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# The nickel content of deep-sea deposits

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ABSTRACT

Determinations of the nickel, manganese and iron present in sediment cores from the Central Pacific Ocean have been made by microchemical methods. The results show (a) that the nickel content is high relative to that of continental rocks and sediments and, (b) that there are considerable variations in the nickel contents with depth below the sediment surface. No correlation with the manganese and iron contents is evident except in cases where a change in the rate of sedimentation appears to have affected all three elements in the same manner.

It is tentatively suggested that this abundance of abyssal nickel may be partly derived from cosmic dust settling over the earth's surface. Arguments for and against this thesis are put forward.

This paper* sets out the results so far obtained in determinations of nickel, manganese and iron in deep sea sediment cores. They have been made partly in the Oceanography Institute, Göteborg and partly in the Geochemical Laboratory of the Swedish Geological Survey (S.G.U.) in Stockholm by Dr. S. LANDERGREN. Although the paper deals primarily with the nickel content which affords points of special interest, the iron and manganese contents are included for purposes of comparison. Iron and manganese are components of red clay and radiolarian ooze but opinions differ regarding their origin and the occurrence of these two elements will be dealt with in a later report [1].

Little has been published on the occurrence of nickel in deep-sea deposits. The earliest reference known to the writers appears in the "Challenger" Reports, vol. 4, Deep-Sea Deposits by Sir JOHN MURRAY and the Rev. A. D. RENARD [2]. Their analyses show only traces of nickel in a few of the sediment samples but in manganese nodules taken from great depth traces of nickel were not unusual. Indications of cobalt were more rare.

The lack of precise data is not surprising considering the low sensitivity of the methods used for determining the nickel content fifty years ago. In the second decade of this century very sensitive microchemical methods for measuring small quantities of nickel by means of colour reactions came into use. By means of colourimetric determinations on an organic nickel compound with the Pulfrich spectrophotometer it is fairly easy to measure quantities of the element as low as a few parts per million.

These modern methods have so far been used chiefly for measuring the nickel in continental rocks and sediments. The only nickel determinations on deep-sea deposits recorded during the present century are those by CLARKE and STEIGER [3]. In a composite sample made up from 51 varieties of red clay from different localities the content was found to be 0·032 % of nickel oxide or 0·025 % of the element. This figure is considerably higher than the average value of 0·008 % Ni given by SANDELL and GOLDICH [4] for the uppermost crust of the earth.

* A full report will appear in the series "Report of the Swedish Deep-Sea Expedition" to be published by the Royal Society of Göteborg.

No explanation has so far been suggested for the origin of this abyssal nickel. A fairly constant supply of nickel should give high values in sediments, such as red clay and radiolarian ooze, which have a slow rate of accumulation. Four sources of nickel seem worthy of consideration:

1. *Directly from the land area*—Terrestrial material in the form of wind- and / or water-borne particles of very small size, such as the finest clay particles from rivers, desert sand is carried seawards by the winds, or ash from continental or island volcanoes. The nickel content of this material is not likely to be appreciably higher than that of similar material hitherto analysed and therefore a much higher content in deep-sea deposits presupposes a concentration *in situ* by some submarine process by which other components are dissolved leaving the nickel in a more concentrated state.

2. *From the sea water*—The nickel content in ocean water according to ERNST and HOERMANN [5] is extremely low, being in the order of $0{\cdot}1\ \gamma$ per litre. This figure is approximately 500 times less than the average for iron. The extraction of nickel by marine organisms has not been observed.

3. *From magma, erupted onto the sea bottom*—If this magma contains a higher concentration of nickel than that found in ordinary rocks or sediments on the continents then this may be a sufficient source of the element. The numerous volcanic islands of the three oceans, and especially of the Pacific, establish that volcanic eruptions on a large scale occur in great ocean depths. Submarine lava beds of considerable size were observed on the "Albatross" cruise both in the Pacific and Indian Oceans [6].

4. *Extra-terrestrial sources*—Meteoric dust settling from shooting stars and dispersed in the higher atmosphere, or unchanged minute particles from interplanetary space; the micro-meteorites of ÖPIK and WHIPPLE [7] and [8], provide a source of nickel. The proportion of extra-terrestrial nickel in the deep-sea deposits can be expected to vary, in general, in inverse ratio to the rate of sedimentation and therefore to rise to a maximum in abyssal sediments having a very low rate of accumulation. On the other hand, such an accrument of cosmic nickel is likely to show local variations due to the incidence of exceptionally heavy meteor showers. (Compare NININGER [9].)

If an extra-terrestrial origin for a considerable, if not the major part of the nickel in red clay and radiolarian ooze is admitted, then large variations of the nickel content along the length of a deep-sea core may be interpreted as indicating significant variations, either in the rate of sedimentation or in the rate of accumulation of meteoric dust and micro-meteorites or perhaps both. These variations would thus depend on the cosmic history of our earth or of the solar system to which it belongs.

This latter suggestion gives a particular interest to the study of abyssal nickel. It seems possible that the rate of accumulation of the supposedly cosmic material, as far as can be determined from measurements of the absolute rate of sedimentation, may afford a check on the accuracy of the conclusions reached by astronomers from meteor counts and estimates of the quantity of cosmic dust in interplanetary space based on the strength of the zodiacal light, a problem in which students of solar radiation and its variations with time are greatly interested [10].

## Results of the Analyses

Sandells [11] method was used for the nickel determinations. It is based on the brown colour obtained when dimethyl-glyoxime is added to an alkaline solution containing nickel which has been treated with an oxidizing agent. Large quantities of aluminium interfere with the colour reaction and manganese, readily oxidized to its dioxide, also gives a brown colour. Iron gives a yellow colour with the citrate which was used. Due to the above complications it was necessary to extract the nickel by utilizing the solubility of the complex nickel-dimethylglyoxime in chloroform in a slightly alkaline solution, a method also recommended by Sandell. By this technique it was possible to measure quantities of nickel considerably less than 10 $\gamma$ by means of the Pulfrich photometer when using quantities of the sample which contained amounts of nickel between 5 $\gamma$ and 50 $\gamma$.

Identical solutions from the samples were used for determining the iron and manganese contents. Iron was estimated by utilizing the red colour produced when potassium thiocyanate is added to an acid solution which has been treated with an oxidizing agent. Oxidation to the permanganate with ammonium persulphate was used for the manganese determinations. This method has been described in detail by F. Nydahl [12].

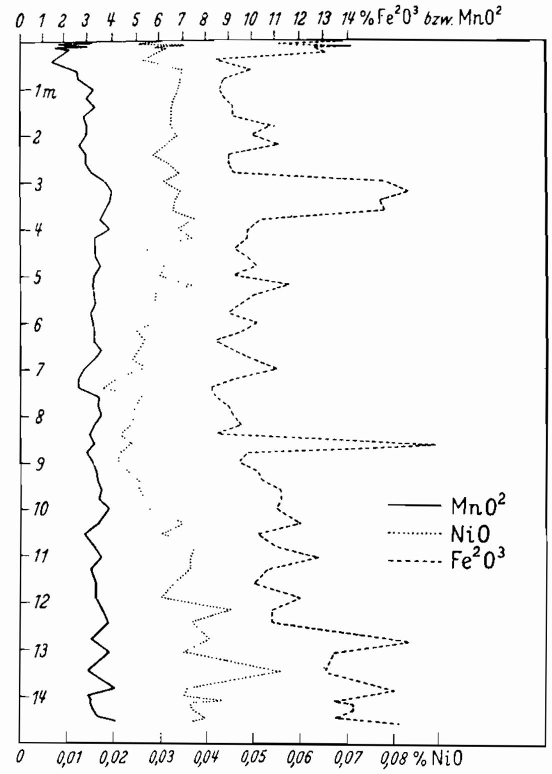

Fig. 1. Core 72.

The first determinations were made on two long cores of red clay taken close together at Albatross Station 105 (Lat. S 7° 38′, Long. W 152° 53′, depth 5000 m) about 1000 nautical miles north of Tahiti. A cursory examination, made on board the ship, showed the deep chocolate brown sediment to be homogeneous. The content of siliceous and calcareous organic remains was low.

The results, which are shown in Fig. 1, varied from a minimum for NiO of 0·041 % at 742 cm below the top of the core to a maximum of 0·089 % at 862 cm. Five distinct maxima of NiO occurred at sediment depths of 322, 862, 1280, 1387, and 1457 cm.

The iron content varied between 3·60 % and 11·17 % $Fe_2O_3$ and the manganese between 1·38 % and 4·14 % $MnO_2$. Apart from a minimum for all three oxides at 742 cm no correlation between the variations is evident from the curves.

The average values from the 77 samples were: NiO—0·056 %; $Fe_2O_3$—6·21 %; $MnO_2$—3·13 %. The average nickel value is almost twice as high as that found by

CLARKE and STEIGER for their composite sample of red clay and the maximum value nearly three times their average.

After publishing these results in a letter to Nature [13] work was resumed in the Autumn of last year on other cores from the central Pacific Ocean. The following is a summary of the results:

*Core 69*—Albatross Station 102 (Lat. S 13° 25′, Long. W 143° 30′, depth 4625 m). A red clay rich in phillipsite crystals and homogeneous in the uppermost 3 m with an increase in lime in the lower parts. Average contents were: NiO—0·029 %; $Fe_2O_3$ 7·83 %; $MnO_2$—2·69 %. At 222 cm below the top of the core there were two maxima of NiO close together, 0·098 % and 0·064 % respectively, with an intermediate minimum in a thin layer apparently of volcanic origin (0·048 % NiO). Below the 300 cm level the sediment becomes richer in lime with a maximum of 60 % whereas the nickel, manganese and iron contents fall off due to the diluting effect of the calcareous remains. Recalculating the values for a sediment free of lime the three curves are found to be independent of each other except for a common minimum at 480 cm.

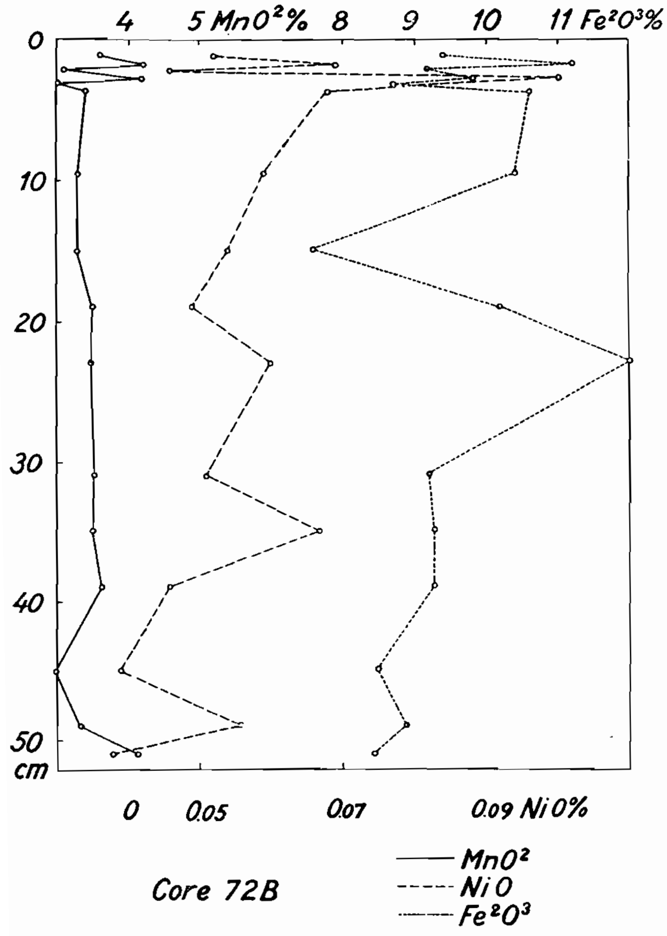


Fig. 2.

*Core 70*—Albatross Station 104 (Lat. S 11° 15′, Long. W 150° 30′, depth 4920 m). A red clay with a low lime content but rich in phillipsite. Average contents were: NiO—0·036 %; $Fe_2O_3$—9·38 %; $MnO_2$—3·24 %. The nickel content was fairly constant with a slight maximum of 0·047 % at 120 cm. The nickel and manganese variations are similar but the iron decreases with depth from a high content in the uppermost portion.

In addition to the long cores described above analyses were carried out on some short cores which were taken by a different technique. In these cores the uppermost surface layer is preserved whereas in the long cores a top layer varying from a few centimetres to ten or more centimetres may be missing. One of these cores, taken at the same locality as core 72 and designated as 72 B, had a length of 56 cm (Fig. 2). The averages from a length of 49 cm were NiO—0·058 %, $Fe_2O_3$—9·54 %, $MnO_2$—3·52 %. Immediately below the sediment surface this core had a maximum of 0·10 % NiO which was the highest observed in any of the cores. On the other hand, short cores taken north of the Equator somewhat further east gave considerably lower values for nickel and manganese. Core 77 B (Fig. 3) (Lat. S 9° 30′, Long. W 148° 40′, depth

5360 m) was poor in lime and rich in radiolarians and had average contents of only 0·009 % Ni, 0·42 % $MnO_2$ and 6·50 % $Fe_2O_3$. The composition of the uppermost 10 cm of the core was extremely variable. The dilution effect due to the silicious remains was evident and the high iron content was remarkable.

Similar results were noted with other short cores, taken between the Equator and Hawaii, which were rich in radiolarians (Fig. 4). Here also low values of nickel and manganese were found.

It is of interest to compare the nickel, manganese and iron contents by calculating the average Mn:Ni and Fe:Ni ratios for each core (Table 2). The Mn/Ni ratio for typical radiolarian ooze is fairly constant between 33 and 41, the same as in cores 72 and 72B, but in cores 69 and 70 it rises as high as 75 and 73.

A few cores which were rich in lime were analysed by MAG. BAUER in the *Oceanographic Institute*, Göteborg. As a result of the dilution by calcareous remains (up to 90 % of the total weight) the nickel content was only a few parts in a hundred thousand and the Mn/Ni ratio was as low as between 13 and 20.

Recalculating the data from these cores for a non-calcareous sediment the nickel and iron values were about as high as those in core 72 but the manganese content is fairly low (three to five times less). The Mn/Ni ratio for red clay is much higher (45–75) and for globigerina ooze much less (13–20). The Fe/Ni ratio was still more variable: from 100 to 250 for red clay; from 190 to 650 for radiolarian ooze; from 68 to 220 for globigerina ooze.

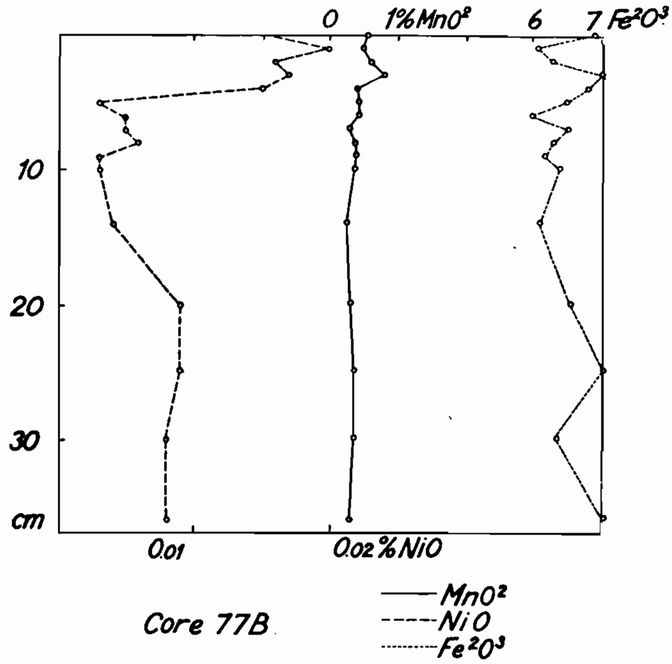


Fig. 3.

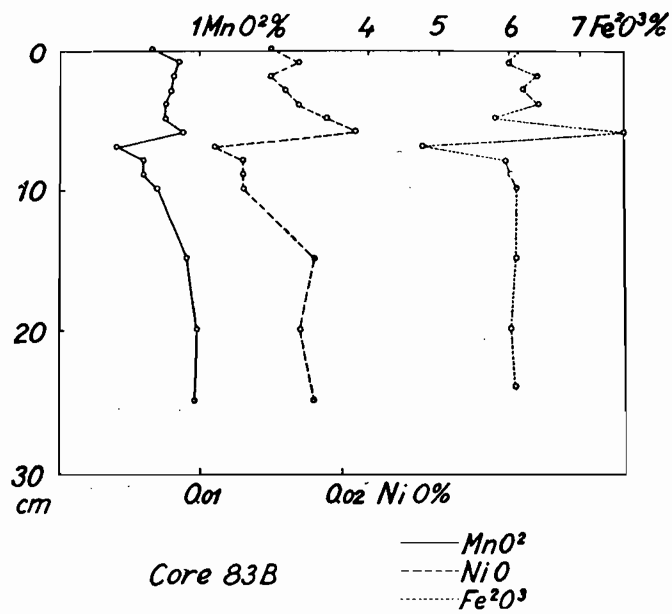


Fig. 4.

Preliminary experiments were carried out to ascertain the nickel content in grains of different size. After the magnetic particles were extracted from the sediment of core 72 between depths of 1360 and 1410 cm a large volume of material was available for further investigations. This residue was submitted to a mechanical separation into fine particles with an equivalent diameter of 0·003 mm and larger particles exceeding 0·006 mm. The results of the determinations on this material was as follows:

| *Particle Size* | NiO % | $MnO_2$ % | $Fe_2O_3$ % |
|---|---|---|---|
| Less than 3 μ . . . . . . | 0·064 | 4·25 | 10·90 |
| Greater than 6 μ . . . . | 0·026 | 1·05 | 3·23 |

One explanation for the concentration of nickel, manganese and iron in the finest particles may be the greater adsorption power of the latter. Another explanation (limited to the nickel and iron content) might be the preponderance of nickel and iron in highly disperse cosmic dust.

In view of the possibility that the nickel in the deep-sea sediments may have its origin in magma extruded over the ocean floor, at the suggestion of the writers, analyses of magmatic material were kindly undertaken by Dr. LANDERGREN. He analysed a number of representative samples for their content of nickel and other elements by means of a precision spectrographic method he has developed. The following were investigated:

(i) A fragment broken from a submarine lava bed in the eastern Pacific Ocean at a depth of nearly 4100 m.

(ii) Two specimens of basaltic rock obtained from the bottom of the Indian Ocean at a depth of nearly 3400 m by the John Murray Expedition. (Given by Dr. J. WISEMAN of the *British Museum of Natural History*, London.)

(iii) Three samples of supramarine basalt from Hawaiian volcanoes. (Given by Dr. J. T. JAGGAR of Honolulu.) The results of these analyses are shown in Table 1. The nickel content varied from 0·005–0·012 % Ni. The submarine lava contained only 0·006 % Ni.

Determinations were also carried out on the nickel content of some basic and ultra-basic rocks from Nukuhiva, Tahiti and Oahu, on the assumption that such rocks, supposedly from deeper layers of the earth's crust, may be fairly rich in nickel. The results, which are shown in the lower part of Table 1, show that in one case (an olivine basalt from Mahaena in Tahiti) a nickel content of 0·13 % NiO was found. In the other rocks of this type the nickel content varied between 0·002 % and 0·018 % NiO.

Summing up the results of the analyses set out in the preceding pages it may be stated that:

1. The nickel content in most of the cores so far investigated was much higher than the average value for continental rocks and sediments with maximum values ten times higher than the continental average. The highest nickel values were found in material with a low rate of sedimentation (in the order of 1 mm in 1000 years). The lower values observed appear largely due to the dilution effect of calcareous or siliceous microfossils. Variations in the nickel content with depth below the sediment surface are conspicuous, especially in long cores of red clay and radiolarian ooze, with maxima two to three times higher than the average value found for the same core.

*Table 1. Spectrographical analyses**

| | Position | Depth m | Ni % |
|---|---|---|---|
| *Submarine material* | | | |
| Lava | 11° 25′ N, 113° 48′ S | 4090 | 0·007 |
| Variolitic basalt | 1° 25′ S, 66° 34′ E | 3385 | 0·005 |
| Basalt with augite and oligoclase | 1° 25′ S, 66° 34′ E | 3385 | 0·01 |
| Variolitic augite basalt | 1° 25′ S, 66° 34′ E | 3385 | 0·006 |
| Hornblende augite dolerite | 1° 25′ S, 66° 34′ E | 3385 | 0·006 |
| Basaltic fragment | 6° 55′ N, 67° 10′ E | 4793 | 0·012 |
| Olivine augite basalt | 8° 35′ N, 94° 10′ E | 2268 | 0·01 |
| *Supramarine material* | | | |
| Basalt | Oahu | | 0·019 |
| Basalt | Hualalai, Hawaii | | 0·022 |
| Basalt | Hualalai, Hawaii | | 0·012 |
| Old pitchstone | Puu Waawaa | | ≤ 0·001 |
| Old pitchstone | Puu Waawaa | | ≤ 0·001 |
| *Microchemical analyses*** | | | |
| Laterite | Pali Oahu | | 0·014 |
| Laterite | Moku au Toto, Nukuhiva | | 0·009 |
| Basalt | Taipi Vai, Nukuhiva | | 0·014 |
| Volcanic Glasses | Nukuhiva | | 0·001 |
| Basaltic lava | Tenua kue Nui, Nukuhiva | | 0·013 |
| Olivine-basalt | Mahaena, Tahiti | | 0.10 |
| Pyroxenite | Mahaena, Tahiti | | 0·014 |

* By Dr. Landergren. ** By Dr. M. Rotschi.

*Table 2*

| Core | NiO % | $F_2O_3$ % | $MnO_2$ % | Mn/Ni | Fe/Ni | Sediment |
|---|---|---|---|---|---|---|
| 69 | 0·029 | 7·83 | 2·69 | 75 | 250 | Red clay |
| 70 | 0·036 | 9·38 | 3·24 | 73 | 235 | Red clay |
| 72 | 0·056 | 6·21 | 3·13 | 45 | 100 | Red clay |
| 77B | 0·009 | 6·51 | 0·42 | 40 | 650 | Rad. ooze |
| 83B | 0·016 | 6·11 | 0·76 | 37 | 330 | Rad. ooze |
| 85B | 0·021 | 5·99 | 1·04 | 41 | 260 | Rad. ooze |
| 86B | 0·022 | 4·69 | 0·87 | 33 | 190 | Rad. ooze |
| 87B | 0·020 | 4·57 | 0·98 | 39 | 200 | Rad. ooze |
| 75 | 0·006 | 0·46 | 0·13 | 17 | 68 | Glob. ooze |
| 93 | 0·008 | 1·04 | 0·12 | 13 | 120 | Glob. ooze |
| 95 | 0·004 | 0·93 | 0·1 | 20 | 220 | Glob. ooze |

2. The iron and manganese contents determined in the same samples also displayed considerable variation with depth. In general, however, these do not run parallel to those for nickel except in some of the short cores (especially manganese). The cause in the few cases where coincident maxima were found for nickel, iron and manganese is probably due to changes in the rate of the sedimentation and dilution effect. The Mn/Ni ratio varied from less than 20 to over 70, with an average of

approximately 45 which in not far from the ratio between the two elements assumed to prevail in seawater. The Fe/Ni quotient varied between 70 and 650 with an average of 200, whereas the ratio in seawater is about 500. Core 93, which was extremely rich in forams, was a rather exceptional case. The Mn/Ni ratio was as low as 13 but the Fe/Ni ratio was 120 and therefore much closer to the average value. Possibly the abundance of calcium of organic origin in the sediment has counteracted the accretion of manganese or has facilitated its disappearance through resolution.

3. Consideration of the above-mentioned sources of nickel as a means of explaining the high nickel content of deep-sea sediments, and especially those having a low rate of accumulation, led the writers to the conclusion that it was improbable that it was due to terrestrial material in the form of wind or waterborne particles because of their original very low nickel content. The same objection applies to a magmatic source from lava ejected over the ocean floor. Both these sources presuppose an extraction process operating in the bottom water, by which the greater part of the matrix becomes dissolved, leaving the nickel in a state of 5- to 10-fold concentration. A magmatic origin would require periodic outbursts of lava especially rich in nickel in order to account for the variation found in the distribution of the element along cores of great length. In view of the present imperfect knowledge of the chemical processes operating on the ocean floor neither hypothesis can be considered impossible. A more detailed study of the content of other elements present in the deposits may yield more conclusive evidence for or against these explanations.

The very low content of the element has already been cited as an objection to the sea-water as a source of the abyssal nickel. On the other hand it must be admitted that the Mn/Ni ratio in some of the cores approaches that assumed to exist in seawater and this lends some support to this theory, for the abyssal manganese is in all probability largely derived by a percipitation and/or adsorption process. The fact that the variations found in the nickel content with depth below the sediment surface do not show any close relationship with those of manganese would seem to refute this explanation. In any case, the adsorption or precipitation process leading to the deposition of nickel must be different from that by which manganese is deposited.

The surprisingly large amounts of nickel found in the sediments of the central Pacific Ocean at first consideration seem to contradict the suggestion that the element is of cosmic origin and derived from meteoric or cosmic dust from interplanetary space. Core 69 may be taken as an example. The average content of NiO in the uppermost 10 cm was 0·051% or 0·040%–Ni. The fresh sediment has a specific gravity of 1·5 and contains 65% seawater and thus the nickel contained in one cc. of the sediment is $200 \times 10^{-6}$ g. By means of a detailed radium analysis of the top layers the rate of sedimentation was found to be 0·04 mm in 1000 years. The accumulation of nickel in this time would be $8 \times 10^{-8}$ g per $cm^2$ of the bottom surface. It is assumed that this nickel remains stationary in the sediment.

Assuming that *the whole* of this nickel is of cosmic origin and that the distribution is uniform within the time-span involved, the total accumulation of the element to the whole surface of our planet becomes $40 \times 10^{12}$ g per 1000 years or 40000 tons per annum. Ascribing an average nickel content of 2% to the cosmic dust, the latter figure has to be multiplied by 50 in order to reach the total contribution of extraterrestrial matter to the earth in the course of one year which gives

2 000 000 tons*. Comparing this figure with that of 1800 tons which was derived from meteor counts according to WATSON, the difference seems disconcerting for the cosmogenic hypothesis.

It was therefore thought desirable to submit the results to authorities on meteors and cosmic dust from interplanetary space. Dr. F. WATSON, in a private communication to one of the writers, states that for various reasons, the figure based on meteor counts can only be taken as a lower limit for the amount of material actually deposited on the earth's surface, and that the true figure may well be higher by one, or even two, powers of ten. Dr. F. WHIPPLE, of the Harvard Observatory, very kindly sent the writers a typescript copy of an unpublished paper on "The theory of micrometeorites." In this paper he discusses the possibility of very small particles, a few $\mu$ or less in diameter, penetrating the atmosphere and being deposited on the earth without being heated to incandescence by aerial friction. Dr. WHIPPLE expresses the opinion that the total mass of these invisible micrometeorites may be 10 000 times greater than that of the visible meteors. Dr. ÖPIK, of the *Armagh Observatory* in N. Ireland and formerly of the *University of Tartu* and the *Harvard Observatory*, had noticed the results published in a letter to Nature and had the kindness to send the writers a typescript copy of a new paper. In this most interesting work, Dr. ÖPIK discusses the total quantity of cosmic dust present in interplanetary space and which is responsible for the zodiacal light. He concludes, from his calculations, that the total quantity of such dust swept up by the earth approaches the value obtained from this study of the nickel in deep-sea deposits and may, with certain assumptions on the composition of this dust, give very close agreement. Dr. ÖPIK, in a personal communication to one of the present writers, stresses the astronomical importance of this research on the deep-sea deposits and the desirability of it being extended to still more material.

These converging opinions from three authorities on meteors and interplanetary dust encourage the view that the main part of the nickel discovered in the deep-sea sediments may be of cosmic origin. Dr. ÖPIK feels that the very considerable variations in the nickel concentrations along the longest cores (Nos. 71 and 72) are more difficult to explain. The above mentioned paper by Dr. ÖPIK deals with these difficulties in detail.

Dr. LANDERGREN, on the other hand, in a recent paper on the results of spectrographic analyses of samples from one of the long "Albatross" cores collected in the Romanche Deep (Lat. S 00° 13′, Long. W 18° 26′, depth 7500 m) where considerable variations in the nickel content were found, expresses the opinion that the nickel may be of terrestrial origin.

* It has been assumed in these calculations that *all* the nickel found in abyssal sediments is of cosmic origin in order to obtain a maximum value for the accumulation of the material required. Obviously this assumption cannot be correct for a considerable part of the nickel is probably of terrestrial origin. Assuming an average of 0·015%–NiO for terrestrial material and deducting this from the 0·051%–NiO found in the top part of core 69 means that the remaining two-thirds or 0·036% would be the net contribution of NiO from extra-terrestrial sources. This would reduce the total quantity of such material carried to the earth from 2 000 000 tons to 1 400 000 tons annually. If a further increase in the terrestrial nickel occurs as a result of secondary processes such as solution, adsorption, etc., a further reduction in the cosmic material is possible. It seems, however, that the maxima of the nickel content, unparalleled by similar maxima in the manganese and iron values, are probably of extraterrestrial origin and consequently of special interest.

One critical experiment which suggested itself was that of analysing red clay samples which were rich in nickel for elements of the platinum group and especially platinum and palladium *. The average platinum content in iron meteorites according to GOLDSCHMIDT should be 20 g per ton and of palladium 10 g per ton, whereas in continental rocks the values are about 10000 times less. In stony meteorites also, the two elements are much less concentrated in relation to nickel.

Dr. HARRISON BROWN of the Institute for Nuclear Physics in Chicago has kindly undertaken to analyse some of the samples for palladium after activation in the neutron pile. It is therefore possible that the question of the absence or presence of platinum metals in a concentration indicative of a cosmic origin may be settled in the near future.

In the meanwhile this investigation is being continued on cores from the Western Pacific Ocean and also from the two other oceans.

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* A qualitative test for platinum and palladium was undertaken by MAG. BAUER with negative results.

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