

## SECTION V.—PAPERS AND DISCUSSION ON “COLLOIDAL PHENOMENA”

### ON CLAYS AS DISPERSE SYSTEMS.

BY SVEN ODÉN.

#### I. THE DEFINITION OF CLAY.

If one searches the literature of geology and agriculture for a definition of the meaning of the term “clay,” one finds a number of attempts to attach the characteristic properties of clay to certain definite substances, which are supposed to be the chief components, and, though more or less impure to be the essential components of the natural clays in all their forms.

As such an essential component the hydrated silicate of aluminium, *Kaolin*, was generally regarded formerly. Kaolin was later replaced by the earthy zeolites. (Th. Way, J. M. van Bemmelen, R. Gans): and more recently by many sorts of soil colloids: colloidal silicic acids, aluminium hydrate, iron oxide, allophane, and so forth (F. Cornu, P. Rohland, H. Stremme, and others).

A view differing in principle was put forward by A. D. Hall.<sup>1</sup> According to him it is not the chemical nature of the substance which characterises clay, but, before everything, the fineness of the constituents. This view was also upheld in principle by A. Atterberg,<sup>2</sup> though he was of the opinion that Muscovite and Biotite were the characteristic minerals in the Swedish quaternary clays.

For the reasons which are set out below I think, in agreement with Hall, that the following definition of clay is in accordance with experimental results: Clays are disperse formations of mineral fragments in which particles of smaller dimensions than  $2\mu$  predominate.

Among these mineral fragments, quartz, felspar, mica, and others may occur, as well as their different weathering-products, kaolin, bauxite, hydrargillite, zeolites, allophane (Stremme), gelatinous silicic acid, iron oxide, and so forth, but none of these constituents is necessary to characterise a sedimentary soil as clay. All that is necessary is that the smallest particles preponderate. The limit is at present set, according to Atterberg, at  $2\mu$  and one may reckon on more than half of the total amount of particles falling about this limit, but these limits are, of course, only arbitrary, and strata where particles of 2 to  $5\mu$  magnitude compose the greater portion, show the transition to sand and silt formations. It is also probable that the limits of magnitude within which the characteristic properties of clay are found are dependent on the chemical properties of the material, and a clay composed

<sup>1</sup> “The Soil” (London, 1912), pp. 34-39.

<sup>2</sup> A. Atterberg, *Intern. Mitt. f. Bodenkunde*, 1, 7 (1911), 7, 718 (1914); *Kolloidchem. Beih.*, 6, 55 (1914).

chiefly of kaolin and quartz must in all probability be appreciably finer than clay consisting of, for example, biotite and hydrated silicic acid, in order to attain the firmness and plasticity typical of clay. In this connection it may be pointed out that it is not any special chemical colloids which comprise the clay colloids. The true soil-colloids are the innumerable fragments of both weathered and unweathered minerals, crystal chips and amorphous substances, which, in a state of fine division, constitute the clay.

By *colloids* or disperse formations, we understand, in physical chemistry, two-phase systems where the one component, for example, the solid phase (here the mineral grains), occurs in finely divided (disperse) form, and is dispersed in the other component, the dispersion medium (for example, water).

But while, in colloidal solutions and in the majority of cases in gels, the disperse phase constitutes the smaller part of the system (often a few per cent. at the highest), in the clays the disperse phase preponderates, and the dispersion medium (water, or water vapour and air) is only to be regarded as a filling for the interstices between the mineral particles.

The clays also, in contrast to the majority of other colloid systems, consist of grains of all possible sizes, from sand grains of one millimetre to particles of one hundred-thousandth part of a mm. ( $0.001\mu = 100\mu\mu = 0.00001$  mm.). The importance of the percentage distribution of the mineral grains over the different sizes for differentiating and classifying clays will be dealt with further on.

An enormous increase in surface area proceeds hand in hand with this higher subdivision, which subject has been treated in my previous note. This large surface also brings about adsorption of different salts, and even gases, by the particles, which are thereby fixed, and protected against washing out.

But while, according to the reasoning set out above, it is not the chemical nor the mineralogical constitution of the material, but only its state of division, which decides the apparent properties of the clays, the chemical nature of the constituents plays, of course, an important role in the agricultural and technical valuation, and in the more minute characterisation of the different clays. We must here call in the aid of chemico-analytical methods. Against the complete analysis ("Bausch analysis") of course, no objection can be advanced. It gives us the complete quantitative composition of the clay. But to proceed, as was formerly usual, to call the portion soluble in hydrochloric acid "zeolitic weathered-silicate," and that soluble in concentrated sulphuric acid "kaolin bodies," is misleading. For it is necessary in all extraction methods to take into consideration the degree of fineness of the material. The finer the mineral grains are, so much the more easily are they attacked by acids, and, with the exception of quartz, small mineral particles of all sorts are attacked by hydrochloric acid.<sup>1</sup>

## 2. THE CHARACTERISATION OF THE CLAYS BY MEANS OF DISTRIBUTION CURVES.

We have seen how the clays are characterised by the fineness of the particles. How shall we now distinguish the degree of this division among the minerals? The answering of this question has been the task of

<sup>1</sup> P. Zemjatschensky, 11 *Journ. de la confér. de natur. et méd.*, St. Petersburg, 1901, p. 73; A. Vesterberg, *Intern. Mitt. f. Bodenkunde*, 5, 37 (1915); S. Odén and A. Reuterskiöld, *Bull. of the Geol. Inst. of Upsala*, 16, 148 (1919); *Intern. Mitt. f. Bodenkunde*, 10, 343 (1920).

*mechanical soil-analysis*, the object of which is to divide up the soil-strata in fractions according to the size of the particles, and to determine the weight of these fractions. But this grading must, for practical reasons, be limited to the isolation of a fairly limited number of fractions, and, as the soil-strata contain particles from the size of sand grains a millimetre large down to amicros, a classification limited in this way to certain groups of particles is always very imperfect. Two soils might give almost identical figures in such a mechanical analysis and yet be very different with regard to the nature of the grains. This is probably the chief reason why classifications grounded on mechanical soil analysis have regularly failed in their object, and must be abandoned. Mechanical soil analysis calls all particles under  $2\mu$  clay. Since it is just these smallest particles which are characteristic of clay, but which, in different clays, may have quite different degrees of smallness, we must lay the greatest stress on the characterisation of just these smallest particles. One might now take into consideration the colloid-chemical methods worked out by Zsigmondy, which consist in counting the number of particles in a limited volume of a suspension of known concentration, but even such methods of procedure are incomplete, for they give a figure only for the mean of the magnitudes of the particles, and these values are often far from the real size of the most numerous particles.<sup>1</sup>

A determination of the distribution, that is to say, of the weight or the number of particles corresponding to each size, here shows itself to be very desirable.

Before entering upon this problem we shall first clearly decide what may conveniently be meant by the term "dimensions," applied to such small mineral particles as these, for to give the length, breadth, and height of all the different mineral fragments is manifestly impossible.

In practice the particles are studied in contact with water, and for this reason I have previously in another place<sup>2</sup> proposed that the "effective radius" of a particle should be defined as the radius of an imaginary sphere of the same material which would sink in water with the same velocity as the particle in question. The detailed argument for this, as well as the laws governing such motions of small particles in a liquid were set out in the same paper.

It was probably Maxwell who first used distribution curves to characterise physical systems, as, for example, to demonstrate the different velocities of the molecules in a gas. The use of the same procedure for soil layers presumes that it is possible to express the number of the particles graphically as a continuous function of the effective radius. This is most easily illustrated by means of a few examples.

If we consider a clay composed of particles of the size of  $1\mu$ ,  $2\mu$ ,  $3\mu$ , . . .  $100\mu$ , as well as of particles less than  $1\mu$ , we may express the mass of particles  $< 1\mu$ ,  $1\mu$  to  $2\mu$ ,  $2\mu$  to  $3\mu$ , and so forth, in percent. of the whole. Such a set of figures would have the same meaning as the results of an ordinary mechanical soil analysis, with the difference that it would be divided into 100 groups, instead of the 3 or 5 of the soil analysis. But to make the figures more easily comprehensible it is more convenient to set them out graphically. This may be done in several ways.

In Fig. 1, are set out as abscissæ the effective radii, and as ordinates a value  $q(r)$ , which indicates the percentage of the sample consisting of

<sup>1</sup> Odén, *Koll. Zeitschr.*, **26**, 109 (1920).

<sup>2</sup> *Intern. Mitt. f. Bodenkunde*, **5**, 257 (1915).

particles larger than this effective radius. For the effective radius 0, this value is 100, for effective radius  $10\mu$  for example, it is 75, for effective radius  $30\mu$ , it is 50. This indicates that 75 per cent. of the sample consists of particles larger than  $10\mu$ , 50 per cent. larger than  $30\mu$ , and so forth.

Between  $0\mu$  and  $10\mu$  lie, therefore,  $100 - 75 = 25$  per cent., and so on. To obtain the percentage between any two abscissæ we, therefore, subtract the ordinate corresponding to the larger effective radius from that corresponding to the smaller, the difference corresponding to the percentage of particles. This graphical function declines therefore from 100 for the smallest particle to 0 for the largest in the sample. If the curve is horizontal it shows that there are no particles at all in this interval.

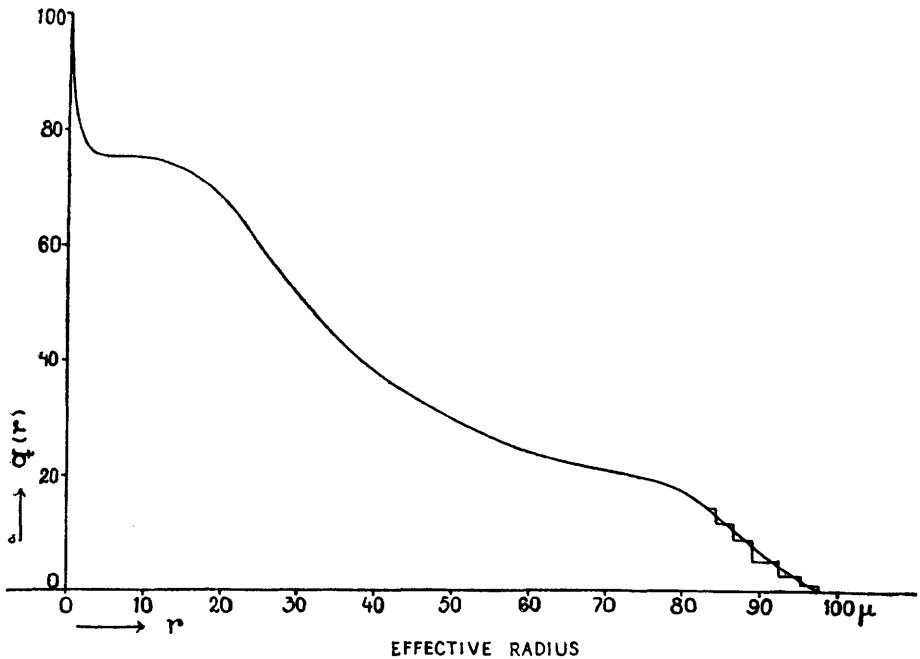


Fig. 1

One entirely theoretical remark may be permitted here. When drawing this curve, if one desires to use a scale such that each particle would be represented, the curve would, strictly speaking, not have this continuity, but would have a step-like appearance such as is represented to the right of Fig. 1, because the number of particles, despite its large amount, is limited, and there must always be size values which have no particles. In reality these intervals of millionths of one per cent. are negligible, and we may give the curve a continuous outline.

The above method of representing the composition of a soil is specially convenient for the treatment of certain problems. In most cases where we wish to get an idea of the numbers of the contents of the different fractions it is better and easier of comprehension to represent the amounts of the particles not as a line but as a surface. This is accomplished by the method shown in Fig. 2, where the effective radius is as before set

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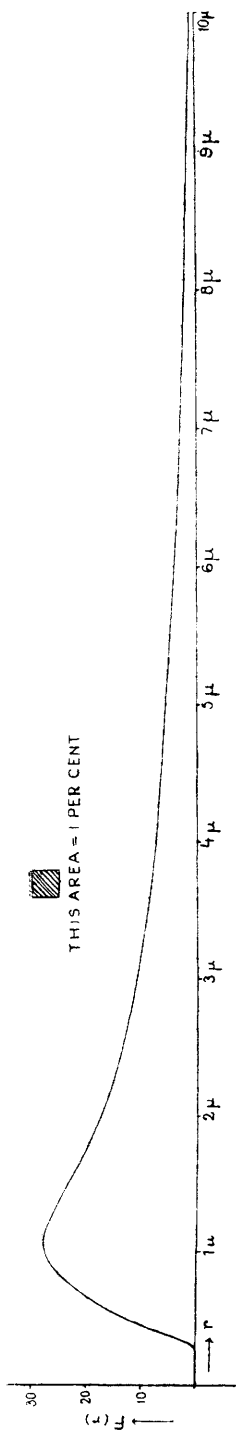


FIG. 2.

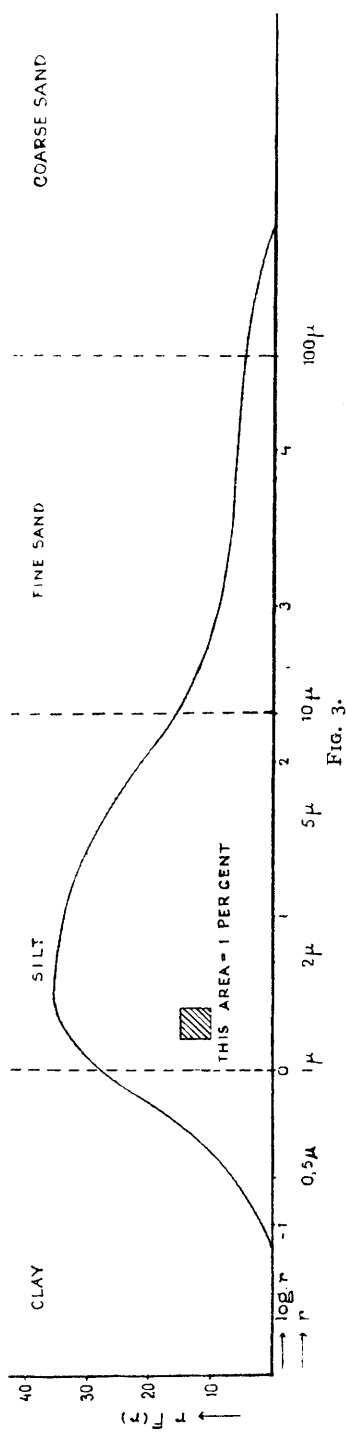


FIG. 3.

off as abscissæ, but where we measure as ordinates, a value  $F(r)$ , such that this value, multiplied by a part of the abscissa axis, gives us a surface which represents the number of particles in the interval of abscissa in question. The number of particles is here represented as an integral, and the number of particles between any two radius-values is obtained simply from the surface encompassed by the axis of abscissa, the corresponding ordinates, and the curve. The easiest way to carry out the graphical representation is to arrange that the total surface comprises 100 units on millimetre-paper, when it is only necessary to reckon the squares for each interval to arrive at the percentage.

If we are considering a typical clay soil, the curve will appear as represented in Fig. 2. If there are, on the contrary, particles of a large range of sizes it would be very inconvenient to use radii as abscissæ, because of the length. If we there use  $r \cdot F(r)$  instead of  $F(r)$  the surface content is not altered. The curve merely assumes a rather different appearance, and is easier to deal with.<sup>1</sup> Cf. Fig. 3.

I have in Fig. 3 drawn lines through  $1\mu$ ,  $10\mu$ , and  $100\mu$ , since according to Atterberg's<sup>2</sup> researches these limits are the most convenient to characterise the groups clay, silt, fine sand, and coarse sand.

*A soil-layer is therefore a clay, a silt, or a sand soil according to whether the greatest part falls within the limits for clay, loam, or sand.*

The properties of firmness, plasticity, and permeability to water, etc., are nevertheless not dependent only on the preponderating constituent and its fineness, but also on substances occurring to a smaller extent, and therefore greatly dependent on the nature and character of the distribution curve.

As we have seen, distribution curves characterise the loose soils far better than the results obtained by mechanical soil analysis. The determination of these curves may be carried out by carefully levigating a clay in water, and then studying the rate of sedimentation. This may be accomplished by suspending a fine plate from a balance beam immediately over the bottom of the sedimentation vessel, and from time to time determining the weight of the deposited particles. The rate of accumulation of these particles is dependent at every moment both on the number of the particles and their weight and distribution, and this may be calculated therefrom.

Before going into this the experimental method may be sketched briefly.

### 3. THE METHOD OF EXPERIMENT.

The experimental method is based, as before mentioned, on the suspension of a metal plate A, over the bottom of the vessel in which the sedimentation is carried out. The plate is suspended by fine gold wire from the balance B, which allows the sediment to be weighed continually.

It is not necessary that the plate should be as large as the real bottom of the sedimentation vessel, but it may simply be regarded as the base of a cylinder whose height  $h$  is identical with the distance from the plate to the surface of the liquid. The remaining parts of the suspension nearer the

<sup>1</sup> Since  $d(\ln r) \cdot rF(r) = \frac{1}{r} \cdot dr \cdot rF(r) = drF(r)$ .

<sup>2</sup> A. Atterberg, *Intern. Mitt. f. Bodenkunde*, **1**, 7 (1911), 7, 718 (1914); *Kolloid chem. Beih.*, **6**, 55 (1914).

sides of the vessel may be disregarded, if we reckon all weight determinations in percentage of the total weight  $P_{\infty}$  after all the particles have finally subsided.

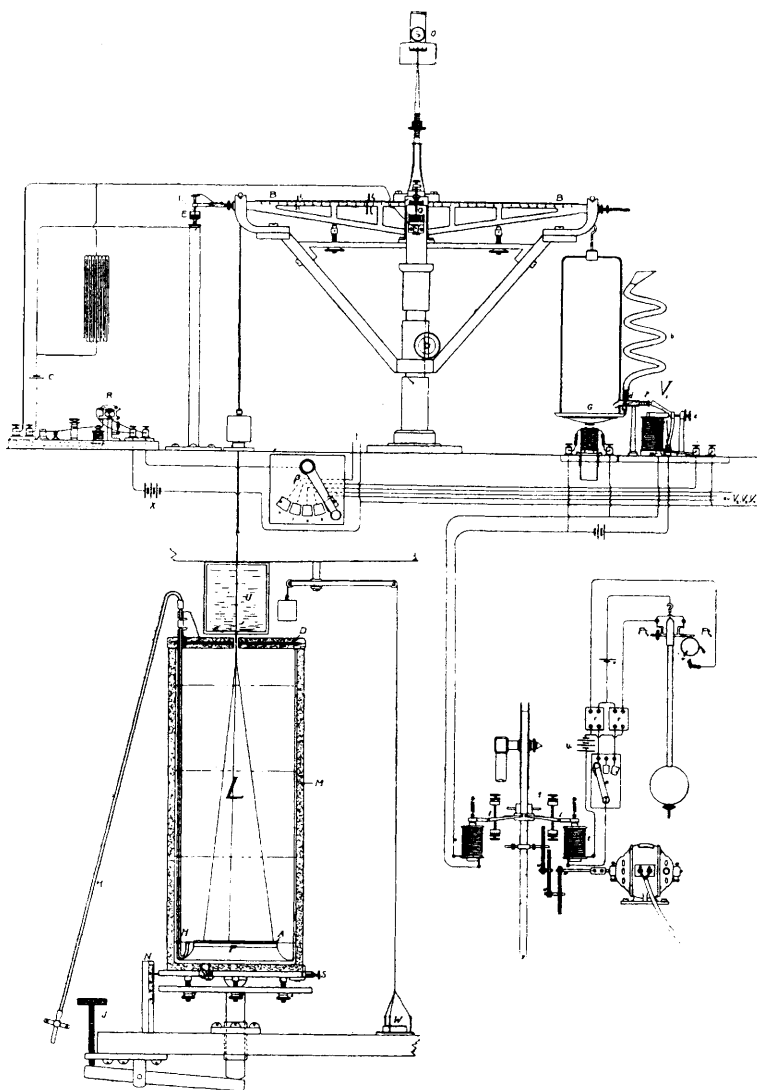


FIG. 4.

If the balance is adjusted at the beginning to equilibrium, and a weight<sup>1</sup>  $d$  is afterwards placed on the balance pan  $G$  and the sediment allowed to deposit on the plate until this weight is counterpoised, then the balance

<sup>1</sup> Steel balls of the weights of (b) 0.1276 g. (c) 0.0544 g. (d) 0.0153 g. were used for weights.

swings to the left, making the electric contact at E, which causes the time to be marked on the telegraph tape by the apparatus RC*nl*, and further causes the automatic feed V to introduce another weight on to the balance pan G. This breaks the contact at E until equilibrium is again attained, and the weight of 2*g* is reached, when the time is again marked.

The times corresponding to equilibrium may be read off at leisure by means of the apparatus—also electric—from the clock Pt. *rtl*, which marks seconds or minutes on the telegraph tape.

By raising the stop and properly adjusting the contact at E we may bring the right and left positions of the balance very near to each other, and also to the equilibrium position, and so avoid unnecessary oscillations in the liquid.

I have in another place<sup>1</sup> described in detail the experimental arrangements, and the best way of recording the results. It suffices to mention here that all the experiments were carried out in a constant-temperature room without heating arrangements, and that generally the variation of temperature per day did not exceed 0.1 to 0.2 deg. C., and consequently no special measures to compensate for varying weight of the plate due to changed specific gravity were taken. And further, that both Th. Schloesing père<sup>2</sup> and I have shown, by purely experimental methods, that, provided that the suspension be not too concentrated, the path of the falls of the individual particles may be regarded as being relatively uninfluenced by those of the others; in other words, that the accumulation curve, after correction to a certain normal height, 10 cm.,<sup>3</sup> and to percentage of the final weight, is *relatively* independent of both the absolute height of fall and the total weight of the levigated substance.

Theoretically, of course, in this case, the conditions necessary to the application of Stoke's law are not experimentally fulfilled, but if we avoid concentrations over 1 per cent. we may in practice assume that the particles fall independently of each other.

The recording was not usually continued for longer than 100 hours. The whole of the sediment had, of course, seldom fallen in this period, but, since it was necessary to determine  $P_{\infty}$ , at the end of this period the remaining liquid was carefully drawn off through the syphon *h*, from the height of the plate, and a new determination made with half of this after addition of a coagulator, whereby the weight of the undeposited residue was determined. The sum of the final weight in experiment 1 and twice the weight found in experiment 2 then give us  $P_{\infty}$ . In the other half the mean size of particle was determined ultramicroscopically, according to Zsigmondy-Siedentopf.

Let us regard the particles as spheres of specific gravity *S* and call the effective radius *r*. If we optically delimit a small volume, *v* cm.<sup>3</sup>, of the diluted clay suspension, we may, if *p* be the concentration of the disperse phase in gm./cm.<sup>3</sup> and *n* the number of particles in the volume *v*, calculate the radius from the equation

$$r = \sqrt[3]{\frac{3}{4\pi} \times \frac{v \cdot p}{s \cdot n}} \quad \dots \quad (1)$$

The specific gravity is always taken as 2.7, *v* is expressed here in cub. cm.

<sup>1</sup> *Bull. of the Geol. Institut.*, Upsala, **16**, 15 (1918).

<sup>2</sup> Th. Schloesing père, *Compt. rend. de l'Acad.*, Paris, **136**, 1608 (1903).

<sup>3</sup> For a fall of 5 cm., it is therefore necessary to multiply the time by 2, for 20 cm. fall, by 0.5, and so on, since the rate of fall is inversely proportional to the height of fall *h*.



#### 4. CALCULATION OF THE DISTRIBUTION CURVE FROM THE ACCUMULATION CURVE.

In the following, by *distribution curve* is indicated the function  $F(r)$ , which gives the relation between percentage weights of the suspended material and the corresponding radii, by *accumulation curve*  $P(t)$ , the weight of sediment (weighed in water) as a function of the time. If  $P_1, P_2, P_3$ , be the weights corresponding to the times  $t_1, t_2, t_3$ , we may, for a small interval, write:—

$$\frac{P_2 - P_1}{t_2 - t_1} = \frac{dP(t)}{dt}.$$

The different values of the angle coefficient, or, in some cases, more conveniently the logarithm of the angle coefficient, of this accumulation curve at different times as a function of the time (or logarithm of the time), is designated *the auxiliary curve*, and by means of this it is possible to determine graphically the other derivatives of the accumulation curve.

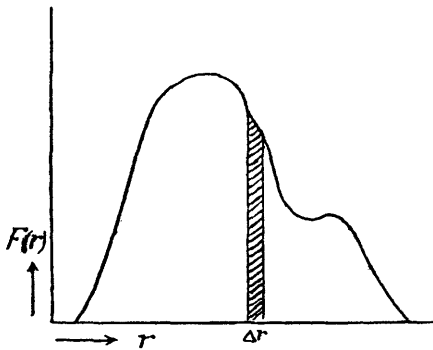


FIG. 6.—Distribution Curve.

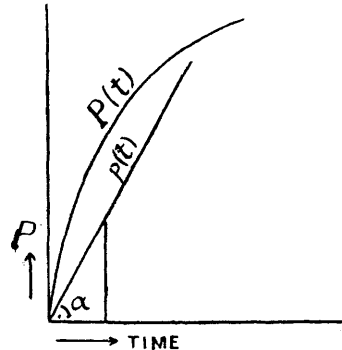


FIG. 7.—Accumulation Curve.

Let us first consider the simplest case, where all the particles have the same effective radii and are similarly distributed in the liquid, and let  $p$  be the amount deposited as a function of the time. The accumulation must then proceed at a rate proportional to the rate of fall  $v$  of the particles, and to the concentration  $c$ . The latter can, if the total amount of the particles after complete deposition =  $A$ , the area of the bottom of the sedimentation cylinder =  $\tau$ , and the height of fall =  $h$ , be written  $c = \frac{A}{h}$ .

The rate of accumulation then becomes  $\frac{dp}{dt} = \frac{A \cdot v}{h} \cdot \text{Const.}$

If  $p$  be here expressed as a fraction of  $A$ , and  $v$  be given in terms of the same units as  $t$  and  $h$ , this constant =  $\tau$ .

The weight of deposited sediment (Fig. 7),  $p$ , after a certain time,  $t$ , is therefore clearly

$$p = t \cdot tga = t \frac{dp}{dt} = t \frac{A \cdot v}{h} \quad (2)$$

This equation holds good, however, only so long as there are any falling particles in the fluid, that is to say, until  $p = A$ . This time during which the equation holds is clearly the same as that which the topmost particle requires to reach the bottom, and is obtained by writing in equation (2),  $p = A$ .



If  $P_1, P_2, P_3$ , and so on, be the weights of the sediment at the times  $t_1, t_2, t_3$ , and so on, we may, provided that we have sufficiently small weights, write

$$\frac{dP}{dt} = \frac{P_2 - P_1}{t_2 - t_1}$$

and thus from the observation material *calculate* the first differential coefficient of the accumulation curve.

If we now set off as abscissæ

$$x = \log_{10} t = \ln_e t \cdot \log_{10} e,$$

and as ordinates

$$y = \log_{10} \frac{dP}{dt} = \ln_e \frac{dP}{dt} \cdot \log_{10} e,$$

we obtain a discontinuous curve consisting of horizontal parts. If we substitute for this a smooth continuous "auxiliary curve," we may determine the second differential coefficient of the accumulation curve from the angle-coefficient for the tangent of this curve. We here use logarithms with the purely practical object of avoiding excessively large and unwieldy figures.

$$\frac{dy}{dx} = \frac{\frac{d^2P}{dt^2}}{\frac{dP}{dt}} \cdot \frac{dt}{dx} \cdot \log_{10} e = \frac{\frac{d^2P}{dt^2}}{\frac{dP}{dt}} \cdot t \quad \dots \quad (8)$$

But equation (7a) may be written in the form

$$F(\rho) = - \frac{2t}{\rho} \cdot \frac{\frac{d^2P}{dt^2}}{\frac{dP}{dt}} \cdot t \cdot \frac{dP}{dt}$$

or, according to (8)

$$F(\rho) = - \frac{2t}{\rho} \cdot \frac{dy}{dx} \cdot \frac{dP}{dt} \quad \dots \quad (9)$$

For the construction of the distribution curve we must in this equation decide upon fitting radius values ( $\rho = 2\mu, 1\mu$ , and so forth), *calculate* the corresponding  $t$ -values according to 7b, *determine graphically* from the auxiliary curve  $\frac{dy}{dx}$  for each value of  $t$ , and, finally, calculate  $\frac{dP}{dt}$ , either from the data of the observation or from the auxiliary curve, by finding the  $y$ -value corresponding to each value of  $\log t$ .

Examples of calculations showing the method of working out have been previously published in the *Proc. Roy. Soc.*, Edinburgh,<sup>1</sup> and I only give in Figs. 8, 9, 9b, 10, 11, 16, 17, 18, and 19, some examples of the distribution curves obtained for various soils.

##### 5. THE EXTENSION OF THE RESEARCHES TO SEDIMENTATION IN MORE VISCOUS LIQUIDS.

The use of sedimentation liquids of smaller specific gravity and viscosity, whereby the deposition would proceed more rapidly and an extension of the registered region to the left might be obtained in a reasonable time, cannot be carried out in practice. For with all liquids which could here come in question there occur, because the greater volatility, and the cooling of

<sup>1</sup> Odén, *Proc. Roy. Soc.*, of Edinburgh, 36 (III.), No. 13, 219-35 (1916).

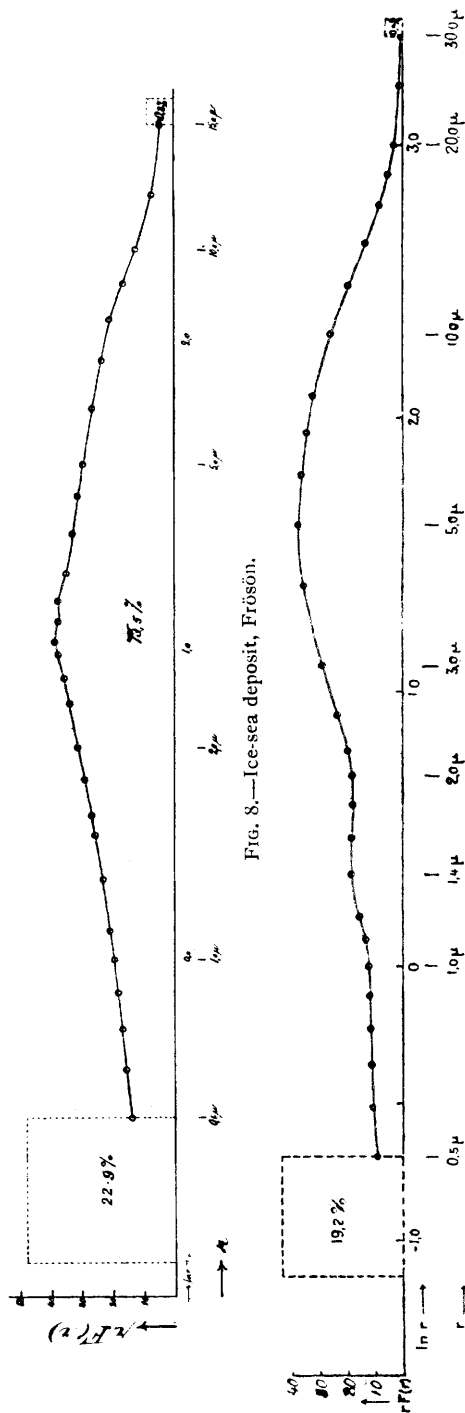


FIG. 8.—Ice-sea deposit, Frösön.

FIG. 9.—"Rhaet" clay, Bornholm.

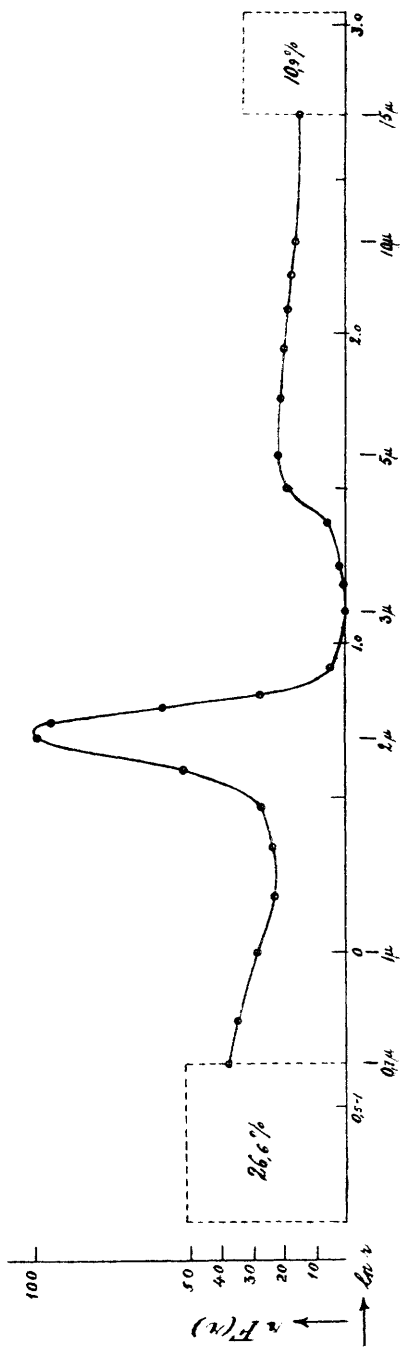


FIG. 9b.—Medium oligocæne silt from Skive.

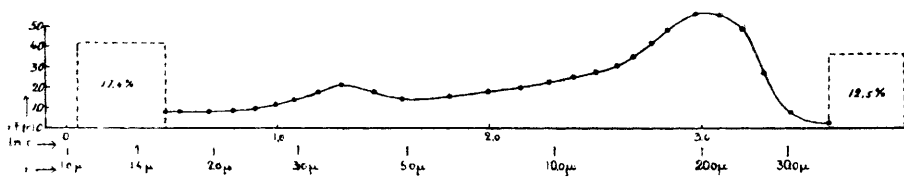


FIG. 10.

the surface caused thereby, convection currents which interfere with the deposition. On the other hand, an addition of glycerine (sp. gr. 1.3, viscosity about 11 at 15° C.) to the suspension under examination delays the deposition of sand and silt particles to such an extent that it becomes possible to observe particles up to  $r = 50\mu$ , that is, up to 0.1 mm. diameter.

An observation of this nature of a fairly coarse Danish mica-clay from the Upper Miocene strata at Vemb (near Lemvig) was carried out at 15° C. in a glycerine-water mixture of  $d = 1.115$  and  $\eta = 0.04925$ , with a height of fall of 30 cm. I refrain from giving the observations and only give, in Fig. 10, the distribution curve calculated therefrom.

From  $30\mu$  the distribution curve may be regarded as fairly reliable. The coarser sand grains, which sink almost at once, are assembled together to the right.

As a proof of the correctness of the above assumptions and as an example of the method of working when it is a question of earths of particles very various in size, we may here give the figures from a research on a kaolin soil 107 Sandbäck.

Since it is impossible to observe the deposition of particles larger than 0.1 mm. in the sedimentation apparatus, the coarser material, "sand," was levigated out to begin with in the Atterberg levigation cylinder with a height of fall of 30 cm., and a time of fall of 15 seconds. After the dry weight had been determined, there had in this way been isolated  $\frac{48.27}{100}$  as fraction A. According both to Oseen's<sup>1</sup> modification of Stokes' formula and to Atterberg's experimental results, this comprises the particles of an effective radius  $> 100\mu$ . It was not further examined, but was recorded as a square to the right in Fig. 11.

One further separation was carried out in the Atterberg cylinder at 15° C. and 20 cm. height of fall, for a period of 20 minutes. The particles no longer in suspension, "Fraction B," amounted to  $\frac{19.87}{100}$ , and the suspended portion, amounting to  $\frac{31.86}{100}$ , was taken as the residue, in good agreement with an analysis which showed 32 per cent. The two fractions, B and C, were then sedimented separately in different liquids, with different heights of fall, and at different temperatures.

	Specific D.	$\eta$ .	Temperature.	Height of Fall.
Fraction B in water + glycerine .	1.125	0.05642	15° <sub>2</sub>	15 cm.
Fraction C in water . . . . .	1	0.01155	14° <sub>5</sub>	10 cm.

In spite of this the curves coincide in a quite unmistakable way, as is seen in Fig. 11, and give a picture of the mechanical constitution of the whole region from  $0.4\mu$  to  $100\mu$ . The two small squares at  $8\mu$  indicate that the separation from the levigation was not quite complete, so that fraction B still contained some still finer particles. The levigations were carried out with a view to make possible the determination of the sand particles in a more viscous liquid. They were, certainly, separated in the Atterberg apparatus, but not according to the times of deposition fixed by him. Another matter, which,

<sup>1</sup> C. W. Oseen, *Arkiv. f. matematik, etc.; Utg. av K. Svenska Vet. Akad. i., Stockholm*, 6, No. 29 (1910); 9, No. 16 (1913).

with regard to what follows, is worth noticing, is that my assistant, when calculating and drawing the curve, was given only the figures of the observation and the physical data, but otherwise received no further information regarding the origin of the sample, nor of any previous researches thereon.

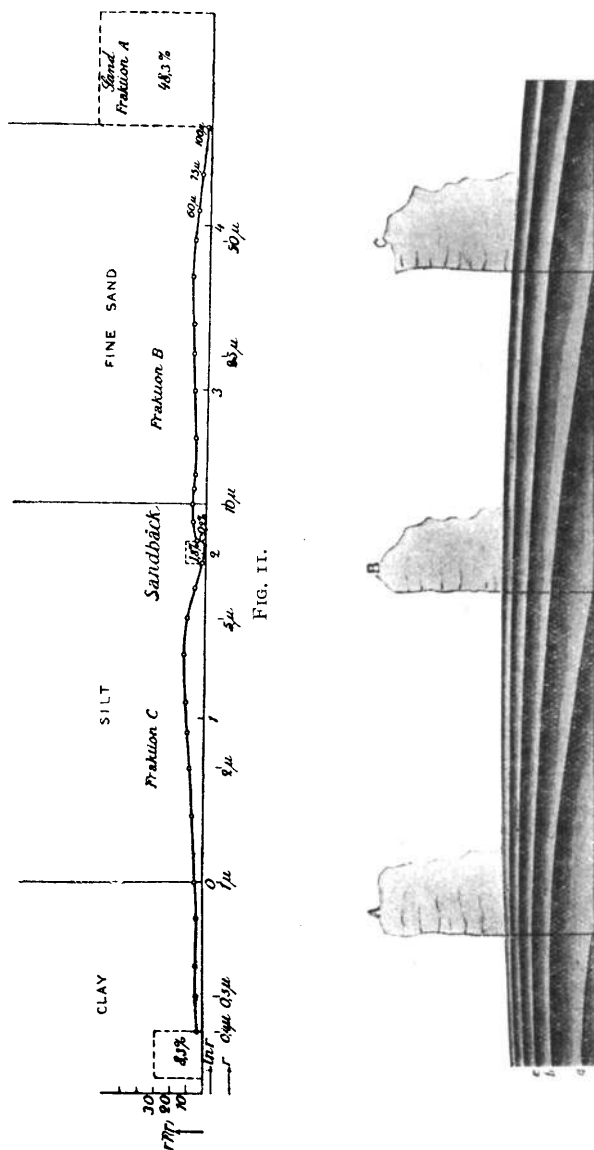


FIG. 11.

FIG. 12.—Diagrammatic section in the direction of the ice-edge through the lower strata of the stratified clay, according to De Geer. Three successive stages of the ice-edge, A, B, C, each with its corresponding layer of clay, *a*, *b*, *c*.

If we draw perpendiculars through the effective radii  $1\mu$ ,  $10\mu$ , and  $100\mu$ , the distribution curve is divided into the International (Atterberg's) groups of clay, silt, fine sand, and sand. If the curve be drawn on millimetre paper the weights may easily be determined by counting the squares, thus giving the figures for a mechanical analysis of the soil.

These gave :—

Clay up to $1\mu$	(diameter $2\mu$ )	.	.	13'0	per cent.
Silt $1.0\mu$ to $10\mu$	( „ $2-20\mu$ )	.	.	21'9	„
Fine sand $10\mu$ to $100\mu$	( „ $20-200\mu$ )	.	.	20'0	„
Sand $> 100\mu$	determined by levigation	.	.	48'3	„
				<u>103'2</u>	„

As we see, the calculations here gave a little more than 100 per cent., which is often the case, due to the uncertainty in the determination of the second differential coefficient of the accumulation curve.

It is of interest to see, not only that the curves plotted according to the new method show a good correspondence with each other, as shown above, but also how the figures for analyses which have been calculated from the distribution curves (obtained with the automatic registering apparatus) compare with others obtained by other means. Two different analyses of the same soil exist now, the one by A. Atterberg<sup>1</sup> and the other by Dr. C. Munthe<sup>2</sup> and I have further, as a check, determined the sand content with the Atterberg apparatus.

If we compare these figures we obtain the following :—

	Per Cent.	Mean Per Cent.
Clay (called both by A. and M. "residue")	Atterberg 14'0	12'85
	Munthe 11'7	
Silt . . . . .	Atterberg 19'6	20'4
	Munthe 20'8	
Fine sand . . . . .	Atterberg 21'1	21'9
	Munthe 22'7	
Sand . . . . .	Atterberg 45'3	45'2
	Munthe 42'1	
	Odén 48'3	
		<u>100'35</u>

It has often been pointed out that the figures of the levigation analyses vary frequently according to the temperature and the operator. Despite this, the mean figures for the older analyses show a surprisingly good agreement with those given above, obtained by my method. It should be remembered that a part of the soil had been deposited in a glycerine mixture of a viscosity five times greater than that of water. The utility and correctness of the hydrodynamic equations is thus made obvious, and we may hope that they will in the future replace the empirical formulæ.

## 6. THE DISTRIBUTION CURVES OF SOME ANNUAL LAYERS OF THE SWEDISH STRATIFIED CLAY.

As a second example we may here describe the use of the new method in the study of a geological problem.

<sup>1</sup> Atterberg, *Intern. Mitt. f. Bodenkunde*, 2, 26 (1912).

<sup>2</sup> *Dissertation*, Uppsala, 1915, p. 37.

We have in the Fennoscandian quaternary deposits a remarkably widely-distributed clay showing a clearly marked stratification, which has from olden times been called "the stratified clay".

Formerly, opinions regarding the cause of this stratification diverged widely, until Gerard de Geer, in the early eighties (1882) connected the stratification with the melting of the great northern inland ice, and characterised the different layers as annual deposits from the melted ice from the glaciers of the retreating ice-line. The later development of this theory has, as is well-known, led up to the geochronological researches on the recession of the ice,

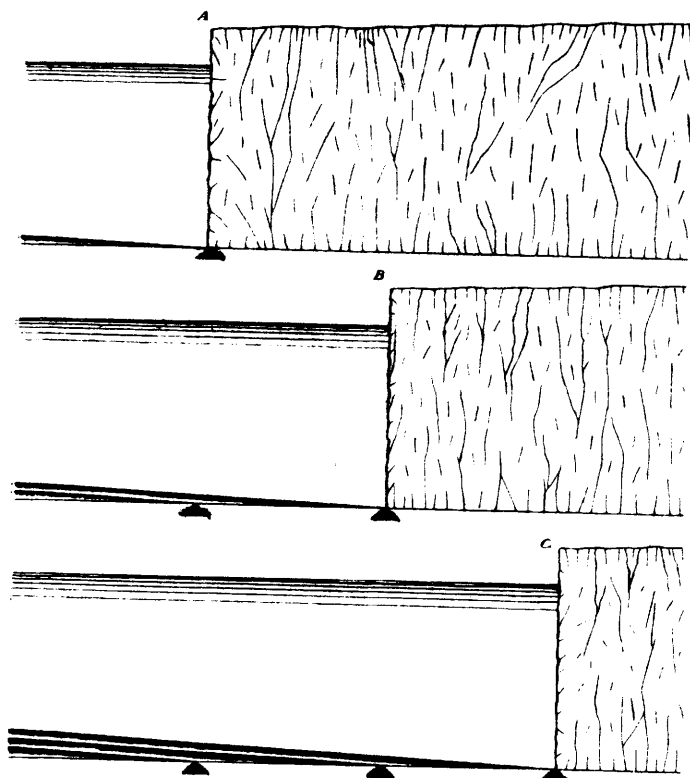


FIG. 13.—Schematic view according to De Geer, showing three successive stages of the ice-edge. The black triangles represent the coarse, stony "äs".

and made it possible to estimate exactly geological time over a period of more than 12,000 years.<sup>1</sup>

This is not the place to give even the outlines of de Geer's magnificent researches. Only as much as is necessary to understand the following mechanical analyses will be reviewed, with the help of two of de Geer's diagrammatic figures (see Figs. 12 and 13). At the time of the melting of the great inland ice, Fennoscandia was covered by an Arctic sea, though poor in salt content, into which large masses of sand, morain, and clay, formed by glacier-erosion, were carried by the glacier streams.

<sup>1</sup> G. de Geer, *Geochronologie der letzten 12,000 Jahre Geol. Rundsch.*, 1912, p. 457.



As far as we can judge, these streams must have flowed in tunnels under the inland ice. The larger stones and blocks were deposited at the gates of the glaziers or near the edge of the ice, and remain still as the mounds (åsar) characteristic for Sweden.

The nearer the ice edge the deposition occurred, the larger and more quickly falling were the particles deposited: while the particles which, on account of the low salt-content of the water, were held in suspension for a long time, were deposited further out over a large surface. As the heaviest gradually subsided, the clay became finer and finer, the farther it was from the ice edge. Since the melting was strongest during the summer, and produced streams of melted ice more rapid, and of greater carrying-power, the structure of the annual deposits shows that the lower part is built up of coarser and lighter-coloured particles of brown to grey clay, which finally merges into a thin, dark, nearly black, so-called winter layer.

During the summer the ice-edge retreated a little (according to de Geer, 50 to 350 metres) northwards, while, during the winter it stood still, or even advanced a little. Due to this movement of the ice-edge each newly formed layer of clay came to rest with the edge nearest the ice on sand, while, further back, it covered the layer of the year before.

The stratified clays resemble in structure fish scales in the respect that each new deposit on the side towards the ice-edge projects a little over the one beneath, the overlap giving the distance that the ice had retreated during the year, and the breadth of ground laid free. This is shown diagrammatically in de Geer's Figs. 12 and 13. A vertical section of the clay, therefore (*cf.* Figs. 14 and 15), shows layers that become thinner and thinner the higher in the section they lie, corresponding to the northward movement of the ice-edge.

De Geer's explanation of the stratification obtained strong corroboration through A. G. Högbom's<sup>1</sup> chemical researches on a calcareous stratified clay. It was shown that the light (grey) "summer deposits" were appreciably richer in lime than the dark (black) winter layers.

The cause of this must be sought in the quicker and more complete solution of the lime during the long time of suspension of the particles, and also in the greater solubility resulting from the larger surface-area due to the greater degree of fineness. Extraction of carbonate of lime during water suspension was also experimentally demonstrated by Högbom.

But the mechanical constitution, too, of the layers, as shown by the distribution curves, must reveal their history. As little work of this nature has been done on the quaternary clays, I have investigated some of these clay-strata.

The thickness of the strata varies a great deal. The lower strata may show a thickness of several decimetres, but this diminishes as we go higher until the thickness is not more than a millimetre, or even a fraction thereof. Higher up the streakiness disappears, and the clay takes a homogeneous grey-brown tone, with here and there a striated appearance. This brown-grey clay merges higher up into the postglacial overlying clay strata, deposited in the Ancylus Lake and the Litorine Sea. Between these two occurs in some places the so-called mottled zone with small lime concretions.

The samples investigated by me were taken from a clay quarry at the St. Erik brickworks, about a kilometre North-West of Upsala. (The

<sup>1</sup> *Trans. of the Geological Soc.*, Stockholm, **11**, 265 (1889); **14**, 291 (1892).

appearance is shown in Fig. 14). Reckoning from the bottom up, the following layers were investigated :—

- A. A 5 cm. summer deposit.
- B. " 4 " " "
- C. " 2 " " "
- D. An area where the striæ were to be measured in mm. (Summer and winter layers.)
- E. An area where the striation could scarcely be distinguished, but where the clay certainly did not belong to the "stained zone".
- F. The over-lying, very thick, *Ancylus* clay, taken just over the mottled zone. (Fig. 17.)



FIG. 14.

The preparatory treatment of all the samples was carried out according to the standard method published by me.<sup>1</sup> A detailed report of the figures of the original determinations would take too much space, I content myself, therefore, with giving graphically in Figs. 16 to 20. the course of the distribution curves.

In setting out the results in Fig 16, the smallest particles ( $<0.6\mu$ ) are shown to the left as a square, proportional to the weight. The coarser particles showing, in places, to the right clearly do not belong to the true clay deposit, but are to be regarded as sandy impurities, dropped by drifting icebergs.

<sup>1</sup> *Bull. Geol. Institut. of Upsala*, 16, 125 (1919); *Intern. Mitt. f. Bodenkunde*, 10, 320 (1920).

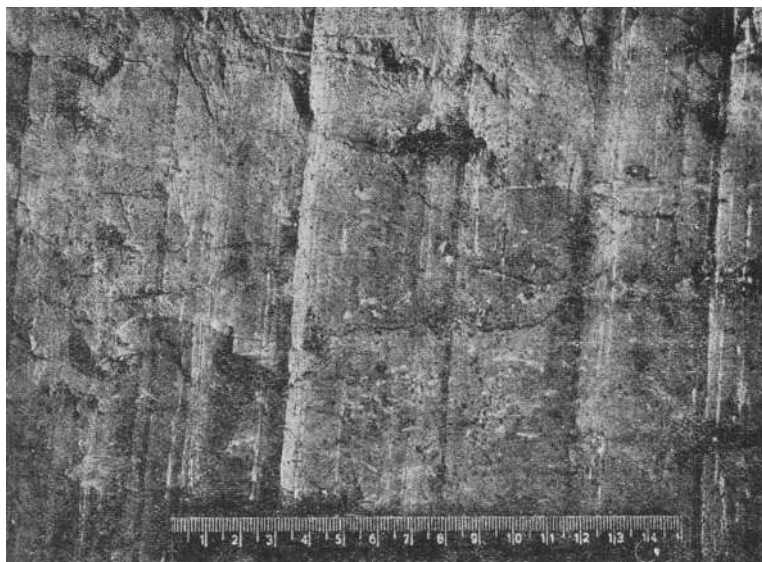


FIG. 15.

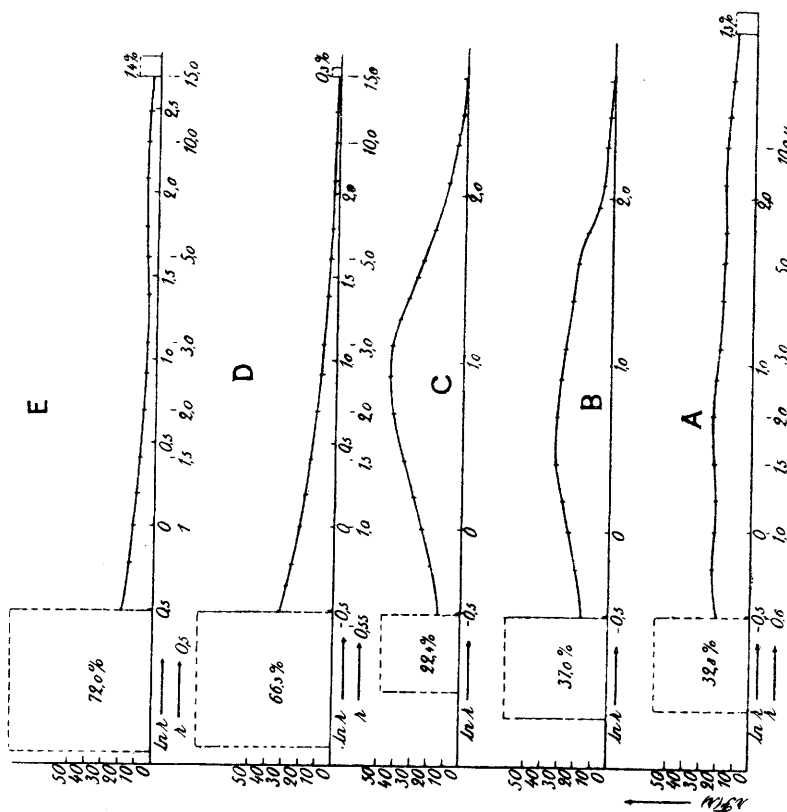


FIG. 16.

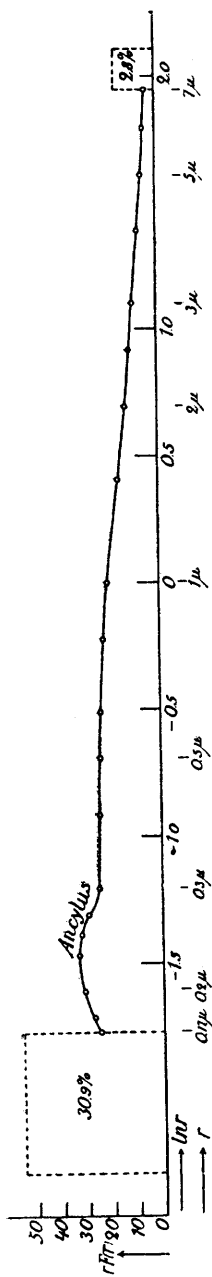


FIG. 17.

If we disregard these irregularities, the samples in Fig. 16 show the increase in coarser particles with increase of thickness of the layer, just as the de Geer theory requires.

The experiment with the Ancyclus clay was allowed to proceed for a much longer time, and the curve was continued a long way to the left, as far as an effective radius of 0.17, as shown in Fig. 17.

After this, the mean diameter of the still undeposited residue was determined by the Zsigmondy-Siedentopf method (*cf.* p. 329). The figures are not completely comparable, since the temperature was not quite the same during all the determinations; and further, there is always a fairly large error in such determinations. But if we examine in turn E, D, C, and A, we find a regular increase in the mean size of the grains. It is also of importance to note that the thicker layers likewise contain appreci-

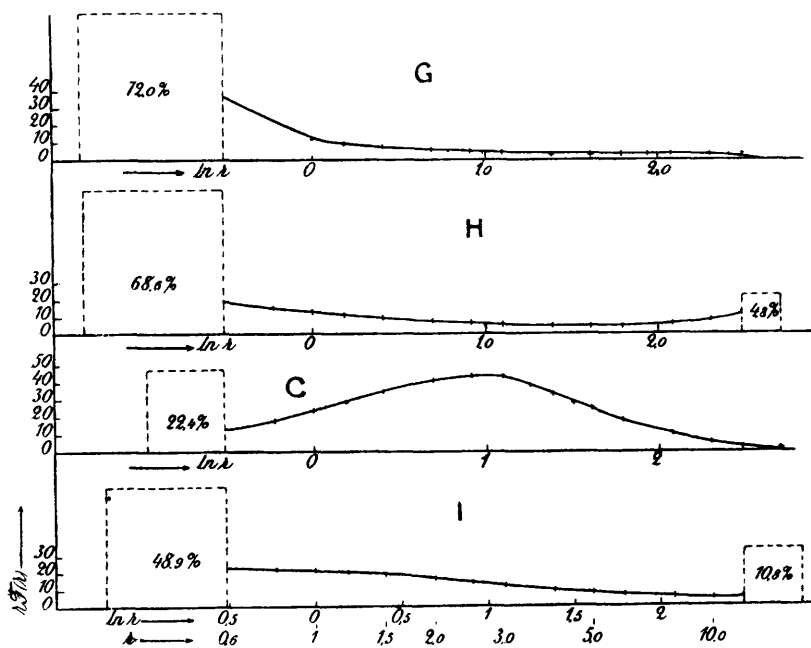


FIG. 18.

able quantities of quite fine particles, which have clearly been deposited together with the coarser grains.

Comparative investigations of "winter" and "summer" deposits were then carried out.

G. A millimetre thick black winter deposit overlying a 1 cm. thick summer deposit.

H. } The winter deposits both overlying and underlying the 2 cm.  
I. } summer deposit C.

The distribution curves for these clays are given in Fig. 18, and clearly show the finer structure of the winter deposit. The appearance of G shows good agreement with the sample E, from the finest deposits, which was to be expected, since the latter consists of both winter and summer deposit.

Further corroboration is furnished by the distribution curves for the samples in Fig. 19.

K. A winter deposit overlying a 4 cm. thick summer deposit.

L. } The winter deposits both overlying and underlying the 5 cm.

M. } thick summer deposit B.

If the winter deposits be compared with each other, they too show a regular decrease in the amount of fine material with increasing depth. The sample, H, certainly is an exception, but this contains considerable quantities (10 per cent.) of coarse sand as impurity.

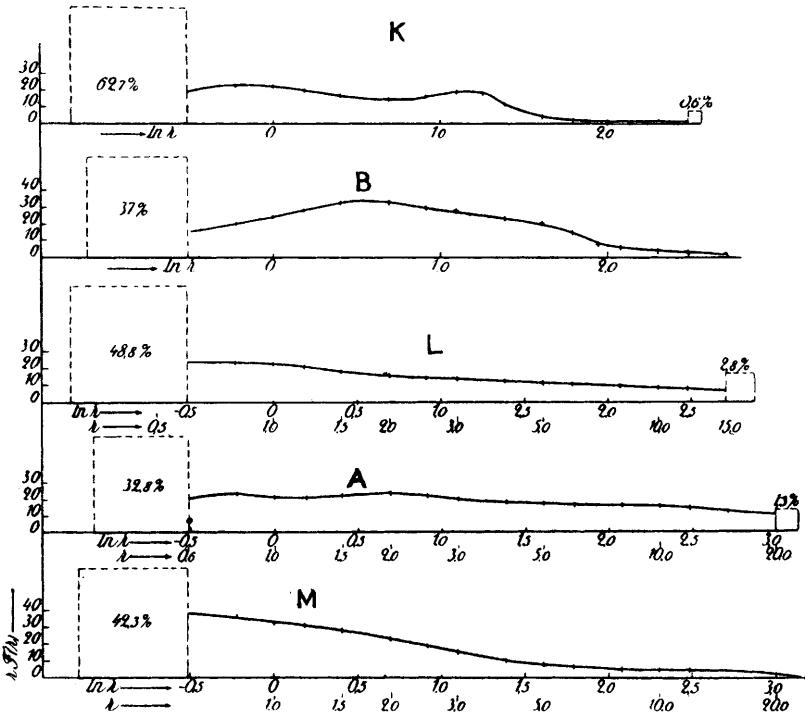


FIG. 19.

With increasing thickness of the layers, the differences between the summer and winter deposits become less marked, as is shown by a comparison of Figs. 18 and 19. This is appreciable, indeed, to the naked eye, since the colour difference between the black winter deposits and the more grey-brown summer deposits give place to a more monotonous grey-brown.

The colour, therefore, is dependent on the size of the particles, and perhaps also on changing chemical constitution dependent thereon.