Magnetic Spherules in Deep-sea Deposits

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FEW OF THE COMPONENTS entering into the sediments covering the ocean floor have attracted so much attention as the magnetic spherules first described in 1876 by Sir John Murray. Struck by the resemblance of their surface structure to that of iron meteorites, Murray called them "cosmic spherules." The number he was able to extract from one quart of the deposit, by means of a small magnet, varied from 20 to 30 in samples of Red Clay, whereas in the same quantity of Globigerina Ooze only one or two were found (Murray, 1876).

Together with A. F. Renard, Murray later published (1897) a more detailed description of the spherules in volume 4 of the "Challenger" Reports, *Deep Sea Deposits.* The higher numbers found in the Red Clay are ascribed to its much lower rate of sedimentation.

Hoping that the method of counting the magnetic spherules present in different kinds of deposits may afford a solution to the difficult problem of determining the rate of deep-sea sedimentation, one of us decided to include in the equipment of the Swedish Deep-sea Expedition (1947–48) special corers of wider diameter than those otherwise used from the "Albatross," viz., with an internal diameter of 90 mm. instead of the usual 46 mm. For various technical reasons this widebore corer was not used until we reached the western Pacific Ocean (Pettersson, 1956). Owing to the higher resistance offered by the

sediment to a thick coring tube, and owing to the necessity of avoiding too heavy a strain on the steel cable used when coring, the length of the thick corer had to be limited to only 6 metres, as compared to the 15 to 20 metres of the narrower coring tubes. The length of the thick cores raised was, therefore, in general only 5 to *5Y2* metres. On the other hand, from a section taken from a thick core about four times more material was obtained than from a narrow core of the same length. In all, a dozen of such thick cores were raised from the cruise with the "Albatross" through three oceans.

Owing to more pressing work on other material collected during the cruise, the working up of the thick cores for magnetic spherules had to be postponed for several years after our return from the expedition. However, in the meanwhile a young technician, T. Laevastu, then in the employ of the Oceanographic Institute in Göteborg, was charged with carrying out preliminary extraction experiments using parts of a narrower core of Red Clay raised from the central Pacific Ocean. For this work an electromagnetic extractor of high efficiency had been obtained from the well-known New York firm of Frantz. Portions of the sediment suspended in water were passed through this extractor. Already in the preliminary experiments with this instrument its great superiority over the primitive method for extraction used by Murray was apparent, the number of spherules extracted from 1 kg. of Red Clay varying between a few hundred and a couple of thousands. Attempts to estimate the efficiency of the method were also carried out by adding to sediment already extracted a counted number of artificial magnetic spherules, made from iron wire with the oxygen flame, having slightly larger dimensions though than the

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natural magnetic spherules found in the sediment. The yield was found to exceed 90 per cent. The results from the preliminary investigation, including a graph showing the sizedistribution of the natural spherules, has been published by Laevastu together with Prof. O. Mellis of Stockholm (1955), who had given him valuable help in the microscopical study of the spherules,

Our first objective when starting our work in November, 1954, was to perfect the method of extraction, involving also the preparation of the sediment samples obtained from the cores. The details of this preparatory work, carried out in the Oceanographic Institute with the excellent help of Mrs. Karin Romlin, technical assistant, will be given in a future publication by one of *us.* It will suffice to mention here that we found it necessary to pass an aqeous suspension of a quantity of the sediment, weighing from 200 to 700 grams (with the narrow corer the sample weight varied between 20 and 300 gr.), at least three times through the extractor. From the magnetic particles thus obtained, which comprised also a large proportion of nonspherical particles of terrestrial origin (largely magnetite), those with strong magnetic properties were separated out by means of a small electromagnet and then passed through sieves of different mesh, separating out the three size-classes: coarser than 60 μ , 60 μ to 30 μ , and less than 30 μ in diameter (see Figs. 1 and 2). The products from this fractionation were then mounted between glass discs in a manner suitable for counting under the microscope. This latter operation was made by one of us in Stockholm, more recently assisted by fil. kand M. Nilsson.

The operations required for a detailed examination of the spherules, like polishing, X-ray examination, etc., were carried out in the Mineralogical Institution of Stockholms Högskola (Fredriksson, 1956). To its director, Professor S. Gavelin, we are much obliged for his support and advice. We are also indebted to Professor F. Hecht of the II Chemisches Institut der Universität Wien, for kindly carrying out for us micro-analyses for nickel and cobalt on some of the spherules submitted to him. His results proved nickel to be definitely present in a percentage varying between 6 per cent and 15 per cent of the iron content. Recently Dr. A. Smales at Harwell kindly investigated samples of deep-sea spherules for nickel and cobalt by means of neutron-activation in the pile.

In order to get comparable results from one sample to another, we found it advisable to concentrate the counts on black spherules of a diameter exceeding 30 μ . The spherules smaller than 30 μ are difficult to count and easy to overlook. Their contribution to the total weight of the spherules appears insignificant. Spherules of the greatest size, i.e., from 60 μ to 250 μ were also counted. Their contribution to the total number of spherules is not great, but, owing to their large size, their contribution to the total weight is considerable.

The following table gives a survey of the cores from which we have extracted and counted spherules.

COMMENTS

CORE 71. This core is of special interest, as it was raised from the vicinity of Challenger Station 274 (S 07° 25' W 151° 15') where the depth was 2,750 fathoms or 5,030 m. The sediment in the surface has been characterized by Murray and Renard (1897) as Radiolarian Ooze with 3.89 per cent $CaCO₃$. The core raised from the "Albatross" has a total length of nearly 10 m., from which sections of 26 to 76 em. in length were taken.

The number of spherules per kg. of sediment, free from salts and lime, varied from a maximum of 1,400 near the surface to a minimum of slightly more than 100 per kg. A second maximum of 600 spherules per kg. appears at the 10 metre level. Control counts made on halves of the section, cut lengthwise, gave fairly large variations in the proportion

TABLE 1

* Separate sample from the surface.

of 1:2. The average number of spherules per kg. for the whole core is about 300 (see the curve in Fig. 3).

According to radium measurements, made by Kröll (1955), in the sediment where cores 71 and 72 were taken, the rate of sedimentation is taken to be between 1 and 2 mm. in 1,000 years.

CORE 72. This core was raised from the immediate vicinity of Core 71, where the sediment had the same character. Its uppermost 3 metres, out of a total length of 14 metres, have been examined for spherules by Laevastu and Mellis (4). In Figure 4 the results from their counts are reproduced from their original paper. In Figure 3 the number of spherules is set out for the same levels in which Core 71 has been investigated by us. The figures, however, have been reduced to 50 per cent to be in conformity with our counts, since Laevastu and Mellis included spherules from diameters 10 μ upwards in their counts, whereas our counts in Core 71 are limited to diameters of from 30 μ upwards. According to Laevastu and Mellis the spherules of less than 30 μ in diameter made up less than one half of the total number.

CORE 90. This is a thick core, 90 mm. in diameter. Its total length is, therefore, very moderate, or only 5 metres. In its upper part the content of lime is low, less than 1 per cent. Below the 190 cm. level the content of carbonates increases abruptly to about 30 per cent, rising still further down to more than

80 per cent. The number of spherules varies greatly. From about 3,300 per kg. near the surface, a maximum of 5,000 is reached between 80 and 90 cm. Below the 110 cm. level the number of spherules per kg. of salt- and carbonate-free sediment is less than '1,000 (minimum 140), with the exception of a secon dary maximum of 1,300 in a depth of about 280 cm.

Two parallel series of samples from this core were extracted. The results obtained from the first five samples in the first series (see full drawn curve in Fig. 5) were much lower than those from the second series. This is probably due to the fact that the technique of extraction had not been fully developed at that time, so that a certain number of the spherules may have become crushed in the extractor and thus escaped the counting.

In certain samples it proved very difficult to disting uish between different types of spherules, of which the black shiny ones were primarily counted. This difficulty may explain the strong variations found by control counts from identical levels, especially as regards the sample between 63 and 80 cm. In the diagram in Figure 5 these variations are indicated by horizontal lines, uniting the highest and the lowest values found at the level in question.

 $CORE 90 B.$ This was a narrow "pilot core" taken from the same vicinity as Core 90. The amounts of spherules in the lime-free portions in the uppermost $4-12$ cm. and $0-6.5$ cm. of Cores 90 Band 90 respectively were found to

FIG. 1. Spherules from Core 90, about 30 to 60 microns in diameter.

be practically the same. The carbonate content was, however, much higher in the uppermost part of Core 90 B, viz., 56 per cent as compared with the corresponding 0-6.5 em. of Core 90.

CORE 92. This core, raised from a depth of nearly 4,000 metres close to the Equator, is also one of large diameter (90 mm.) with a length of only 5 metres. The sediment is a calcareous ooze, its content of $CaCO₃$ varying between 71 and 82 per cent. The uppermost 64 em. of the core were missing, having been lost in transport. The rate of sedimentation can only be tentatively estimated at 50 mm. in 1,000 years.

The number of spherules extracted was very low. Recalculated to 1 kg. of lime- and salt-free sediment, the numbers varied from o in a depth of 201-217 em. to slightly over 200 between 64-81 cm., 231-248 cm. and 275-293 em. The average for the whole core was 80 spherules per kg. of sediment. (See diagram in Figure 6.)

CORE 133. This core, the only thick core (90 mm.) raised from the central part of the Indian Ocean at a depth of 5,200 m., had a total length of 326 cm., from which the uppermost 3 em. were missing. Down to a depth of 200 cm. below the top of the core it had a moderately high content of $CaCO₃$, between 14 and 40 per cent, whereas in the lower parts of the core the lime content was only between 5 and 10 per cent, especially near the lower end where it varied over 45 em. between 5 and 5.6 per cent. The number of cosmic spherules extracted from the uppermost 3-18 em. was high, viz., nearly 700 per kg., whereas in the lower parts, at depths between 50 and

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220 em., the numbers were low, rising again to somewhat higher values below 220 cm. The average number for the whole core was 130 spherules per kg., whereas between 52 and 220 em. it was only 30, compared to the average for the uppermost 50 cm. of 500 per $kg.$ (Fig. 7).

To ascribe these very remarkable variations in the numbers of the spherules only, or even mainly, to changes in the rate of sedimentation seems unwarranted. That even in the uppermost layers the rate of sedimentation was higher than in the central Pacific Ocean is indicated by radium measurements in a neighbouring short pilot core, Number 133 B, which contained on an average 11 units of the 12th decimal place of Ragr/gr, compared to from 40 to 50 units in Red Clay from the central Pacific. Thus the rate of sedimentation can be estimated for the upper parts of Core 133 at 10 mm. in 1,000 years.

CORE 187. This core, raised from a depth of 2,500 metres in the eastern Mediterranean southwest of Cyprus, was a narrow one and had a total length of *9Y2* metres. Hence fairly long sections of 30 to 70 cm. had to be used for extracting the spherules. The lime content was moderately high, varying between 20 and 40 per cent of $CaCO₃$. The number of spherules per kg. of lime- and salt-free sediment varied considerably along the length of the core, from 190 near the surface to an absolute maximum of $1,130$ in the section $165-235$ cm., declining from there, both upwards and downwards, to 5 and 10 per cent respectively of the maximum value.

At another maximum, in section 438-506 cm., 491 spherules were found per kg. of sediment. In the very lowest parts of the core, 860-950 em., only 38 and 49 spherules respectively were found. It must, however, be emphasized that where control samples from

FIG. 3. Number of spherules per kg. of dry substance from Core 71.

the same levels were taken, the values varied considerably . *inter se.* The average for the whole core is 190 spherules per kg.

Regarding the rate of sedimentation in this core vety little is known. O. Mellis (1954), from a study of the volcanic ash horizons present in cores from the eastern Mediterranean, has identified a layer of volcanic ash in Core 187, situated at a depth of 40 em. below the top of the core. This layer probably owes its origin to the catastrophic outbreak of the island volcano Santorin, which occurred some time between 1800 and 1500 B.C., i.e., about 3,700 years ago. This assumption would make the rate of sedimentation in the upper layers of the core about 10 em. in 1,000 years, leaving aside possible disturbances in the sedimentation due to slumping, etc.

Regarding the two maxima found in Core 187, it should be noted that the spherules they contain were especially difficult to identify, resembling some of the samples in Core 90. The values given here must, therefore, be stated with due reserve and have only been .indicared in the curve in Figure 8. Nevertheless there is little doubt that the maxima are real, even though the numerical finds are

somewhat uncertain. Attempts to find such maxima in other cores from the vicinity are at present being undertaken.

Considering the abrupt decrease in the number of spherules on both sides of the principal maximum, one is tempted to assume that at the time when the maximum occurred one or several meteoritic showers fell upon the eastern Mediterranean. An alternative explanation, that at the time when the maximum occurred there can have occurred an abnormally low rate of sedimentation, appears less probable.

It would be most interesting to investigate other cores, preferably of larger diameter, taken in the vicinity of Core 187.

MAGNETIC SPHERULES FROM THE ATMOSPHERE

The problem of finding by direct observation the amount of magnetic particles settling

FIG. 4. Number of spherules per kg. of salt- and carbonate-free sediment from Cores 71 and 72.

FIG. 5. Number of spherules per kg . of salt- and carbonate-free sediment from Core 90.

on the Earth's surface has repeatedly been attacked by various investigators, mainly in the United Srates of America. The method adopted has generally been that of introducing magnetic "collectors" into the water spouts carrying rainwater from extensive roofs. The number of magnetic spherules accumulated in this manner has been counted with results which are surprisingly large. Thus Warren J. Thomsen (1953) finds the weight of such magnetic spheres, from observations made at Iowa City, to correspond to a total mass of 2,000,000 tons per year for the whole Earth. Still higher values have been reported by W. D. Crozier (1955) working in New Mexico, viz., 35,000 tons per diem during

August 1955, or nearly 13 million tons for the whole year. These figures are several thousand times higher than those obtained from our counts of the spherules in deep-sea deposits. However, neither of the authors mentioned could find any trace of nickel in the magnetic spheres analyzed.

We have tried to collect magnetic spherules by means of magnetic collectors introduced into water spouts mounted at Bornö Station in the Gullmar Fjord, on the west coast of Sweden, and also from the roof of the Oceanographic Institute in Göteborg. At the latter locality we found the number of magnetic spherules collected to be very high, whereas at Bornö the number was considerably less but

FIG. 6. Number of spherules per kg. of salr- and carbonate-free sediment from Core 92.

still higher than the numbers found in deepsea deposits. No nickel was found to be present in these spherules.

We infer, therefore, that most of the spherules thus collected are of terrestrial origin and are artifacts from industrial plants and welding operations, which can be carried over large distances by wind.

Hoppe and Zimmerman (1954), collecting magnetic spherules at Jena and other localities in West Germany, have arrived at the same conclusion and consider it impossible to avoid such contamination in localities in or near industrial districts.

Based on these results we have planned to collect magnetic particles from the atmosphere on ocean islands situated far from any industrial plants and as remote as possible from the shipping lanes across the sea. Such measurements, which we hope to organize on Pacific Ocean islands during the Geophysical Year, may be expected to afford evidence of

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true extraterrestrial spherules settling over the ocean surface and thus complement our studies of the magnetic spherules in deep-sea deposits. Calling attention to the interesting results 'from the attempts at a magnetic drag over the ocean bottom from the "Galathea" Expedition, we consider the evidence in favour of the magnetic particles reported by A. Bruun and his co-workers (1955) as being largely of cosmic origin to be very strong. On the other hand this method of using a superficial collector can only be expected to bring up extraterrestrial particles of *recent* origin. Moreover, a certain risk of contamination with spurious spherules from coal- or oil-driven ships passing near the course of the "Galathea" cannot be altogether excluded. No such risk's are involved when extracting magnetic spherules from undisturbed long cores provided one gets below the uppermost few centimeters of sediment. This method alone seems to allow of definite conclusions regarding the frequency of meteoritic falls and their variations in the remote past.

Quite recently the accruement of meteoritic fragments and dust to the surface of the Earth and its oceans has attracted the interest of meteorologists and climatologists.

Mainly through the work of Bowen in Australia (1953) and of his co-workers in

FIG. 7. Number of spherules per kg. of salt- and carbonate-free sediment from Core 133.

FIG. 8. Number of spherules per kg. of salt- and carbonate-free sediment from Core 187.

different parts of the world (Bracewell, 1954) strong evidence has been produced for the influence of cosmic dust as condensation nuclei for raindrops. According to these finds there occur distinct maxima of precipitation on certain dates like January 12, 22, and 31 to February 1. These peaks of rainfall occur 29 to 30 days after the Earth has passed through meteor showers. According to Bowen the dust into which these meteors are dispersed at great heights in the atmosphere takes about 30 days to descend to the upper troposphere, where a condensation of waterdrops is produced which ultimately reach the Earth as rain. Through this condensation a depletion of water vapour in the upper atmosphere is occasioned, decreasing the "greenhouse" effect on the radiation balance.

It has even been surmised that an abnormal increase in the incidence of meteors may give rise, by a trigger action, to a deterioration of climate and possibly release a glacial epoch .

No wonder that students of the atmosphere and its radiation balance are taking a growing interest in meteoric dust.

THE ANNUAL ACCRUEMENT OF COSMIC SPHERULES

In trying to work out the total weight of the cosmic spherules settling on to the surface of our planet the numbers of spherules found in the cores investigated can be used, converting the numbers of spherules per kg. into weights. But already this conversion from numbers into weights implies an element of uncertainty.

We have counted all spherules of a diameter exceeding 30 μ and then computed the total number accruing to the Earth for one year. These values are set out in column 7 of Table 3. In converting numbers into weights we have assumed a specific weight of 5.0 and a diameter of 40 μ , assuming that all the spherules belong to the size-class $30-60 \mu$. When taking the total counted number instead of the number of the size-class 30–60 μ , we add about 10 per cent as a correction for the weight of the spherules of a diameter less than 30 μ . In this way the values set out in column 8 have been obtained. Finally, column 9 shows values of the total accruement of black spherules of all sizes, the value being three times higher than those in column 8. The factor 3 we obtain by considering that the number of spherules between 30 and 60 μ is 8 times higher than the number of spherules exceeding 60 μ in diameter. (See Table 2.) The weight of the spherules of 30-60 μ is assumed to be equal to the weight of the fraction 60–125 μ and 125–250 μ .

TABLE 2

CORE NO.		TOTAL NUMBERS OF SPHERULES COUNTED	NUMBERS $30-60 \mu$		
	$30 - 60 \mu$	$> 60 \mu$	NUMBERS 60μ		
71	349 2,634	69			
90		303	9 5 11		
92	115	23			
133	326	29			
187	507	58	9		
	3,931	482	8		

Another still greater uncertainty enters into the calculation owing to the imperfectly known rate of sedimentation. Here one has to use approximate values involving possible errors by the factor of 2 to 5, **in** some cases even more.

The following values worked out at the present stage of our investigation must be given, therefore, with due reservation. Compared to earlier figures given by other investigators our estimates are very moderate but are necessarily subject to an revision when more extensive investigations now pending have been carried out.

cores. **In** general one gains the impression from these tables that the meteor frequency has been considerably above the average **in** recent times, as is also indicated in most of the frequency diagrams. However, we must emphasize the great difficulties inherent in the method of sampling. First, the surface layer may be missing, a layer which may quite well represent a sedimentation time of tens of thousands of years. Further, owing to different rates of sedimentation, a whole core like No. 187 probably represents a sedimentation time of less than 100,000 years, which corresponds only to a couple of em. in Core 7l. Hence, it is desirable to investigate a series of cores from different localities in order to arrive at really dependable values for the accruement of spherules and by this means to interrelate the results obtained from different

Apart from the results given **in** the diagrams and **in** Table 3 we also have values from four other samples, two from the upper layers of two western Mediterranean (Cores 17 and 18), and two from the western Pacific Ocean (Cores 87 and 89). From Cores 17 and 18 the number of spherules per kg. lime- and salt-free sediment is 260 and 150 respectively. Assuming that the frequency of spherules is inversely proportional to the rate of sedimentation at a certain time we can, by comparison with Core 187 from the eastern Mediterranean, form an estimate of the rate of sedimentation **in** the western Mediterranean. We thus find a sedimentation rate of about 80 mm. in 1,000 years for Core 17 and a rate of 130 mm. in 1,000 years for Core 18, which are both quite reasonable figures. For Cores 87 and 89 the numbers of spherules were 1,725 and 1,165 respectively, and through a comparison with Cores 90 and 90 B we obtain a sedimentation rate of 6 and 8 mm. respectively **in** 1,000 years. These figures are three to four times higher than those found by Kröll (1955) from radium measurements. However, we do not know how far the uppermost samples in different cores represent the same span of time.

SUMMARY

The present paper gives the results from a study of the number of "cosmic spherules" present in sediment cores obtained from great depths in different parts of the ocean.

TABLE 3

						ANNUAL ACCRUEMENT OF BLACK SPHERULES		
Core No.	Depth in cm.	Rate of sedimentation mm/1000y	CaCO ₃ per cent	Span of time in years	Number of $\left \text{diam.}\right.\geq 30\right $ per kg.	Number $(> 30 \mu)$ (10^{15})	Weight in tons $(< 60 \mu)$	Total weight in tons
187	$0 - 39$	100	35	$0.2 - 3,700$	190	4.7	790	2,400
90	$0 - 6.5$	3		$0? - 20,000$	3,350	1.6	265	800
90 B	$4 - 12$	6	56	7,000-20,000	3,350	1.6	270	800
92	$64 - 81*$	50	76	13,000-16,000	220	1.1	190	600
133	$3 - 18$	10	15	3,000-18,000	700	1.0	160	500
71	$0 - 26$	1.5		$0? - 200,000$	1,400	0.31	60	175
72	$0 - 300$							125 [†]

* Section 0-64 cm. lost in transport.
† According to Laevastu and Mellis.

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The number of such spherules obtained by means of a powerful electromagnetic extractor is tens of times greater than the numbers found by Sir John Murray and A. F. Renard by more primitive means.

Spherules in considerable numbers are found also in depths of several metres below the sediment surface, i.e., in sediments already deposited in Tertiary Time. This definitely refutes the hypothesis that meteoritic falls, from which the spherules are assumed to be derived, have been limited to a relatively recent past, the last 25,000 years.

On the other hand there are strong indications that the frequency of such spherules deposited in recent times, say in the last few thousand years, has been higher than in a more remote past.

There are also indications of intermediate maxima which may correspond to a greater frequency of meteoritic falls during certain times covered by our material. A systematic correlation of such maxima *inter se* has not been made.

Comparing the frequency of spherules found in recent times, i.e., near the surface, in certain cores affords means of estimating the approximate rates of sedimentation.

Converting the number of spherules found per kg . of sediment into weights makes it possible to estimate the total accruement of spherules to the whole Earth, the results being a moderate figure of 2,400 metric tons annually (with a possible rise to 5,000 tons), i.e., only a small fraction of figures obtained by earlier investigators from the number of magnetic spherules collected from the atmosphere. Means of obtaining more reliable figures for such spherules by establishing collection stations on mid-ocean islands are indicated.

In cases where the number of spherules from deep -sea cores have been analysed for iron, nickel, and cobalt, the presence of nickel has given a definite proof of the cosmic origin of the deep-sea spherules.

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