly estimated in the following way:—Suppose that a very thin lamina of marked molecules could be produced in still air. The concentration should be arranged to be greatest in the central sheet and to diminish towards the outer sheets according to the law of error. The thickness of the lamina, as measured by a standard deviation from the mean, would then increase so that*

(standard deviation) $= \sqrt{(2Kt)}$ where t is the time from indefinite thinness.

Now, the value of K due to molecular diffusion is about 0·17 cm.^{2} sec.$^{-1}$. Hence, we have the following:—

time in seconds	0	0·001	0·01	0·1	1
thickness as measured by standard deviation, cms.	0	0·018	0·058	0·18	0·58

These numbers show clearly that molecular diffusion is very effective when the lamina is 0·01 cm. thick and much less so when it is 0·1 cm. thick. Now, if we look at cigarette smoke in the open air and ask ourselves at what separation molecular motion will produce rather more effect than the eddies, it is not difficult to make a guess. I put it at $l = 5 \times 10^{-2}$ cm. The integral power of ten is really all that matters.

* 'Phil. Trans.,' A, vol. 221, p. 6 (1920).

The data are summarised in the following table :—

Reference.	K cm.^{2} sec^{-1}	l cm.
K from molecular diffusion of oxygen into nitrogen (Kaye and Laby's 'Physical and Chemical Constants'). For l see preceding discussion.	$1{\cdot}7 \times 10^{-1}$	5×10^{-2}
K at 9 metres above ground from anemometers at heights of 2, 16 and 32 metres (W. Schmidt, 'Wien. Akad. Sitzb.,' IIa, vol. 126, p. 773 (1917)).	$3{\cdot}2 \times 10^{3}$	$1{\cdot}5 \times 10^{3}$
K from anemometers at heights of 21 to 305 metres (Åkerblom, F., 'Nova Acta Reg. Soc. Upsaliensis' (1908)).	$1{\cdot}2 \times 10^{5}$	$1{\cdot}4 \times 10^{4}$
K from pilot balloons at heights between 100 and 800 metres (Taylor, 'Phil. Trans.,' A, vol. 215, p. 21 (1914), also Hesselberg and Sverdrup, 'Leipzig Geophys. Inst.,' Ser. 2, Heft 10 (1915)).	6×10^{4}	5×10^{4}
K from tracks of balloons either manned (L. F. Richardson, 'Weather Prediction by Numerical Process,' p. 221) or not manned (Richardson & Proctor, 'Royal Meteorological Society Memoirs,' No. 1).	10^{8}	2×10^{6}
Volcano ash, same reference as last	5×10^{8}	5×10^{6}
Diffusion due to cyclones regarded as deviations from the mean circulation of the latitude (Defant, 'Geog. Ann.,' H. 3, also (1921), 'Wien. Akad. Wiss. Sitzb.,' IIa, vol. 130, p. 401 (1921)).	10^{11}	10^{8}

Since, when not obstructed by the ground, smoke spreads about as much horizontally as it does vertically,* the observations at the smaller values of l, though made in the vertical, can be treated as applicable to the horizontal. Thus the whole collection is coherent.

The logarithms of K and l when plotted on a graph (fig. 8) are seen to lie close to a line of slight curvature. It is hardly worth while to discuss details until observations have been made in a manner appropriate for the determination of F (l) rather than of K. How such observations could be obtained will be discussed in § 7.

The straight line on the logarithmic diagram which corresponds to $K = 0{\cdot}2\ l^{4/3}$ also fits the observations almost as well as the curve in the limited range between l = a metre and l = 10 kilometres. For mathematical simplicity this formula will be used in the illustrations which follow.

Thus in this range $F(l) = 0{\cdot}4\ l^{4/3}$ approximately, when the units are centimetres and seconds.

* G. I. Taylor.

The equation for the changes in the (l, q) graph is then

$$\frac{\partial q}{\partial t} = \varepsilon \frac{\partial}{\partial l}\left(l^{4/3}\frac{\partial q}{\partial l}\right), \tag{1}$$

where the constant ε is of the order of $0{\cdot}4$ cm.$^{2/3}$ sec^{-1}. This equation summarises the subject.

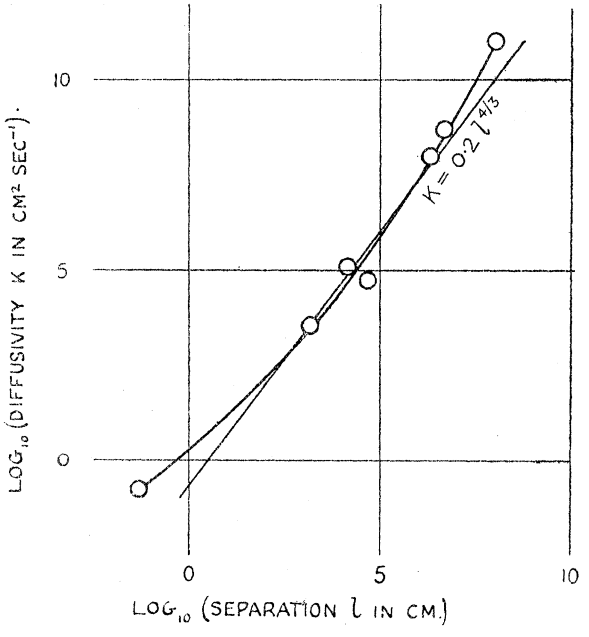


FIG. 8.

§ 5.3. *Analogy with the Diffusion of Heat.*

The fundamental equation § 5.2 (1) can be brought into touch with some standard mathematical forms by changing the variable l to $l^{1/3} = \alpha$ say. For this transforms the equation into

$$\frac{\partial q}{\partial t} = \frac{\varepsilon}{9}\left\{\frac{2}{\alpha}\frac{\partial q}{\partial \alpha} + \frac{\partial^2 q}{\partial \alpha^2}\right\}, \tag{1}$$

which is Fourier's equation for the diffusion of heat in a homogeneous solid, where the isothermal surfaces are concentric spheres of radius α, and the diffusivity is $\varepsilon/9$. The equivalence of the equations in α and l is complete, except at $l = 0$, where a source of sink might occur. In choosing a solution we must be sure whether it makes the whole number of marked molecules independent of time.

§ 5.4. *Non-Fickian Diffusion of an Initial Point-Cluster on a Line.*

A solution due to Fourier,*

$$q = \mathrm{A}\,(4t\varepsilon/9)^{-3/2} \,.\, e^{-\frac{\alpha^2}{4t\varepsilon/9}}, \tag{1}$$

in which A is independent of t and α, represents a process in which at $t = 0$ all neighbours are indefinitely close, and as time proceeds they spread out continually. The corresponding value of N is

$$\mathrm{N} = \int_{-\infty}^{+\infty} q\, dl = \mathrm{A}\,(4t\varepsilon/9)^{-3/2} \int_{\alpha^2 \to -\infty}^{\alpha^2 \to \infty} e^{-\frac{\alpha^2}{4t\varepsilon/9}} \,.\, 3\alpha^2 \,.\, d\alpha.$$

Putting

$$\beta^2 = \frac{\alpha^2}{4t\varepsilon/9}$$

it is found that

$$\mathrm{N} = 3\mathrm{A} \int_{-\infty}^{+\infty} e^{-\beta^2} \beta^2\, d\beta. \tag{2}$$

Thus N is independent of time, as required, and there is no source at $l = 0$, except at $t = 0$.

Fig. 9 exhibits this function in the special form

$$q = 10^5 \,.\, t^{-3/2} \,.\, e^{-\frac{l^2/3}{4t\varepsilon/9}} \tag{3}$$

when ε is given its observed value of 0·4 C.G.S. units.

At $t = 0$ the graph would consist of an infinity of q at $l = 0$, and $q = 0$ elsewhere. One graph shows the distribution at $t = 100$ seconds. Neighbours as distant as 4 metres are now not scarce. Five minutes later, at $t = 400$ seconds, neighbours at 40 metres are noticeable. In the corresponding distribution in space the isopleths of concentration are parallel planes. The standard deviation of the marked molecules from their mean position will be investigated in § 7.1.

§ 6. Returning from Number of Neighbours as a Function of Separation towards Concentration as a Function of Position.

§ 6.1. *Introduction.*

In the course of the theory we began with concentration ν given as a function of position x. We wished to know what became of the distribution when

* See E. W. Hobson, 'Wärmeleitung, Encyk. Math. Wiss.,' vol. 4, p. 195.

diffusion occurred. Fick's equation being contrary to the facts, and no suitable adjustment of it being in sight, we had to change the variables from (x, ν) to separation l, and mean number of neighbours per length q. This was easily done. Then the equation $\frac{\partial q}{\partial t} = \varepsilon \frac{\partial}{\partial l}\left(l^{4/3} \frac{\partial q}{\partial l}\right)$ gave the changes in the (l, q) graph. But, after all, it is ν as a function of x that we should like to know; so that, after the diffusion has occurred, we wish, if possible, to change the variables (l, q) back again to (x, ν). This is not so easy, and can only be done in part.

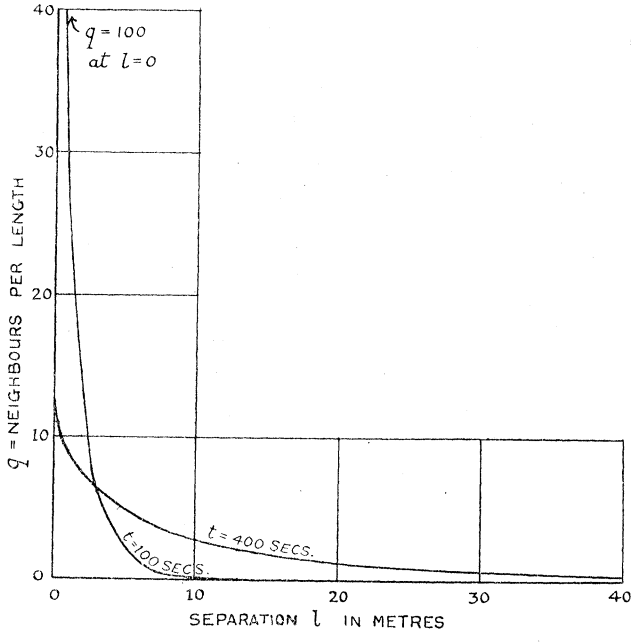


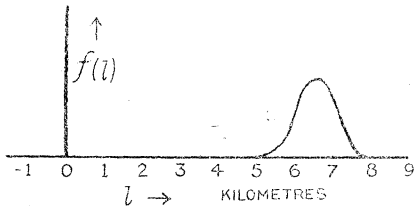
FIG. 9.

As with the problem of integration, a general method is lacking, and so it seems desirable to give typical examples and a variety of processes suited to different circumstances.

It would be too much to expect that the (l, q) graph should give us enough information to allow the distribution of particles in space to be reconstructed in all its details. For the process of taking a mean has been used in forming q to get rid of a superabundance of detail. The process is irreversible. We cannot evolve the detail again from the mean.

The origin of x is not represented by anything in the (l, q) graph.

§ 6.2. *Not every Even Function $f(l)$ which is Positive for all Values of l can serve for the Number of Neighbours per Length.*



Try, for instance, to imagine how the population could be distributed on a line so as to produce a (l, Q) diagram like fig. 10. If you have neighbours at many distances lying between 5 and 8 km., some of them must have neighbours at distances less than 3 km., so the diagram would have to have a central hump. As drawn it cannot be an (l, Q) diagram.

However, when, as usual, q_B has been produced by diffusion from q_A, and q_A corresponded to ν_A, then if the mathematics fits the physics, as it appears to do, there must exist a ν_B corresponding to q_B.

§ 6.3. *The Central Value of q.*

This is evidently $q(0) = \frac{1}{N}\int_{-\infty}^{+\infty} \nu^2\, dx$ when ν vanishes at infinity.

§ 6.4. *Uniformity.*

Given that $\nu = b$ a quantity independent of x.

Therefore $\nu_1(x, l) = b$ also.

And by the definition of § 4.3

$$q = \underset{\substack{\theta \to \infty \\ \lambda \to \infty}}{\text{Limit}} \frac{b^2 \int_{-\theta}^{\lambda} dx}{b \int_{-\theta}^{\lambda} dx} = b.$$

So q is independent of l.

As to the converse, see under Fourier series.

§ 6.5. *A Single Linear "Town."*

Given that

$$\nu = \frac{1}{\sqrt{(2\pi)}}\, e^{-\frac{1}{2}x^2}.$$

Then

$$N = \int_{-\infty}^{\infty} \nu\, dx = 1.$$

Also

$$\nu\nu_1 = \frac{1}{2\pi} e^{-\frac{1}{2}\{(x+l)^2 + x^2\}} = \frac{1}{2\pi} e^{-\frac{1}{2}(\sqrt{2}x + (l/\sqrt{2}))^2} \cdot e^{-\frac{1}{4}(l^2/2)}.$$

Therefore

$$\int_{-\infty}^{\infty} \nu\nu_1\, dx = \frac{1}{\sqrt{(2\pi)}} e^{-(l^2/4)} \frac{1}{\sqrt{2}\,.\,\sqrt{(2\pi)}} \int_{-\infty}^{+\infty} e^{-\frac{1}{2}(\sqrt{2}x + l/\sqrt{2})^2}\, d\left(\sqrt{2}x + \frac{l}{\sqrt{2}}\right)$$

$$= \frac{1}{2\sqrt{\pi}} e^{-(l^2/4)} = q.$$

See also a note at the end of § 6.7.

§ 6.6. *Expansion in a Fourier Series.*

There may be distributions of concentration, extending indefinitely in both directions, for treating which Fourier series will be suitable.
Let

$$\nu(x) = \tfrac{1}{2}A_0 + \sum_{n=1}^{n\to\infty} \{A_n \cos nx + B_n \sin nx\}, \qquad (1)$$

n being a positive integer and the constant A_0 being so chosen as to make ν everywhere positive. Then it may be shown that

$$q = \operatorname*{Limit}_{\substack{\lambda\to\infty \\ \theta\to\infty}} \frac{\tfrac{1}{2}(\theta+\lambda)\left\{\frac{A_0^2}{2} + \Sigma(A_n^2 + B_n^2)\,.\,\cos nl\right\} + \text{finite terms}}{\tfrac{1}{2}A_0(\theta+\lambda) + \text{finite terms}}$$

$$= \tfrac{1}{2}A_0 + \frac{1}{A_0}\sum_{n=1}^{n\to\infty}(A_n^2 + B_n^2)\cos nl. \qquad (3)$$

The fundamental wave-length in q and ν has been taken as 2π. Any other value could be introduced by changing the units of x and l in the same ratio, and the relations between A_n, B_n and C_n would remain as stated.

In particular if $q = \frac{1}{2}C_0$ simply, then $\nu = q$ is a solution, and on account of the generality of the Fourier series it appears to be the only possible one.

§ 6.7. *The Correspondence of Diffusion from Points on the x and l Axes.*

Time is not involved in the connection between the position-concentration graph and the distance-neighbour graph ; but we can bring in time as an aid to finding the connection. Suppose, for example, that the diffusing substance is initially concentrated in five masses each consisting of n particles near five equidistant points on the x-line at intervals b of x. Then there are neighbours near $l = 0, \pm b, \pm 2b, \pm 3b, \pm 4b$, but for no other values of l. The relative numbers of neighbours in these five classes are easily counted.

Next, suppose that Fickian diffusion occurs. Both graphs change, but at

the same instant they must always correspond to one another. Fig. 11 shows the two graphs at one instant.

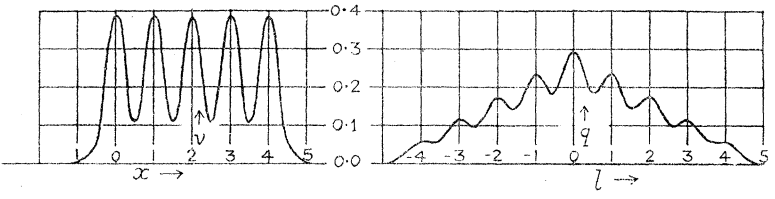


FIG. 11.

Now, a given distribution of ν in x may be seen from the definition of q to lead to a unique function q of l. Thus, $q\ (l)$ does not depend on how $\nu\,(x)$ came to be, but only on what $\nu\,(x)$ is. We have arrived at the correspondence in this example by Fickian diffusion. But if the same $\nu\,(x)$ had been produced by non-Fickian diffusion, or in any other way, it would correspond to the same $q\ (l)$.

The simpler problem of § 6.5 may also be solved in this way.

§ 6.8 *Moments. Standard Deviation.*

Formulæ are usually fitted to frequency curves by way of moments (*vide* Karl Pearson, 'Biometrika,' vol. I, p. 263). Consider, therefore,

$$\frac{1}{\mathrm{N}}\int_{-\infty}^{+\infty} l^n \,.\, q\, dl = \mu_n, \text{ say,} \tag{1}$$

which is the "nth moment-coefficient" of the $q\ (l)$ distribution about its mean $l = 0$.

We shall treat only the case in which N and all the moment-integrals are finite, as occurs when q and ν vanish entirely in the outer regions.

Therefore, in accordance with the definition of q in § 4.3,

$$\mu_n = \frac{1}{\mathrm{N}^2}\int_{l\to-\infty}^{l\to\infty} l^n \int_{x\to-\infty}^{x\to\infty} \nu\,(x)\,.\,\nu_1\,(x,\ l)\,dx\,dl. \tag{2}$$

Now, the limits of integration are independent of one another, so that we may change the sequence of integrations without changing anything else. Let us integrate first with respect to l, remembering that $\rho\,(x)$ is independent of l. Therefore

$$\mu_n = \frac{1}{\mathrm{N}}\int_{x\to-\infty}^{x\to\infty} \nu\,(x)\left\{\int_{l\to-\infty}^{l\to\infty} \frac{1}{\mathrm{N}}\,\nu_1\,(x,\ l)\,.\,l^n\,dl\right\} dx. \tag{3}$$

Since ν_1 is the concentration at the point $(x + l)$, the inner integral is the nth moment-coefficient of the distribution of concentration taken about the point x. On comparing equations (1) and (3) it is seen that :—

> *the* n*th moment-coefficient of the distribution* $q(l)$ *of neighbours, about its centre* $l = 0$, *is the mean of the* n*th moment-coefficients of the distribution of marked molecules taken about every marked molecule in turn.* (4)

Since $q(l)$ is an even function of l its moments of odd order about $l = 0$ all vanish. That is,

$$0 = \mu_1 = \mu_3 = \mu_5 = \mu_7, \text{ etc.} \tag{5}$$

For the even moments the expression can be simplified. The most interesting case is $n = 2$. Let x_m, the centre of the distribution $\nu(x)$, be defined as usual by the equation

$$\int_{-\infty}^{+\infty} \nu(x) \,.\, (x - x_m)\, dx = 0. \tag{6}$$

There is a familiar theorem in mechanics concerning radii of gyration round parallel axes. In like manner it can be proved that if σ_m^2 be the second moment-coefficient of $\nu(x)$ about the centre x_m and σ_x^2 be that about any other point x, then

$$\sigma_x^2 = \sigma_m^2 + (x - x_m)^2. \tag{7}$$

From (7) and (4) it follows that

$$\begin{aligned} \mu_2 &= \frac{1}{N}\int_{-\infty}^{+\infty} \nu(x) \,.\, \{\sigma_m^2 + (x - x_m)^2\}\, dx \\ &= 2\sigma_m^2. \end{aligned} \tag{8}$$

Now the standard deviations of $q(l)$ and $\nu(x)$ are respectively $\sqrt{\mu_2}$ and σ_m. Hence, we have proved that : *the standard deviation of the* $q(l)$ *area from its centre* $l = 0$ *is* $\sqrt{2}$ *times the standard deviation of the marked molecules from their centre* x_m. A glance at the distance-neighbour graph thus gives us a good impression of the size of the cluster on the line.

The skewness of the $\nu(x)$ distribution is not given us by the (l, q) graph, because skewness depends upon a moment of odd order. But the higher even moments could be found. For the fourth moment-coefficient I find

$$\frac{1}{N}\int_{-\infty}^{+\infty} \nu(x) \,.\, (x - x_m)^4\, dx = \tfrac{1}{2}\mu_4 - \tfrac{3}{4}\mu_2^2. \tag{9}$$

The proof of this is omitted for brevity.

§ 7. The Determination of the Diffusivity by Observation.

§ 7.1. *The Standard Deviation of a Linear Cluster formed by Diffusion on a Straight Line from a Point, the Diffusivity for Neighbours being* $\mathrm{F}(l) = \varepsilon l^{4/3}$, *where* ε *is independent of* l. *The Determination of* ε *from Observations.*

It has already been proved in § 5.3 and § 5.4 that under these circumstances we have q given by § 5.4 (1).

To find the second moment μ_2 which is defined in § 6.8 (1), put, as before,

$$\frac{\alpha^2}{4t\varepsilon/9} = \beta^2. \tag{1}$$

Then it may be shown that

$$\mu_2 = 3\mathrm{A}\,(4t\varepsilon/9)^3 \int_{-\infty}^{\infty} e^{-\beta^2}\beta^3\, d\beta. \tag{2}$$

Now if we denote

$$\int_{-\infty}^{+\infty} e^{-\beta^2}\beta^n\, d\beta \text{ by } \mathrm{S}_n \tag{3}$$

it may be proved by integration by parts that

$$\mathrm{S}_n = \frac{2}{n+1}\mathrm{S}_{n+2}. \tag{4}$$

Also it is well known that $\mathrm{S}_0 = \sqrt{\pi}$. And so

$$\mathrm{S}_8 = \frac{7\times 5\times 3\times 1}{2\times 2\times 2\times 2}\sqrt{\pi} = \frac{105}{16}\sqrt{\pi} = 11{\cdot}63. \tag{5}$$

Also for finishing the calculation of N which was begun in § 5.4 we shall need

$$\mathrm{S}_2 = \tfrac{1}{2}\sqrt{\pi} = 0{\cdot}886. \tag{6}$$

With these substitutions it follows that

$$\mu_2 = \frac{3\mathrm{A}\,(4t\varepsilon/9)^3\,\frac{105}{16}\sqrt{\pi}}{3\mathrm{A}\tfrac{1}{2}\sqrt{\pi}} = (4t\varepsilon/9)^3\,\frac{105}{16}. \tag{7}$$

But by the theorem (§ 6.8 (8)) of the previous section σ_m, the standard deviation of the marked molecules from their mean position on the line is $\sqrt{(\tfrac{1}{2}\mu_2)}$.

So

$$\sigma_m = \sqrt{\left\{(4t\varepsilon/9)^3\,\frac{105}{32}\right\}} = \left(\frac{70}{243}\right)^{\frac{1}{2}}(\varepsilon t)^{3/2}. \tag{8}$$

Or solving for ε

$$\varepsilon = \left(\frac{243}{70}\right)^{1/3}\frac{\sigma_m^{2/3}}{t} = 1{\cdot}51\,\frac{\sigma_m^{2/3}}{t}. \tag{9}$$

This formula enables the coefficient ε in the diffusivity for neighbours $\varepsilon l^{4/3}$ to

be determined from the scatter of particles on a straight line. The particles will really move in three dimensions, but the formula can be applied to their projections on the line, as was shown in § 4.7.

Suppose now that the formulæ we have thus deduced from the non-Fickian diffusion of neighbours with diffusivity $\varepsilon l^{4/3}$ represents the true sequence of events, but that the observations have been reduced instead by the formula* which is a necessary consequence of the Fickian diffusion of concentration with constant diffusivity K, namely,

$$K = \frac{\sigma_m^2}{2t}, \tag{10}$$

what values will be obtained for K ?

Eliminating t between equations (9) and (10) it is found that

$$K = \tfrac{1}{2}\left(\frac{70}{243}\right)^{1/3} . \varepsilon . {\sigma_m}^{4/3} = 0{\cdot}330\varepsilon{\sigma_m}^{4/3}. \tag{11}$$

This shows that K, obtained in this way, will increase as the 4/3 power of the size σ_m of the scatter, as, in fact, K does (see fig. 8).

If we regard F (l) as what we wish to find from published values of K, then we must put

$$F(\sigma_m) = \varepsilon{\sigma_m}^{4/3} = \frac{K}{0{\cdot}330}\, 3{\cdot}03\ K. \tag{12}$$

As a necessary preliminary to finding out that F (l) was nearly of the form $\varepsilon l^{4/3}$ I made, in § 5.2, the guess that we should obtain the right order of magnitude for F (l) and l by putting F (l) = 2K and $l = \sigma_m$. These imply that F (σ_m) = 2K. It is seen that the guess is amply justified.

But we may now revise the value of ε from $0{\cdot}4$ to $0{\cdot}4 \times 3/2 = 0{\cdot}6$ cm.$^{2/3}$ sec.$^{-1}$. This is only a mean value roughly applicable under average circumstance in the range one metre $< l <$ 10 kilometres. A more detailed study will reveal variations of 10 times or more in ε according to the up-gradients of temperature and mean-wind and other circumstances. Even so, ε will be remarkably more constant than the diffusivity K for concentration, which, as we have seen, varies with l about a billion times.

* This formula was deduced by Einstein in connection with the Brownian motion ('Ann. der Phys.,' vol. 17 (1905)). Something like it was employed by G. I. Taylor for reducing the Scotia kite ascents ('Phil. Trans.,' A, vol. 215, p. 10). The formula was given explicitly and much used by the present writer in "Some Measurements of Atmospheric Turbulence" ('Phil. Trans.,' A, vol. 221). In the latter paper there are two independent proofs of the formula, one of which in Section IV is a correct deduction from Fick's equation, the other in Section V is quite spoilt, alas, by a wrong sign in equation 3, p. 9, and a risky assumption about correlations. This error affects equation (32) on pp. 15 and 27 of the aforesaid paper, but the rest of the paper holds good independently.

Various observers (Dobson, Richardson, Roberts) have separately noted the fact that the width of an individual trail of smoke from a point source, when measured by its standard deviation σ_m from its mean line, is such that σ_m is roughly proportional to $t^{1/2}$. This implies that the diffusion is Fickian in certain short ranges of l, the diffusivity for neighbours F (l) being there independent of l. When an enormously wider range of l is considered we have seen that F (l) is proportional to $l^{4/3}$ roughly.

[*Note added December* 7, 1925.—The observations collected by Richardson and Proctor in the range σ_m = 3 km. to 86 km. fit well with the slope of the smooth curve in fig. 8.

These apparently contradictory facts may perhaps be reconciled if we regard variations in K, defined by §1.1 (1), as being due to variations in the type of mean chosen in forming $\bar{u}$, $\bar{v}$, $\bar{w}$ in the same equation. As long as the mean is always taken over the same length and time, K may well be more or less constant.]

§7.2. *Theory of a Second Method by which the Diffusivity* F (l) *might be observed.*

It has just been shown that the value of ε in $\varepsilon l^{4/3}$ = F (l) can be extracted from observations made with a wide range of l all in operation together. While this is possible mathematically, it seems, from the standpoint of practical physics, to mix too many phenomena and unnecessarily to assume that ε is independent of l. It would be better to observe separately at or near each selected l_0. This can be done by a process which will be derived from the non-Fickian equation

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial l}\left\{\mathrm{F}\,(l)\,\frac{\partial q}{\partial l}\right\}.$$

Suppose that the "marked molecules" are initially concentrated in two plane parallel laminæ, distant l_0 from one another and similar to one another. The neighbour-distance diagram then consists of three sharp peaks, as suggested qualitatively in fig. 12. The central peak shows the very close neighbours

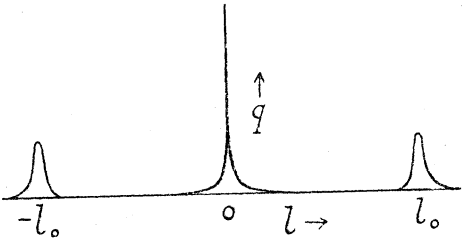


FIG. 12.

which each marked molecule has in its own lamina. The lateral peaks show the neighbours in the other lamina. The spreading of the central peak will go on very much as if there were only a single lamina (§ 5.4, § 7.1), and does not concern us now. If the laminæ are observed during a time such that they spread through only a small fraction of l_0, then we may regard F (l) as a constant and equal to F (l_0) in this short range of l, and may write accordingly

$$\frac{\partial q}{\partial t} = \mathrm{F}(l_0) \cdot \frac{\partial^2 q}{\partial l^2}.$$

An appropriate solution of this equation is

$$q = \mathrm{A} t^{-\frac{1}{2}} e^{-\frac{(l-l_0)^2}{4t\mathrm{F}(l_0)}},$$

which implies that the peak on the (l, q) diagram has the form of the normal curve of error, q playing the part of the "frequency." The standard deviation of the curve from its mean l_0 is $\{2t\mathrm{F}(l_0)\}^{1/2}$.

In determining the standard deviation from the observations it is, of course, essential that the individual values of $(l - l_0)^2$ should be weighted in the proper way. On referring back to the definition of Q in § 3.2 it is seen that we must form every possible pair of marked molecules, one from each lamina, so obtaining a set of distances l. Let $[\quad]_Q$ denote the mean for all members of the set. Then in ordinary circumstances $[l]_Q = l_0$. We next form the mean squared deviation from the mean and thus find

$$\mathrm{F}([l]_Q) = \frac{[(l - [l]_Q)^2]}{2t}. \qquad (1)$$

In practice the "marked molecules" could be replaced by balloons, for those values of l_0 which are many times the diameter of a balloon. This is so because F (l) increases notably with l. C. H. Ley* has invented a valve which allows a balloon to rise to a pre-arranged height and then lets out some gas so that the balloon ceases to move through the air. That is the type of apparatus required.

When there are only one or two observers, they could not manipulate many balloons at once, but they might observe pairs of balloons on successive days. Let us try to adapt the observation to this situation. Imagine first, for the sake of the argument, that the observation with many balloons arranged initially in two parallel laminæ l_0 apart, is made, and let us denote $(l - l_0)^2$ by p

* 'Quart. Journ. Roy. Meteor. Soc.,' p. 247 (1911).

for short. Thus many values of p are obtained on one occasion. Next imagine that this observation were repeated once a day for a year, the time t being in all cases the same ; and not too small nor too large. In this way a double set of values of p is obtained, thus :—

Days.	Pairs on one day $\longrightarrow$				
$\downarrow$	p_{11}	p_{12}	p_{13}	p_{14}	$\cdots$
	p_{21}	p_{22}	p_{23}	$\cdots$	
	p_{31}	$\vdots$	$\vdots$		
	$\vdots$				

The mean value of $\mathrm{F}(l_0)$ for all these observations is $\mathrm{F}(l_0) = [p]_{QS}$ the double suffix denoting the double mean, Q for rows, S for columns. We cannot proceed further without making *the assumption that we obtain the general mean of p if we select at random one value of p from each row of the double set and take the mean of them.* This looks passable.

If so

$$\mathrm{F}(l_0) = \frac{[p]_S}{2t},$$

which can be determined by flying a pair of balloons on each of the many days.

This measure of atmospheric diffusion is in agreement with that to which we were led by a search for a natural mean (§ 2).

§ 8. Summary, Conclusion and Abstract.

The atmospheric diffusivity in Fick's equation has been found by various investigators to increase from 0·2 to 10^{11} cm.^{2} sec.$^{-1}$ as the size of the cluster of diffusing particles increases from 10^{-2} to 10^8 cm. The effect is due to eddies of many sizes acting together. There is apparently no way of modifying Fick's equation in order to describe this phenomenon. But a new mathematical method is here developed in which instead of thinking about concentration as a function of position, we think about q, the mean number of neighbours per length, as a function of l, their distance apart. Formal definition is given to this idea, and various properties of it are investigated. For simplicity only distributions on an unbounded straight line are considered, or projections of three-dimensional distributions on to the line. If the movement of concentration ν is described by Fick's equation,

$$\frac{\partial \nu}{\partial t} + \bar{u}\frac{\partial \nu}{\partial x} = \mathrm{K}\frac{\partial^2 \nu}{\partial x^2},$$

where t is time, x is distance, $\bar{u}$ is mean velocity, and K is diffusivity. Then it is proved that

$$\frac{\partial q}{\partial t} = 2\mathrm{K}\frac{\partial^2 q}{\partial l^2}.$$

If, however, the diffusion is "non-Fickian," as in the atmosphere, then the former of these equations cannot be generalised, but the latter can, taking the form

$$\frac{\partial q}{\partial t} = \frac{\partial}{\partial l}\left\{\mathrm{F}(l)\frac{\partial q}{\partial l}\right\}.$$

A discussion of existing observations shows that a rough average value is $\mathrm{F}(l) = 0{\cdot}6\ l^{4/3}$ cm.^{2} sec.$^{-1}$ for the atmosphere, when l lies between one metre and 10 km. The diffusion of a lamina is worked out from these principles. The diagram obtained in this way exhibits the size of a cluster because it is proved that the standard deviation of the (l, q) area from its centre $l = 0$ is $\sqrt{2}$ times the standard deviation of the cluster from its centroid.

Two methods are prepared for the observation of $\mathrm{F}(l)$ by balloons or smoke.

Various allied topics are examined.

§9. List of Recurring Symbols with the Sections where they are defined.

K, $\bar{u}$, x, y, z, t, § 1.1; Δ, § 1.2; [ ], § 1.3; Q, h, N, $\mathrm{A}_{n,n+1}$, l, § 3.2; ν, § 4.2; q, ν_1, § 4.3; Π, § 4.5; $\mathrm{F}(l)$, § 5.1; ε, § 5.2; α, § 5.3; β, § 5.4; σ_m, μ_n, § 6.8.