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PLUTONIUM IN THE BIKINI WATER SAMPLES

This is a preliminary report on the plutonium isotopes in the Bikini water samples collected and processed on the research vessel "Palumbo" from October 1972 - December 1972. The procedure followed for the water samples was as follows: About 100 - 400 liters of unfiltered water were acidified to pH 1 - 2 with Con-HCl in the steel settling towers 80" high. The water was stirred by a rising column of air bubbles. Spikes of Pu<sup>242</sup> and Am<sup>243</sup>, along with 10 mg Fe/liter as a chloride and 0.25 ml/liter of 0.3% Separan as a coagulant, were added. The spikes contained 4.442 dpm of Pu<sup>242</sup> and 1.697 dpm Am<sup>243</sup>. The pH was raised to a value of 8 - 9 by addition of ammonium hydroxide, the stirring continued for an additional 30 - 40 minutes and the precipitate was allowed to settle for two to six hours. Each precipitate was removed thru a valve at the bottom of the settling tower and centrifuged to a final volume of 100 ml of flocculant precipitate.

*Separan should be 1%  
~ 0.4 dpm/L.  
losses to sides?*

The ferric hydroxide precipitate was dissolved in minimum amount of Con-HCl, warmed and centrifuged. Undissolved residue was treated with HNO<sub>3</sub> and HF to remove organic matter and silica. After repeated evaporation with HNO<sub>3</sub>, the residue was dissolved in a minimum amount of 8N HCl and added to the original solution. In a few earlier samples, fuming perchloric acid was used, but is now being abandoned

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in favor of nitric acid. Hydrochloric acid gas was then passed for 15 - 20 minutes in an ice-cold solution of the above sample and iron was removed by ether extraction. The iron-free solution was evaporated and dissolved in 8N HNO<sub>3</sub>. The majority of the samples were clear at this stage, but when turbidity persisted, HF, HNO<sub>3</sub> process was repeated. 2 ml 5% NaNO<sub>2</sub> solution was added to the hot 8N HNO<sub>3</sub> solution of the sample and after cooling was passed thru 5 ml Ag1x-8, (100 - 200) mesh, NO<sub>3</sub> form anion exchange column. After washing the column with 8N HNO<sub>3</sub> and Con-HCl respectively, Pu was eluted from the column with a mixture of 75 ml Con-HCl + 1 ml HI. The eluate, after repeated evaporation with HNO<sub>3</sub> and HCl, was passed thru another 2 ml column of anion exchange resin and then plated on a polished stainless steel disc at a current density of 1 amp/cm<sup>2</sup>. The plutonium sample was deposited on 100 mm<sup>2</sup> area and counted on an alpha-spectrometer system (the spectrometer consisting of four diode detectors of 200 mm<sup>2</sup> surface area). The electronic system consisted of Tennelec power supply (TC 908), a pre-amp (TC 164), a linear amplifier (TC 211), biased amplifier (TC 252), Nuclear Diode discriminator (ND 531), Nuclear Diode router (ND 521) and a Nuclear Data 512 channel analyzer (Series #130). The background levels, based on 3 - 6 day counts, were as follows: at 4.90 mev (Pu<sup>242</sup>) (0.005 - .0034 cpm); at 5.15 mev (Pu<sup>239+240</sup>) (0 - .0034 cpm); at 5.49 mev (Pu<sup>238</sup>) (0.000 - .0005 cpm). Resolution of the detectors was about 65 -

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67 kev (FWHM). Each sample was counted for 48 - 72 hours to collect reasonable counts for acceptable statistics.

RESULTS:

Table 1 gives the data on B series of sea water samples which were collected inside of Bikini Atoll. Table 2 is for the C series of samples collected in the crater areas of Bikini Atoll and Table 3 is for the D series samples in the open ocean surrounding Bikini Atoll. The sample locations and concentrations of plutonium are plotted in Fig. 1, a map of Bikini Atoll. Following are some of the salient points of the preliminary report:

- (1) (B) series samples exhibited concentrations of  $Pu^{239+240}$  from 35 dpm/1000L to 283 dpm/1000L. Higher concentrations were found in the northwest and lower in the southeast.
- (2) The concentration of plutonium in bottom waters was generally higher than the surface concentration at the same location; the only exceptions were sample B-7, which was collected at the mouth of Pukoji Pass, and sample B-26 from the east-central area of the Lagoon. The presence of excess plutonium in most samples from depth is consistent with the premise that the primary source of Pu is from contaminated crater sediments and the physical and biological sedimentation and the

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association of plutonium with sinking particles is of major importance in the marine distribution of plutonium.

- (3) The mean ratio of  $\text{Pu}^{238}/\text{Pu}^{239+240}$  in the (B) series samples was  $(0.020 \pm 0.004)$  which agrees well with the ratio of 0.024 used by Hardy, Kray and Volchok (Nature Vol. 241, 444, 1973) as an average weapons  $\text{Pu}^{238}/\text{Pu}^{239+240}$  ratio (Fig. 2).
- (4) The concentration of plutonium in the (C) (crater) samples varied from 70 - 270 dpm  $\text{Pu}^{239+240}/1000\text{L}$  and the average  $\text{Pu}^{238}/\text{Pu}^{239+240}$  ratio in these samples was  $(0.056 \pm .006)$  which is much higher than the mean of the P series samples. ← why?
- (5) In the (D) series samples the concentration of plutonium was the lowest found in this suite of samples. The Pu content varied from 4 - 16 dpm  $\text{Pu}^{239+240}/1000\text{L}$ , except in D-9 surface sample, which was collected in the Dokororyuru Pass and which contained  $97.4 \pm 6.8$  d/m  $\text{Pu}^{239+240}/1000\text{L}$ . The high value in this sample reflects surface flow of water out of the lagoon to the west. The average  $\text{Pu}^{238}/\text{Pu}^{239+240}$  ration was  $(0.041 \pm .017)$  in the open-sea samples.
- (6) The water from the Zuni and Bravo craters exhibited high  $\text{Pu}^{238}/\text{Pu}^{239}$  ratios (Zuni surface 0.13; Bravo surface

0.39; bottom, 0.057, 0.063, 0.076), but the water from the Tewa crater, the south side of which is open to the lagoon, exhibited low ratios (surface 0.013, bottom 0.016) which were characteristic of the ratios for the central lagoon (Fig. 2).

In addition to the water samples, analyses have been made on 8 plankton samples from stations at which water samples were also collected. The results of the plankton analyses are as follows:

Sample #	d Pu <sup>239,240</sup> /m/g ash	d Pu <sup>239,240</sup> /m/g wet	d Pu <sup>238</sup> /m/g ash	
B-2	84 ± 2	7.9 ± 1	1.0 ± .1	.012
B-21	47 ± 2	2.6 ± .1	1.1 ± .2	.024
B-22	138 ± 5	5.7 ± .2	1.1 ± .1	.030
B-25	115 ± 7	5.3 ± .3	.9 ± .2	.075
B-27	28 ± 8	1.5 ± .4	.7 ± .5	.025
B-31	25 ± 1	1.0 ± .04	.4 ± .06	.046
C-3	130 ± 7	9.5 ± .5	5.1 ± .5	.039
D-6	6 ± .3	.24 ± .01	.3 ± .1	.040

The ratios  $\text{Pu}^{238}/\text{Pu}^{239,240}$  followed the ratios found in the sea water with the C-3 water sample from the Bravo Crater having a surface water ratio of .04 and a bottom water ratio of .06 and the plankton a ratio of .04. The open sea water outside of Bikini Lagoon had a ratio of approximately .04 and the plankton at Station D-6 had a ratio of .05. Three of

the plankton samples taken in the water of the lagoon had ratios of .01, two near the edge had ratios of .02 and the sample at B-27, which is directly in the flow of open sea water through which you pass into the Lagoon, had a ratio of .03.

The plankton samples analyzed thus far also reflect the levels of activity in the water mass in which the plankton were collected. The concentration factors calculated on the basis of amounts of  $\text{Pu}^{239,240}$  in the water and in a corresponding weight of plankton were remarkably uniform. The concentration factors ranged from  $2.4 \times 10^4$  to  $3.6 \times 10^4$  and the average concentration factor for the plankton samples analyzed thus far averaged  $3 \times 10^4$  x the concentration of the radionuclides in the water. These concentration factors appear to be high by approximately a factor of 10 over those generally reported in the literature.

CF =  $10^4$

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TABLE 1

CONCENTRATION OF PLUTONIUM IN THE B SERIES WATER SAMPLES

Sample	Volume (L)	Pu <sup>239+240</sup> dpm/1000L	Pu <sup>238</sup> dpm/1000L	Pu <sup>238</sup> /Pu <sup>239+240</sup>
B 2 Surface	200	217 ± 15	4.5 ± 0.7	0.021 ± .003
B 2 Bottom	200	258 ± 13	3.8 ± 0.5	0.015 ± .002
B 3 Surface	100	208 ± 16	3.4 ± 0.9	0.016 ± .004
B 3 Bottom	100	226 ± 9	4.6 ± 0.6	0.020 ± .003
B 7 Surface	200	64 ± 4	1.3 ± 0.2	0.020 ± .003
B 7 Bottom	200	50 ± 3	1.5 ± 0.3	0.030 ± .006
B 8 Surface	200	78 ± 4	1.8 ± 0.4	0.023 ± .005
B11 Surface	200	72 ± 4	1.4 ± 0.3	0.019 ± .004
B15 Surface	400	109 ± 9	2.0 ± 0.3	0.018 ± .002
B15 95'	200	168 ± 16	3.7 ± 0.9	0.022 ± .005
B18 Surface	200	152 ± 9	3.3 ± 0.5	0.022 ± .003
B18 94'	100	235 ± 19	3.0 ± 0.9	0.013 ± .004
B21 Surface	200	113 ± 8	2.4 ± 0.5	0.021 ± .004
B21 Deep	200	130 ± 7	3.6 ± 0.5	0.028 ± .004 - <sup>h</sup>
B25 Surface	200	148 ± 8	2.0 ± 0.4	0.014 ± .003
B25 Deep	200	283 ± 21 - <sup>h</sup>	3.3 ± 0.6	0.012 ± .002 - <sup>h</sup>
B26 Surface	200	117 ± 6	1.7 ± 0.3	0.015 ± .003
B26 Bottom	200	106 ± 13	2.4 ± 0.9	0.023 ± .009
B30 Surface	200	123 ± 7	2.0 ± 0.4	0.016 ± .003
B30 Bottom	200	181 ± 21	4.4 ± 1.1	0.024 ± .006
B31 Bottom	200	35 ± 2 - <sup>h</sup>	0.8 ± 0.2	0.023 ± .006

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Mean = (0.020 ± .004)

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TABLE 2

CONCENTRATION OF PLUTONIUM IN THE C SERIES WATER SAMPLES

Sample	Volume (l)	Pu <sup>239+240</sup> dpm/1000l.	Pu <sup>238</sup> dpm/1000L	Pu <sup>238</sup> /Pu <sup>239+240</sup>
C 1 Bottom	100	229 ± 8	13.1 ± 0.8	0.057 ± .003
C 2 Bottom	100	269 ± 14	17.0 ± 1.4	0.063 ± .005
C 3 Surface	100	71 ± 3	2.8 ± 0.5	0.039 ± .007
C 5 Bottom	100	231 ± 15	17.6 ± 1.8	0.076 ± .008
C 8 Bottom	100	147 ± 5	2.4 ± 0.4	0.016 ± .003
C11 Surface	100	81 ± 3	10.9 ± 0.8	0.13 ± .01
C 8 Surface	100	151 ± 7	1.9 ± 0.4	0.013 ± .003

Mean = (0.056 ± .006)

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TABLE 3

CONCENTRATION OF PLUTONIUM IN THE D SERIES WATER SAMPLES

Sample	Volume (L)	Pu <sup>239+240</sup> dpm/1000L	Pu <sup>238</sup> dpm/1000L	Pu <sup>238</sup> /Pu <sup>239+240</sup>
D 1 300m	66	11.2 ± 1.5	0.6 ± 0.1	0.054 ± .009
D 2 Surface	300	4.2 ± 0.4	0.3 ± 0.1	0.071 ± .023
D 2 300m	86	10.5 ± 1.1	0.3 ± 0.2	0.024 ± .017
D 2 1550m	84	8.2 ± 0.9	0.4 ± 0.2	0.050 ± .025
D 6 Surface	100	8.2 ± 0.7	0.2 ± 0.1	0.023 ± .013
D 6 Bottom	88	15.9 ± 3.0	0.7 ± 0.4	0.044 ± .025
D 9 Surface	100	97.4 ± 6.8	2.4 ± 0.8	0.025 ± .008
D 9 900m	74	8.7 ± 0.7	0.3 ± 0.1	0.037 ± 0.016

Mean = (0.041 ± .017)

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○ 11.2 (300M)

71 SURFACE  
229 BOTTOM  
269 BOTTOM  
231 BOTTOM

○ 217 SURFACE  
258 BOTTOM

○ 208 SURFACE  
226 BOTTOM

○ 42 SURFACE  
10.5 300M  
8.2 1530M

○ 113 SURFACE  
130 BOTTOM

97 SURFACE  
9 BOTTOM

○ 164 SURFACE  
30 BOTTOM

○ 78 SURFACE

○ 81 SURFACE

152 SURFACE  
235 BOTTOM

○ 151 SURFACE  
147 BOTTOM

7.8 SURFACE

109 SURFACE  
160 BOTTOM

○ 148 SURFACE  
183 BOTTOM

123 SURFACE  
181 BOTTOM

○ 35 BOTTOM

○ 72 SURFACE

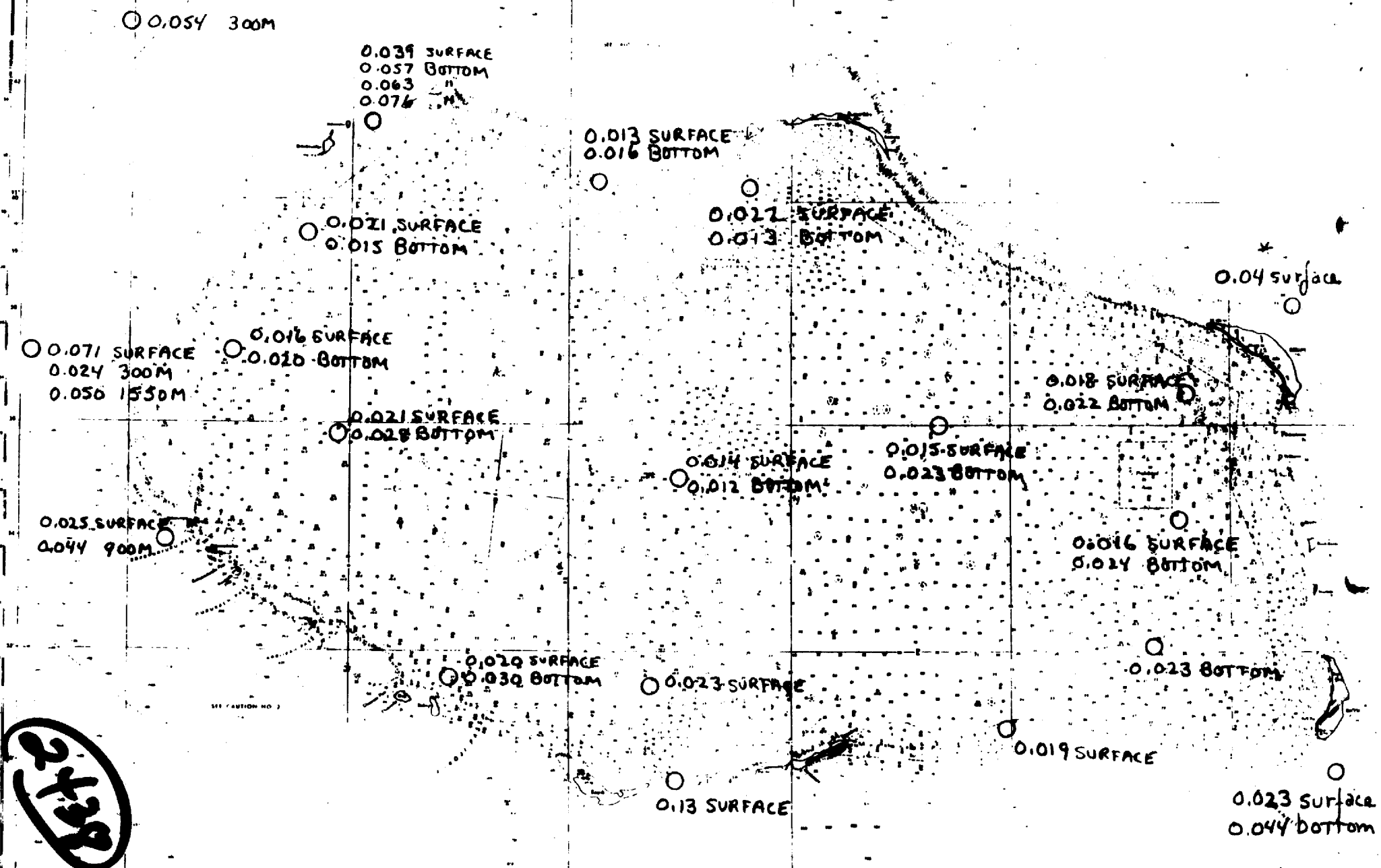
○ 8.2 SURFACE  
15.9 BOTTOM

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Pu 239.240 in sea water in and near Bikini Lagoon in d/m/100 liters.

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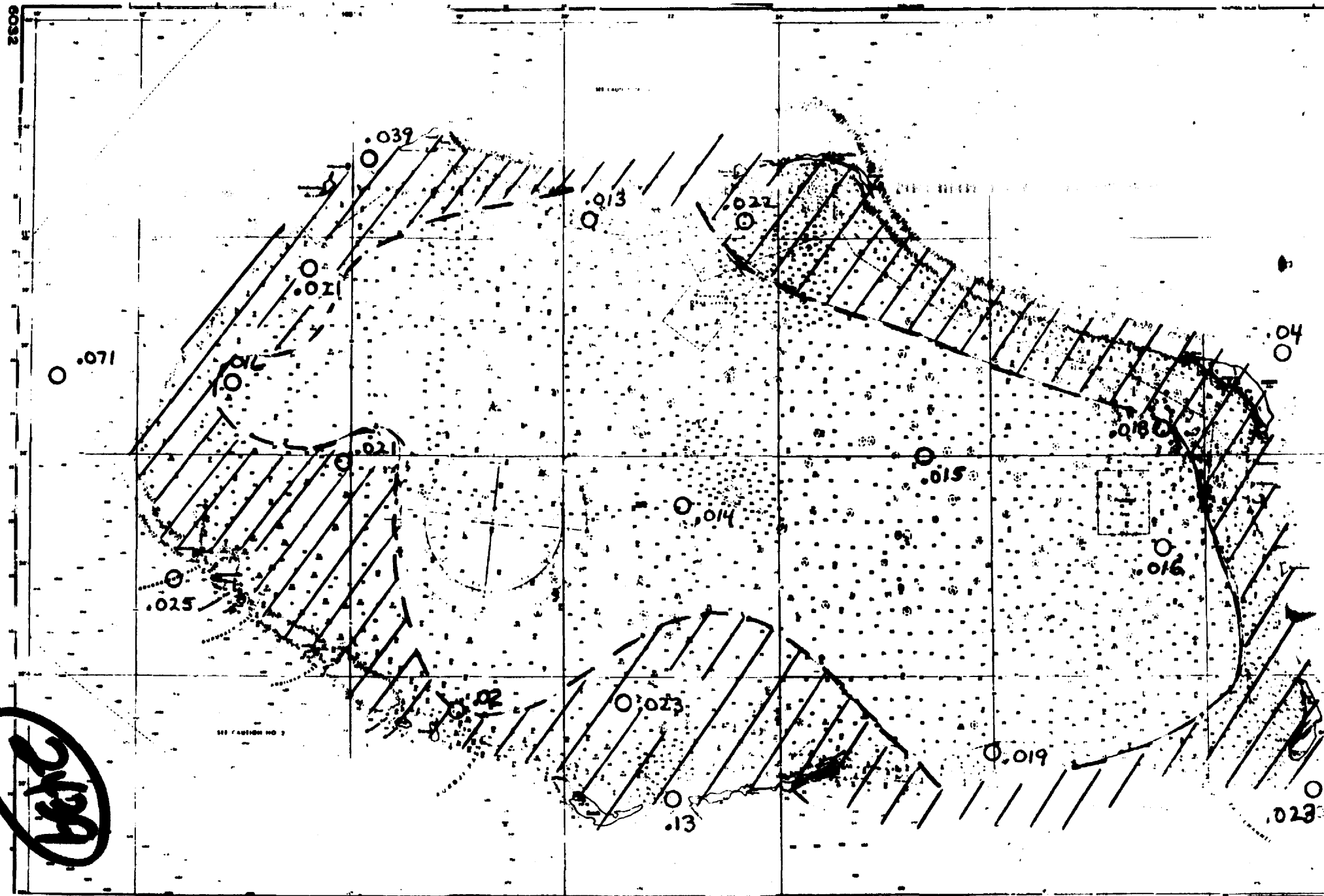
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Ratio Pu<sup>238</sup>/Pu<sup>239,240</sup> in waters of Bikini Atoll and nearby areas.

\* Livermore analysis

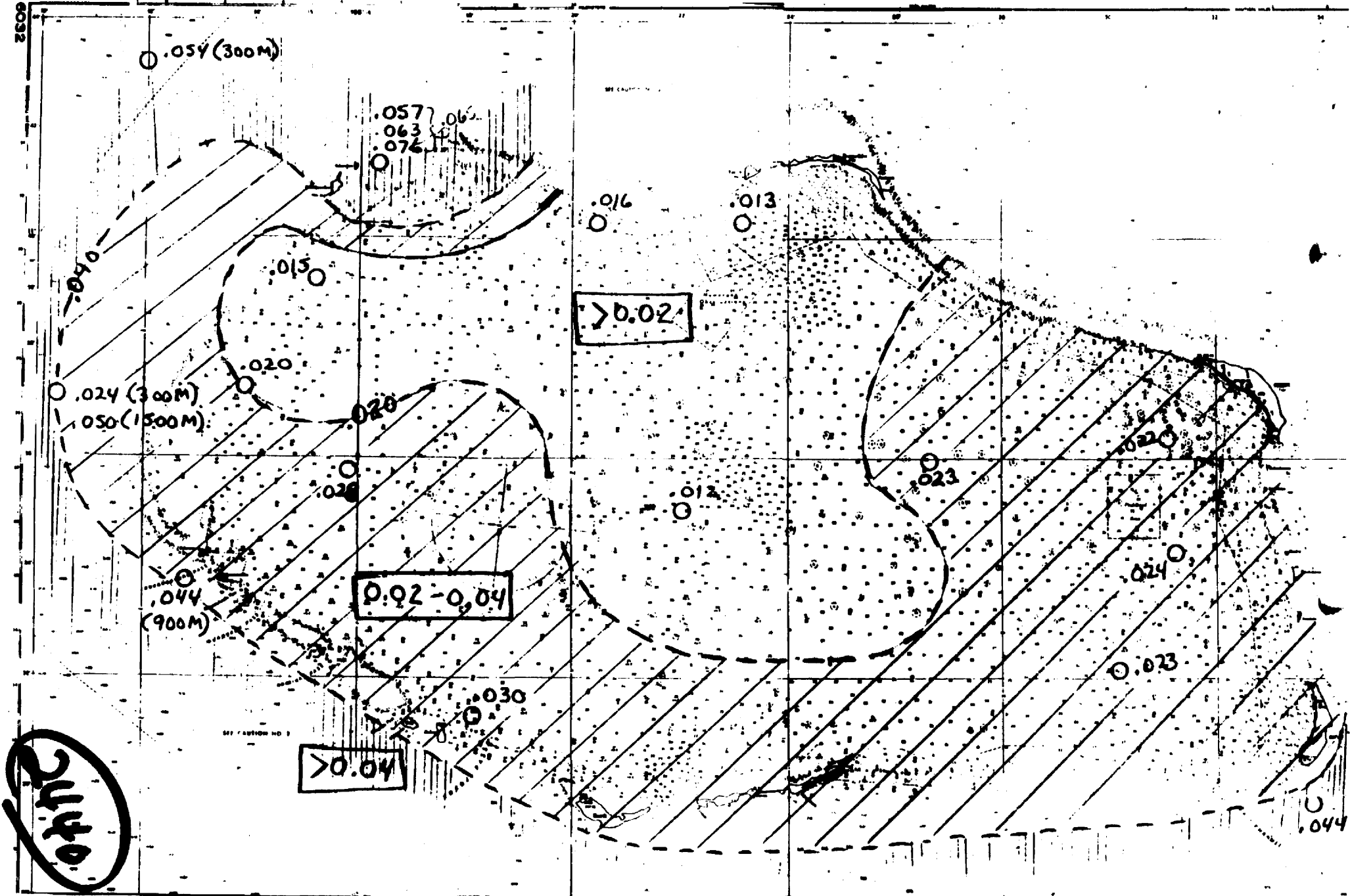
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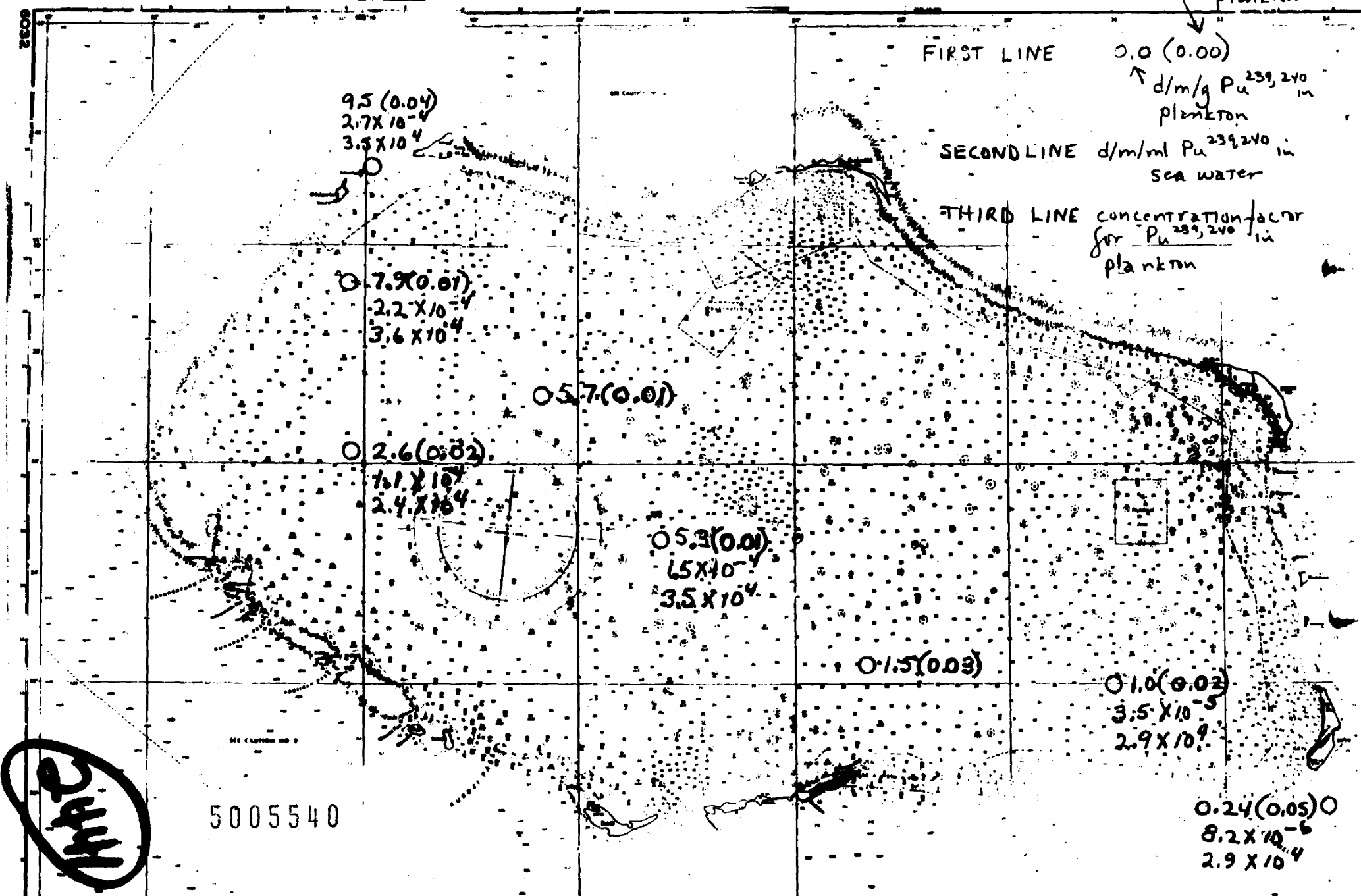
Ratio  $\text{Pu}^{238}/\text{Pu}^{239,240}$  in surface water of Bikini Atoll. Dashed line 0.02 ratio line. Center of lagoon had ratios all less than 0.02 and outer rim had values all greater than 0.02.

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Ratio  $Pu^{238}/Pu^{239,240}$  in deep water of Bikini Atoll and near-atoll areas as determined from available data.

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The amounts of  $Pu^{239,240}$  and the ratio  $Pu^{238}/Pu^{239,240}$  in plankton from Bikini Lagoon (first line); the amounts of  $Pu^{239,240}$  in water from the same stations (second line) and the concentration factors for  $Pu^{239,240}$  by the plankton (third line) based on the d/m/g in the plankton and d/m/g in the water