# U<sup>254</sup>/U<sup>255</sup> ACTIVITY RATIOS IN SOUTH PACIFIC OCEAN WATERS

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Received October 27, 1969

#### ABSTRACT

Analyses of uranium extracted from South Pacific waters by the *in-situ* extraction method point to a uniform depth independent value of  $1\cdot 14 \pm 0\cdot 02$  for the  $U^{224}/U^{238}$  activity ratio. This value is also the same, within one standard deviation, as that determined for the N. Pacific, S. Indian and Atlantic waters.

#### Introduction

THE radioactive disequilibrium between uranium-238 and its daughter uranium-234, found in minerals and rocks by Russian workers<sup>1</sup> and in corals, mollusks and oolities by Thurber<sup>2</sup> has led to useful methods of dating Pleistocene events.<sup>3</sup>

Uranium in sea-water is believed to exist in the hexavalent state in the form of the carbonate complex,  $UO_2$  ( $CO_3$ )<sub>3</sub>-4. It is relatively inactive in this state and has a long residence time of  $5 \times 10^5$  years. Because of this long residence time, the uranium concentration in ocean waters as well as the activity ratio of  $U^{234}/U^{238}$  is expected to be uniform in open ocean waters. Koide and Goldberg<sup>4</sup> and several others<sup>5-8</sup> have analyzed coastal and open ocean waters from the North Pacific, Atlantic, and Indian Oceans and found the  $U^{234}/U^{238}$  activity ratio to be remarkably uniform at a value of  $1 \cdot 14 \pm 0 \cdot 02$ . In this paper we report on  $U^{234}/U^{238}$  ratios in South Pacific waters, surface to 3500 m. depth. These results give credence to the generally accepted uniformity of  $U^{234}/U^{238}$  ratios in oceans.

#### EXPERIMENTAL PROCEDURE

During the Nova Expedition of the Scripps Institution of Oceanography ferric hydroxide deposited spongin matrix<sup>9</sup> was towed in sea-water at several locations and depths using butterfly stainless steel samplers.<sup>10</sup> Sea-water was flushed through the matrix at the desired depth by raising and lowering

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the sampler for 10-15 hours at mean speeds of about 10 m/min. The samplers descended to the profile depth in an orientation which did not allow water to flow through the matrix. Corrosion of a magnesium strip at the profile depth swung the samplers in the correct orientation for free flow of water. During ascent, the sampler was in the open position and part of the uranium picked up is from shallower depths. However, considering the operational times and the fact that the pick-up rate decreases with time, more than 90% of uranium picked up corresponds to the profile depth. Leaching of uranium from the towed matrix and its purification and plating were done according to the procedures published earlier.6, 11 A few counts per minute activity of U232 was added as a chemical yield tracer to each of the samples before processing; the plated samples were assayed for their alpha-activity using a surface barrier detector operated in conjunction with a 128 channel analyzer. Chemical efficiencies were around 30%. activity of uranium in control samples of untowed ferric hydroxide matrix was found to be less than about 10% of the activity observed for the samples; the correction for any uranium activity originally present in the treated spongin matrix is therefore small.

### RESULTS AND DISCUSSION

A typical alpha-spectrum due to a uranium sample is shown in Fig. 1. Samples were usually counted long enough to obtain a statistical error of  $\pm 0.02$ –0.03 on the activity ratio. The  $U^{234}/U^{238}$  activity ratios were determined from such spectra in the usual manner,<sup>4</sup> correcting for

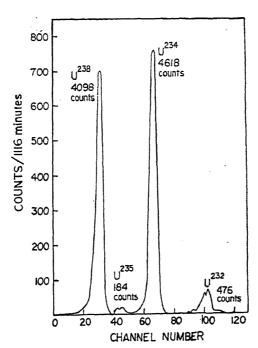


Fig 1. A typical alpha-spectrum of uranium activity extracted from South Pacific waters.

inclusion of alphas from U<sup>235</sup> (4·12 MeV—5%) in the U<sup>238</sup> peak (4·20 Mev). All results along with the sample details are given in Table I.

TABLE I  $U^{234}$ ,  $U^{238}$  Ratios in South Pacific Waters

Station and location	Depth (meters)	U <sup>234</sup> /U <sup>238</sup> Activity ratio
NOVA VI-5	50-150	$1.11 \pm 0.02$
	500-600	$1.13 \pm 0.02$
23 42·5′ S	1100-1200	$1.14 \pm 0.02$
176 02·6′ E	2200-2300	$1.13 \pm 0.02$
	3400-3500	$1.12 \pm 0.02$
NOVA VI-4	50-150	$1.15 \pm 0.03$
	500-600	$1.12 \pm 0.03$
27: 20-9' S	1100-1200	$1.16 \pm 0.03$
175° 25•0° E	2200–2300	$1.17 \pm 0.03$
	3400-3500	$1.16 \pm 0.03$
NOVA VI-1	50-150	$\frac{1.13 \pm 0.03}{1.13 \pm 0.03}$
	500-600	$1.17 \pm 0.02$
31° 41′ S	1100-1200	$1.16 \pm 0.02$
177 16-2 W	2200-2300	$1.13 \pm 0.02$

Average Value:  $1.14 \pm 0.02$ .

The activity ratios of the individual samples of the three profiles ranged from  $1\cdot 11 \pm 0\cdot 02$  to  $1\cdot 17 \pm 0\cdot 02$ . The mean of all the results is  $1\cdot 14 \pm 0\cdot 02$  and thus within the errors of measurements, the activity ratio the mean value of  $1\cdot 14 \pm 0\cdot 014$  reported for North Pacific, South Indian and the Atlantic waters.

# ACKNOWLEDGEMENTS

Acknowledgement is made to the donors of the Petroleum Research Fund administered by the American Chemical Society for the support of this research. One of us (B. L. K. S.) was an ACS-PRF Fellow during the Nova Expedition. We thank P. K. Talekar for his assistance in the sponge processing.

## REFERENCES

1. Cherdynstev, V. V., Orlov, D. P., Isabev, E. A. and Ivanov, V. I.

Geokhimiya, 1961, 10, 848 (Russian).

2. Thurber, D. L.

. J. Geophys. Res., 1962, 67, 4518.

3. Veeh, H. H.

.. Ibid., 1966, 71, 3379.

4. Koide, M. and Goldberg, E. D.

In Progress in Oceanography, (ed. M Sears), Pergamon Press, Oxford and New York, 1965, 3, 173.

5. Blanchard, R. L.

.. J. Geophys. Res., 1965, 70, 4055.

6. Somayajulu, B. L. K. and Goldberg, E. D.

Earth Planet. Sci. Let., 1966, 1, 102.

7. Veeh, H. H.

.. Geochim. Cosmochim. Acta, 1968, 32, 117.

8. Sarma, T. P. and Krishnamoorthy, T. M.

Curr. Sci., 1968, 37, 422.

9. Lal, D., Arnold, J. R. and Somayajulu, B. L. K.

Geochim. Cosmochim. Acta, 1964, 28, 1111.

Craig, H., Dixon, F. S.,
Lal, D., Somayajulu,
B. L. K. and Weiss,
R. F.

1969, (in preparation).

11. Bhat, S. G., Krishnaswamy, S., Lal, D., Rama and Moore, W. S. Earth Planet. Sci. Let., 1969, 5, 483.