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Manganese nodules and oceanic radium

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The manganese nodules found by the *Challenger* Expedition in great depths have been studied by Sir John Murray and A. F. Renard (1891). They were recognized as products of a very slow growth, from manganese dioxide and ferric hydroxide, accumulated round a nucleus, a fragment of waterlogged pumice, or a shark's tooth or some other object lying on the surface of the deposit. Chemical analyses carried out by Brazier *et al.* proved a considerable number of trace elements like copper, cobalt, nickel, barium and lead to be present in the nodules. Of special interest was their relatively high content of radium first proved through measurements made by J. Joly (1908).

As I have been engaged on determinations of the radium present in deep-sea deposits since 1921 (1930; 1939; 1953), and later also of the radioactivity of sea-water, my interest was attracted by the radium contained in the nodules. The close affinity between radium and manganese,* well known from deposits at thermal sources on the continents, is no doubt responsible for the entry of radium into the manganese nodules. Granting this to be the case, I assumed that this radium is " unsupported " by its parent element ionium, in which case it should decay with age and consequently also with the depth below the surface of the nodule, according to the half-value period of radium, about 1,600 years.

Thanks to the courtesy of the British Museum of Natural History I obtained from their *Challenger* collections two small nodules, and through a similar courtesy of the Mineralogical Department of the Riksmuseum in Stockholm one half of a much larger nodule, also from the *Challenger* Expedition.

The results of my measurements on samples from these nodules, taken from thin layers in the surface and further inwards, confirmed my assumption. The radium content per gram of the samples fell off from a very high value in the uppermost surface, sometimes exceeding 100 units of the 12th decimal place, to a few units 7 to 10 mm deeper down. The rate of decline proved to be more rapid on the lower, flattened, side of the nodule, on which it had presumably been lying on the bottom, than on the upper, convex, side which must have been exposed to the settling of sediment from above, increasing the rate of growth over the value due only to aggregation into the nodule. The rate of radial growth, calculated from the decrease in radium, proved to be rather less than 1 mm in 1,000 years on the lower side and about 50% more on the upper, convex, side (1943).

The conclusion drawn from these measurements was, that the radium in the nodules is accumulated from the surrounding sediment. These results made it most desirable for me to obtain also other nodules from other parts of the ocean. Through the kind

* An addition of small quantities of powdered " braunstein " and a thorough shaking of the mixture has been proved through experiments in Göteborg, to be a most efficient method for extracting radium and its isotope ThX from extremely dilute solutions.

intervention of Dr. BIGELOW I obtained from the Harvard Museum of Comparative Zoology three manganese nodules, raised out of great depths in the SE Pacific Ocean, from Dr. AGASSIZ's expeditions with the *Albatross*.

Measurements of the radium content of these nodules were started in 1944, soon after they had arrived in Göteborg. However, due to pressure of other work in connection with preparations for a Deep-Sea Expedition with the Swedish *Albatross*, the values I found have not previously been published. As the results in some respects extend those of my earlier publication I have found the occasion propitious for publishing them here, expressing at the same time my sincere gratitude to Dr. BIGELOW for his generous help, not only in this connection but also in other matters, and for the most valuable discussions it has been my privilege to have with him.

NODULES FROM THE "ALBATROSS" PACIFIC CRUISES UNDER DR. AGASSIZ

The nodules obtained were from three stations visited during the Third Expedition. The following descriptions are quoted from J. MURRAY and G. V. LEE (1909, p. 144):

Station 13. Lat. 09° 57′ N, long. 137° 47′ W, depth 2,690 fathoms. "At this station a large quantity of nodules was dredged, one of which is figured in Dr. ALEXANDER AGASSIZ's preliminary report. . . . They average 4 to 6 inches in diameter, and are all irregularly spherical or irregularly cubical, and never tend to form slabs. Their most conspicuous feature is the high degree of mammillated structure: the whole surface is covered with large protuberances, which may be as much as one inch in diameter. Sometimes these protuberances may be compounds, that is, formed of smaller ones, grafted the one on the other. On the one side of the nodule the mammillae have a relatively smooth surface, on which rubbing will develop a shining lustre; on the other side the mammillae are more dull, with a shagreen-like appearance."

Table I

Station Albatross 13. *Lat. N* 9° 57′. *Long. W* 137° 47′. *Depth* 4923 *m*

Average "depth"			*Radium in* 10^{-12} *g/g*	MnO_2	*Fe*
0·2	mm	upper side	38	18·2%	13·3%
3·3	mm		5·4	29·3	13·2
11	mm		3·6	20·9	18·2
1·0	mm	lower side	102		
3·5	mm		19·5		
5·8	mm		9		
13	mm		3·5		
0·15	mm	upper side	61		
0·45	mm		33		
0·75	mm		26		
1·1	mm		34		
1·6	mm		16·7		
2·1	mm		7·2		
4·0	mm		8·2		
4·6	mm		10		
0·15	mm	lower side	189		
0·2	mm		180		
0·6	mm		103		
1·0	mm		85		
1·5	mm		67		

As in earlier experiments, thin layers were removed from the surface and from layers immediately below it. Weighed quantities from these samples were dissolved in hot hydrochloric acid, leaving a small undissolved residue, which was practically free from radium. In some of the samples the content of manganese and of iron were measured by colorimetric methods. The first column in Table I gives the average " depth " of the sample below the surface of the nodule, the second column gives the content of radium in the usual units of the 12th decimal place in gram per gram of the sample. The maximum values obtained from the lower surface of the nodule are seen to be 102 and 189 units of Ra, respectively, whereas from the upper surface the values come out much lower, i.e. 38 and 61 units respectively. The fall-off in radium inwards is quite steep, to one half of the surface value or less for an increase in depth

Table II

Station Albatross 173. *Lat. S* 8° 55′. *Long. W* 146° 32′. *Depth* 4462 *m*

Average " depth "	*Radium in* 10^{-12} *g/g*	MnO_2	*Fe*
0·2 mm upper side	64	23·2%	14·9%
0·2 mm lower side	114	19·1%	15·8%
0·1 mm upper side	51	19·1%	15·8%
0·35 mm	31	20·8%	15·65%
1·35 mm	15	20·6%	50·8%
0·05 mm	35		
0·14 mm	28		
0·28 mm	25		
0·7 mm	15		
0·2 mm lower side	81		
0·5 mm	65		

of 1 mm. Attempts to calculate the rate of radial growth from the rate of this fall-off gave values ranging from 0·6 mm to 1·4 mm in 1,000 years. The percentage of iron is fairly high, between 13% and 18%, whereas the content of manganese is of the same order varying between 18% and 29% of MnO_2.

Station 173. Lat. 18° 55′ S, long. 146° 32′ W, depth 2,440 fathoms. The description of the nodules from this station given by Murray and Lee (1909, p. 146) runs as follows:

" These nodules . . . constitute one of the most remarkable hauls of the cruise. At this station, where the deposit is Red Clay, immense numbers were dredged, varying in size from that of a small hazel-nut to potato-shaped nodules, three or four inches in diameter, and slabs over six inches in length. . . . Although the shape varies, the surface characters are absolutely constant, and belong to a type which was met with only at this particular spot. The surface of all the nodules, large and small, is *even*, that is to say, not covered with protuberances, but it is not *smooth*, being covered with numberless, closely set mammillae, giving it the appearance of shagreen or coarse leather. On account of the presence of these mammillae—1 mm in diameter, as a rule—the surface is dull, not shining. The colour is brown, never black. The external layer, from 1 to 2 mm in thickness, peels off easily. In some cases the layer coming next to it shows shagreen structure; in other cases it does not, being smooth. This external layer is generally the only one that can be removed: the rest of the nodule is very compact, and so hard as to be scratched with a knife only with difficulty. When whole and with no fracture previously existing, the nodules cannot be broken in the hand, even when great strength is exerted; the specific gravity is very high.

This nodule arrived in Göteborg broken into fragments. However, by piecing the fragments together it was easy to reconstruct the nodule and decide which had been its surface. The results from the measurements are set out in Table II, showing maximum values (presumably on the lower side)* of 114 and 81 units of Ra, whereas samples from the upper side contained much less radium, 64, 51 and 35 units respectively. The content of manganese was somewhat lower than in the nodule from Station 13, the content of iron being much the same except for one sample taken at a depth of 1·35 mm where an unusually high value of over 50% Fe was found. Two attempts to calculate the rate of radial growth from the fall-off in radium content gave a value of 0·4 mm in 1,000 years in each case.

The third nodule, taken from Station 4658, was the largest and, according to my view, the most remarkable of the three, both with regard to its shape and its colour. It is described by MURRAY and LEE (1909, p. 29):

> " The nodules from this station are as remarkable for their large size as for their constant shape, which is on a definite pattern. These nodules have each two surfaces, which are, roughly speaking, respectively dome-shaped and cone-shaped. Looking normally at one of the surfaces, the wider portion is circular, so that there is practically an axis of symmetry passing through the apex of both surfaces. The dome-shaped one is due to the aggregation of a few smoothly undulating bosses or protuberances of large radius. It is very smooth and black, with a metallic lustre, and has a distinctly scaly structure. The other surface is mammillated, has a dull colour and is incoherent, breaking up with little exertion of the fingers. A certain amount of clay is, moreover, mixed with the oxides, filling the cavities between the mammillae."
>
> " A section across a nodule shows it to be formed of successive concentric layers following exactly in their distribution the contours of the smooth surface. The innermost layer has absolutely the same shape as the outer one. The difference between the alternate layers is mainly one of hardness. In the samples cut, it has not been possible to find what was originally the centre of accretion; it probably consisted of some material which has since been transformed, or rather, replaced by the oxides.
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> " This particular kind of nodule does not appear to have been described before; the nearest approach to it, as regards shape, is one represented in Fig. 4, Plate 3, of the ' *Challenger* Report on Deep-Sea Deposits ' " (MURRAY and LEE, 1909, p. 30).

A greater number of samples from this nodule than from the two others were submitted to analyses. Before sampling, the nodule was cut in two practically identical parts. The appearance of the section is shown in natural size by the drawing reproduced in Fig. 1, which reveals the concentric arrangement of the different layers in this cauliflower-shaped nodule. Its domed surface, presumably the upper one when lying on the sediment,† was smooth, whereas the lower, conical surface was scoriaceous, which made the sampling of this part rather difficult. Adopting MURRAY'S views on the position of the nodule on the sediment surface, viz. that the scoriaceous, conical surface was the lower one, the higher radium values are ascribed to the lower surface

* According to a footnote given in the paper by MURRAY and LEE their views on the original position of this nodule differed (see above).

† Dr. LEE holds that the logical conclusion is that the cone-shaped mammillated surface is the upper surface, the smooth shining one being embedded in the clay, whereas Sir JOHN MURRAY takes the view, from his *Challenger* experience, that the smooth surface was the upper one, and points to Fig. 1, Plate IX, of the " *Challenger* report on deep-sea deposits " as confirmation of this, the smooth surface in his opinion being formed above the level of the deposit.

with maxima varying between 85 and 330 units of Ra. The upper surface had much lower values, ranging from 25 to 61 units. The fall-off in the radium content with depth is more difficult to estimate, owing to the uneven, mamillated lower surface. With due reserve for this uncertainty we find the rate of radial growth in the downward direction comes out at only 0·15 mm in 1,000 years, which is only a fraction of the rate of growth in the upward direction, 0·4 mm in 1,000 years. The content of manganese in this nodule is seen to be very high, ranging from 30% to 75% of MnO_2.* On the other hand the content of iron is very low, averaging 1·3% of Fe, with an absolute minimum of only 0·03%.

Low values of radium content were found in the nucleus and its vicinity, viz. 5 to 6 units of Ra. Still lower values were found in depths of from 4·5 to 15 mm in a series of samples taken from the back of the nodule, near the lower limit of the domed

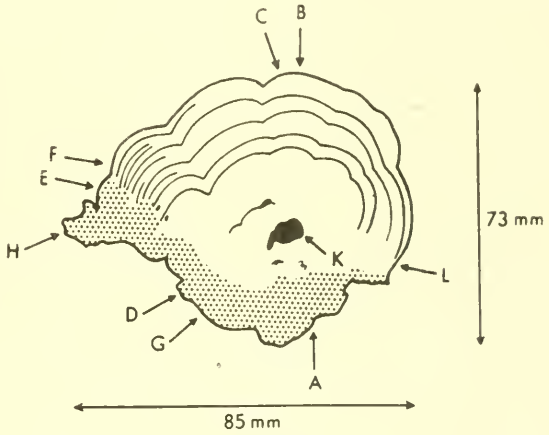

Fig. 1

surface, where radium values as low as 2·2 to 2·4 units were measured. The results found with this extraordinary nodule, when summarized, seem to tell of an intense transfer of both manganese and radium from the sediment to the nodule, especially to the scoriaceous parts.

It would have been most interesting to measure the radium, manganese and iron contained in the sediment surrounding this nodule, but unfortunately no sediment from the station was available. I collected instead a sample of the sediment entrapped within the nodule and measured it (last entry in Table III). The radium content was moderate (24 units), whereas the manganese dioxide was relatively low (9·1%), and the content of iron quite high for this nodule, viz. 4·1%. However, too much importance should not be attached to these figures, since an exchange between this entrapped

* Only in one nodule, taken by the *Challenger* Expedition in the Pacific Ocean lat. 33° 31′ S, long. 74° 43′ W, depth 3,950, was a content of manganese dioxide as high as in this nodule reported by MURRAY and RENARD, viz. 77% MnO_2 in a nodule from blue mud. The iron content of that nodule was low, 4·5%.

sediment and the surrounding layers of the nodule may have occurred. With due reserve for the limited number of samples investigated, one may say that there is strong evidence for a transfer of radium from the sediment via the bottom water to the nodule, whereas the accretion of radium from the water column above is not likely to have occurred to a notable extent.

By what kind of chemical action the solution of radium and other elements has taken place is unknown, owing to our ignorance regarding the chemical reactions, proceeding with immense slowness, in the contact region between sediment and bottom water.

Table III

Station Albatross 4658. *Lat.* 14° 29′. *Long. W* 81° 24′. *Depth* 4960 *m*

Series	*Average "depth"*	*Radium in* 10^{-12} *g/g*	MnO_2	*Fe*
a	0·3 mm lower surf.	186	49·8%	3·2%
b	0·2 mm upper surf.	58	43·4	6·0
b	3·2 mm upper surf.	3·0	64·9	0·4
d	0·2 mm Scoriacae	85	53·6	1·15
d	12 mm	29	58·3	1·3
e	0·2 mm edge	46	68·2	0·95
f	0·2 mm shell	31	71·2	1·0
c	0·5 mm summit	25	66·7	1·7
c	0·5 mm summit	9·5	68	1·1
c	2·5 mm	3·0	74·5	0·55
	0·5 mm back of nodule	25	74·8	0·67
	1·5 mm	9·6	70·4	0·47
	2·5 mm	3·3	62·2	0·44
	3·5 mm	3·0	70·0	0·55
	10 mm	2·4	69·8	—
	15 mm	2·2	69·8	—
m	0·2 mm	61	—	—
m	0·5 mm	17·0	61·6	—
m	0·85 mm	10·0	61·4	—
k	14 mm nucleus	5·5	64·9	—
k	14 mm cavity	9·0	30·8	
g	0·15 mm Scoriacae	160	44·7	1·0
g	0·7 mm Scoriacae	13·7	64·4	0·03
g	1·3 mm Scoriacae	3·0	62·1	0·14
h	0·2 mm Scoriacae	330	44·5	0·35
	sediment	24	9·14	4·1
k	12 mm nucleus	6·1	64·9	

It does not seem unlikely that submarine volcanic activity, which has undoubtedly played a dominant part in the building up of the islands and the "guyots" in the Pacific Ocean, may have given rise to magmatic acids, converting calcareous sediments into Red Clay and, at the same time, dissolved also other sediment components, thus facilitating their transfer into manganese nodules, manganese crusts and other concretions. It seems reasonable to assume that this acidulation has been concentrated to a relatively thin bottom layer over low-lying parts of the ocean floor.

OCEANIC RADIUM

The early hypotheses, put forward by JOLY and other early workers in the field, for the origin of deep-sea radium, have not withstood later criticism. This applies also to other explanations for the surprisingly high radium content in the deepest deposits.

(A) A precipitation of radium as insoluble sulphate from the water (JOLY).

(B) A production *in situ* from the uranium present in the sediment (JOLY).

(C) A cosmic origin of the radium, like that of the cosmic spherules (MURRAY).

(D) A magmatic origin from submarine volcanoes (MURRAY).

(E) A biological extraction from sea water by plankton organisms (EVANS, *et al.*, 1938).

Only the two last alternatives need to be considered seriously. The possibility that some of the deep sea radium is actually derived from volcanoes on the ocean floor cannot be denied. In several instances abnormally high radium values have been found both in the Tyrrhenian Sea and in the equatorial Atlantic, for which a magmatic origin seems probable. That radium to a certain extent becomes adsorbed by marine plankton has been supported by measurements in Göteborg and elsewhere. However, this biological extraction and subsequent transfer to the bottom deposits with siliceous or calcareous tests cannot be a *major* cause of the high radium content in deep-sea deposit. Such " unsupported " radium must have a highly transient existence in the very uppermost surface layer of the deposit.

In order to find a better explanation for the high radium content in the Red Clay and in the Radiolarian Ooze, found by JOLY and confirmed through my own measurements, teamwork on the radioactive elements present in ocean water was started by me over 20 years ago in Göteborg in collaboration with specialists from Vienna and Oslo (1939). By a special technique, extraction of radium from large-volume samples of sea water (20–40 litres), through co-precipitation with radium-free sulphate of barium, dependable values for the radium content were obtained. They turned out to be surprisingly low, in general less than one unit of the 13th decimal place. At the same time, accurate measurements of the content of uranium in sea water were, for the first time, carried out by means of a fluorescence method, developed by HERNEGGER and KARLIK (1935) of the Institut für Radiumforschung in Vienna. Their results indicated a fairly constant uranium content averaging $1{\cdot}3 \times 10^{-7}$ gr uranium per litre sea water of normal salinity (35‰). In a state of radioactive equilibrium this uranium content would correspond to over $0{\cdot}4 \times 10^{-12}$ g Ra per litre of sea water, or to 6 times more than the radium actually found.

In order to explain this partial disappearance of radium from the sea water, I suggested in 1937 (1938), that its parent element, ionium, is being constantly removed from the water through a co-precipitation with ferric hydroxide, which is known to go on in the sea. This transfer of the ionium produced from dissolved uranium to the ocean bottom would explain the high radium content in deep-sea deposits as due to ionium-supported radium. From this ionium-precipitation hypothesis it would necessarily follow that the radium content in the deposit should increase to a near-surface maximum, attained about 9,000 years after precipitation, characterized by a radioactive equilibrium between ionium and radium. From there downwards in the sediment the content of the two elements should decrease together, according to the half-value period of ionium, about 82,000 years.

The first attempt to utilize the ionium-precipitation hypothesis for a submarine geochronology, based on radium measurements, was made in the early 70's by C. S. PIGGOT and W. D. URRY (1941) and by URRY (1949). Their first results, obtained from fairly long sediment cores from the Caribbean and the N. Atlantic Ocean (published as curves only), gave hopes of realizing a dependable method of dating different layers of a sediment core and of obtaining values for the rate of sedimentation from radium measurements. More extensive measurements on cores from the Swedish Deep-Sea Expedition, started after the return of the Expedition, are now being published by V. KRÖLL, who has for some years been working in Göteborg (1955). The results prove that the vertical distribution of radium in the cores is much more complicated than the ionium-precipitation hypothesis alone would imply. Often two, three, or even more maxima, beside that to be expected from radioactive equilibrium, Ra/Io, are actually found.

The opportunities offered by the Swedish Deep-Sea Expedition for studying the radioactivity of the ocean were used for an extensive sampling for radium analyses of large-volume water samples. After the return of the Expedition, the barium-sulphate precipitates were analyzed for radium, partly by the present author in Göteborg and partly in Vienna by Dr. TRAUDE BERNERT. The results, which have not been published before, are here set out in Table IV. They are seen to confirm our earlier results, viz. that the radium present in the ocean waters is only a small fraction of the quantity in equilibrium with dissolved uranium. In addition, uranium measurements on most of the water samples were carried out in Vienna by G. KOCZY (1950), with results agreeing with those found earlier by HERNEGGER and KARLIK. There cannot then be any doubt about the partial disappearance from the ocean waters of the radium produced from dissolved uranium.

However, a missing link in the chain of evidence concerns the presence of ionium in the deposits and in the water. Thanks to an ingenious method for photographic (nuclear-plate) determinations of ionium, Dr. E. PICCIOTTO and his co-workers in the Institut des Recherches Nucleaires of the Université Libre in Brussels, have been able to measure the ionium contained in sediments from the Swedish Deep-Sea Expedition (1954). It was found that, within the limits of experimental accuracy, the ionium present is in radioactive equilibrium with the radium in the same sample, except in the very uppermost surface layer, where there is an excess of ionium. On the other hand, more recent measurements of the ionium present in sea water, carried out by PICCIOTTO and F. KOCZY, have proved the ionium content to be extremely low, lower even than what could correspond to the radium content, which in itself, as we have seen, is only a fraction of the equilibrium value with dissolved uranium.

This result raises a problem of considerable interest. Since the scarcity of ionium in ocean water negatives the assumption that the radium in the water is produced *in situ* from its parent element ionium, the question arises, from what source is the radium contained in the ocean water derived?

Dr. F. KOCZY has proposed an explanation, put forward in a paper read before the meeting of the I.U.G.G. in Rome in Sept. 1954, that oceanic radium is dissolved through a chemical interaction between the bottom water and the uppermost 3–5 cm sediment layer. From the bottom water the radium is spread upwards through the water masses by diffusion, aided by turbulence. This view KOCZY finds supported by the curves in Fig. 3, giving the vertical distribution of the radium in the water

column found from the measurements already quoted and set out in Table IV. The curves are seen to have maxima in the uppermost water layers, then to fall to intermediary minima at a depth of about 1,500 metres (possibly due to biologic extraction) and, after rising again to a maximum in a depth of 2,000 to 4,000 metres, attaining even higher values in still greater depths.

As has already been pointed out here on pp. 335 and 339, the passing over of radium from the sediment surface-layer to the manganese nodules resting on it speaks in favour of a solution of radium near or in the sediment surface actually taking place.

Accepting Dr. KOCZY's explanation as plausible, we arrive at a rather unexpected picture of the origin of the radium in ocean water.

The ancestral element uranium is being carried into the ocean by rivers from denuded rocks on the continents. The ionium produced from the dissolved uranium

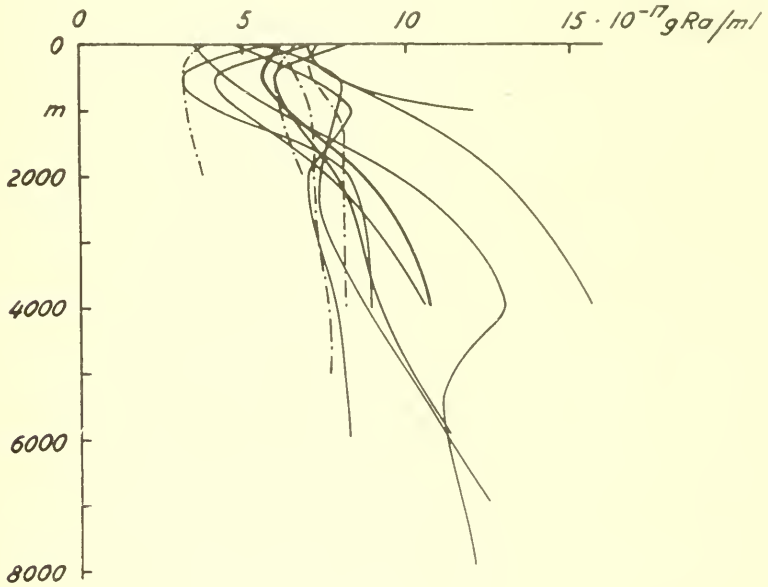

Fig. 3

is very efficiently removed from solution through precipitation, and settles over the ocean bottom, there giving rise to its descendant radium. The radium in the uppermost few centimetres of the sediment passes readily over into solution, some of it becoming reprecipitated or adsorbed together with manganese in the nodules. Another part which has become dissolved from the sediment is distributed through the supernatant water masses, giving them a radium content intermediate between that in equilibrium with the dissolved uranium and the much lower quantity in equilibrium with dissolved ionium.

Table IV

Lat.	*Long.*	*Depth*	*Ra g/Lit.* $\cdot 10^{12}$	*U g/Lit.* 10^{6}
N 00° 13′	W 18° 26′	25 m	0·07	1·1
		500 m	0·08	1·2
		2500 m	0·07	1·2
		5000 m	0·10	1·3
		7500 m	0·115	1·4
N 14° 15′	W 71° 02′	0 m	0·06	
N 14° 27′	W 66° 13′	1000 m	0·07	1·3
		3000 m	0·075	1·2
		5000 m	0·075	1·2
N 28° 05′	W 60° 49′	25 m	0·045	1·15
		500 m	0·07	1·2
		1000 m	0·08	1·4
		2000 m	0·07	1·3
		4000 m	0·08	1·3
		6000 m	0·08	1·1
N 43° 04′	W 19° 40′	25 m	0·05	
		500 m	0·06	
		1000 m	0·065	
		2000 m	0·065	
		4000 m	0·08	
		6000 m	0·11	
N 21° 24′	W 46° 24′	0 m	0·035	
		100 m	0·035	
		400 m	0·035	
S 02° 52′	W 89° 56′	20 m	0·09	1·1
		500 m	0·09	1·1
		1000 m	0·15	1·3
S 13° 24′	W 149° 30′	0 m	0·10	1·1
		1000 m	0·12	1·4
		2000 m	0·13	1·3
		3000 m	0·17	1·1
		4000 m	0·17	1·1
N 05° 10′	E 127° 53′	0 m	0·035	
		500 m	0·050	
		2000 m	0·10	
		4000 m	0·13	
		5700 m	0·11	
		8200 m	0·12	
S 01° 09′	E 126° 22′	3000 m	0·07	
S 00° 00′	E 88° 18′	10 m	0·055	
		500 m	0·05	
		2000 m	0·07	
		4000 m	0·09	1·3
N 10° 06′	E 52° 15′	20 m	0·05	
		500 m	0·04	
		2000 m	0·08	
		3750 m	0·11	
N 21° 09′	E 38° 07′	20 m	0·05	1·1
		500 m	0·06	1·3
		2000 m	0·06	1·1
N 35° 41′	E 21° 50′	50 m	0·065	
		1000 m	0·11	
		2000 m	0·07	
		4000 m	0·09	

Average: 0·078

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