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Soluble and Particle-Associated Fallout Radionuclides in Mediterranean Water and Sediments

Hugh D. Livingston

Susan A. Casso

Vaughan T. Bowen

and

John C. Burke

Woods Hole Oceanographic Institution
Woods Hole, Massachusetts 02543

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Woods Hole Oceanographic Institution
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Summary From measurements in Mediterranean seawater and sediments, fallout ^{137}Cs, ^{90}Sr and $^{239,240}Pu$ behaved mostly as "soluble" nuclides tracing Mediterranean water movement, whereas ^{55}Fe and ^{241}Am behaved more as "particle-associated" nuclides and were relatively rapidly removed to the sediments. Patterns of nuclide distribution within sediments showed depths of penetration in the order ^{55}Fe > ($^{239,240}Pu$, ^{137}Cs) > ^{241}Am and were thought to result from biological mixing on nuclides whose arrival rates varied in the same order.

Résumé Après des mesures d'eau de mer et de sédiments méditerranéens, on voit que les inventaires de retombées radioactives ^{137}Cs, ^{90}Sr et $^{239,240}Pu$ réagissent plutôt comme des nucléides solubles en suivant les movements de l'eau en Méditerranée, alors que le ^{55}Fe et ^{241}Am réagissent plus comme des nucléides à "particules associées" et sont transféré relativement rapidement aux sédiments. On pense que les formes de distribution de nucléide à l'intérieur des sédiments avec un mode de pénétration dans l'ordre suivant ^{55}Fe>($^{239,240}Pu$, ^{137}Cs)> ^{241}Am, sont le résultat d'un mélange biologique des nucléides dont le taux d'arrivée varie dans le même ordre.

Text Distributions and inventories of fallout derived ^{137}Cs, ^{90}Sr, $^{239,240}Pu$ and ^{241}Am have been measured in Mediterranean sediment cores and water columns from locations representing the major hydrographic regimes which control Mediterranean circulation and biological productivity. ^{55}Fe distributions and inventories were measured in four sediment cores. These measurements were used to infer the processes which move these nuclides horizontally and vertically in Mediterranean water columns and, for the sedimentable nuclides, which deliver them

to and distribute them within the sediments.

Nuclides which behaved as "soluble" nuclides and are moved with such water transport processes as a) thermoconvective mixing in the N.W. Mediterranean, b) eastward inflow of Atlantic water, and c) westward flow of Levantine Intermediate Water, include ^{137}Cs, ^{90}Sr and, as previous studies have indicated, $^{239,240}Pu$ (1,2). Comparison of sediment inventories of ^{55}Fe and ^{241}Am with fallout delivery to the Mediterranean and examination of ^{241}Am water profiles relative to those of $^{239,240}Pu$ lead to the conclusion that these nuclides appear to have behaved as "particle-associated" nuclides and have been moved vertically towards and into the sediments in association with sinking particles. Compared to their proportions relative to plutonium in integrated global fallout, ^{241}Am and ^{55}Fe were enriched in the sediments by factors of 4-6 and 6-14 respectively. The lower enrichment of ^{241}Am than of ^{55}Fe in the sediments was not thought to result from a differential rate of sedimentation but in consequence of the delay between Pu and ^{55}Fe delivery and the in situ production of ^{241}Am by ^{241}Pu decay from Pu isotopes throughout the water columns. Once produced, ^{241}Am may well be removed to the sediments at similar rates to those of ^{55}Fe and possibly in association with the same population of sinking particles.

The distribution patterns of $^{239,240}Pu$, ^{137}Cs, ^{241}Am and ^{55}Fe within the sediments seem to depend upon the relative rates of arrival of each nuclide and the extent of biological mixing. Sediments underlying biologically more productive water - such as in the Western Mediterranean - showed patterns of deeper nuclide penetration. The ratios of $^{239,240}Pu$ and ^{137}Cs concentrations within sediment cores were generally rather constant. $^{55}Fe/^{239,240}Pu$ ratios increased smoothly with distance from the sediment/water interface, generally by about an order of magnitude. $^{241}Am/^{239,240}Pu$ ratios, in contrast, mostly decreased with distance from this interface. The patterns of the depth of nuclide penetration, $^{55}Fe > (^{239,240}Pu, ^{137}Cs) > ^{241}Am$ are believed to result from the effects of biological mixing, at rates and to depths which vary according to sediment organism populations (3), on sediment nuclide inventories which accumulated at rates which varied in the same order.

References

1. Noshkin, V. E. and V. T. Bowen. "Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments." In Radioactive Contamination of the Marine Environment, IAEA (Vienna), (1973), 671-686.
2. Livingston, H. D., V. T. Bowen and J. C. Burke. "Fallout radionuclides in Mediterranean sediments." Rapp. Comm. int. Mer Médit. 24, 3, (1977), 37-40.
3. Guinasso, N. L., Jr. and D. R. Schink. "Quantitative estimates of biological mixing rates in abyssal sediments." J. Geophys. Res., 80,(1975), 3032-3043.

RADIONUCLIDE INVENTORIES IN MEDITERRANEAN WATER COLUMNS

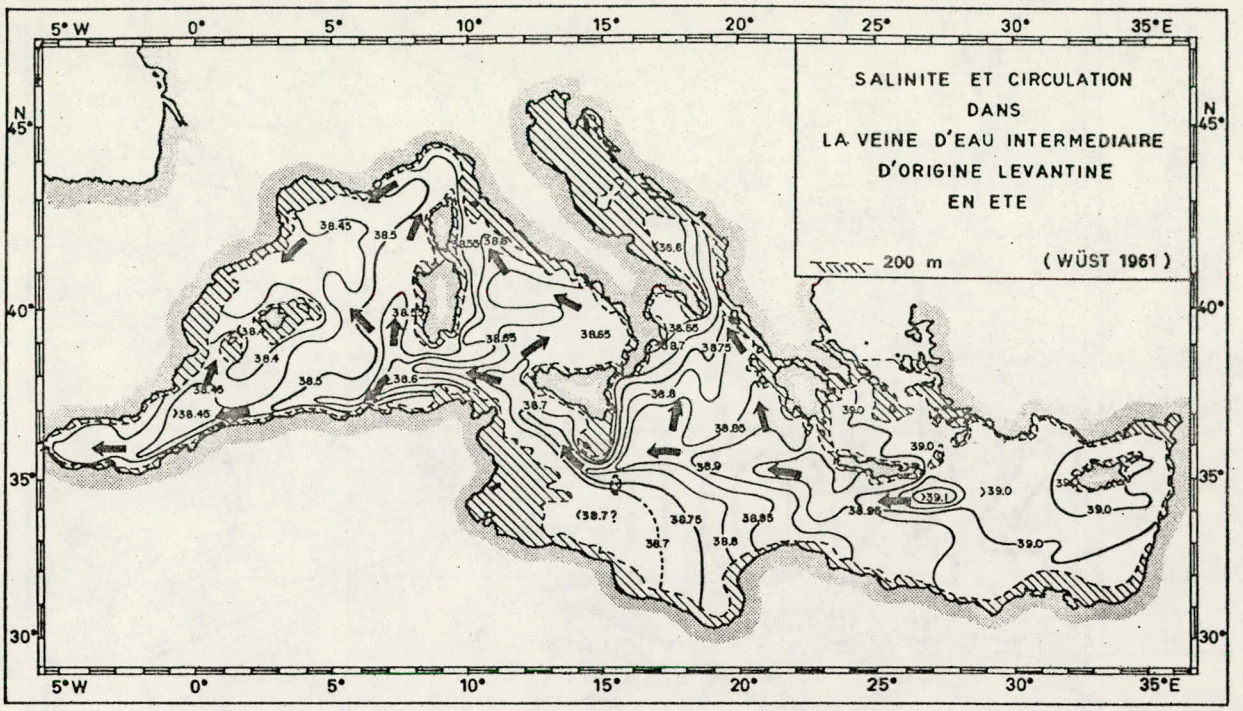
mCi/km^2 (FIGURE IN BRACKETS IS PERCENTAGE FALLOUT DELIVERY)

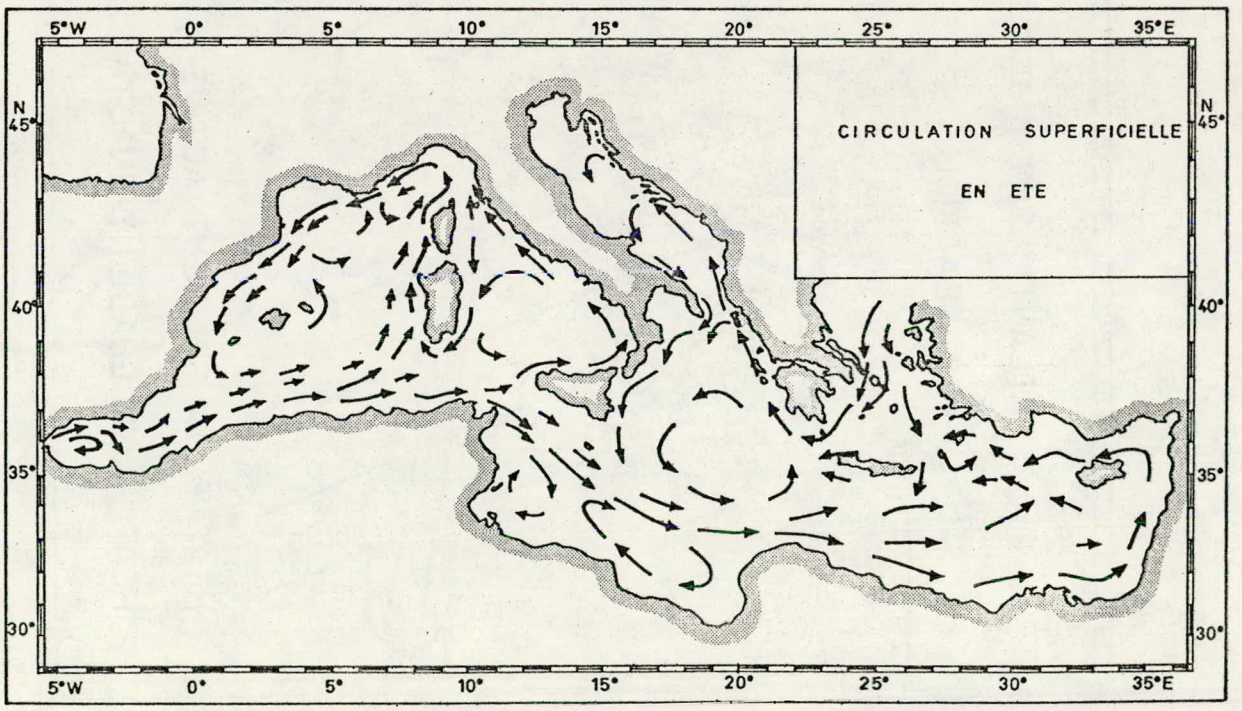
	WESTERN MEDITERRANEAN					BALEARIC SEA		S. IONIAN SEA	AVERAGE MEDITERRANEAN DELIVERY (37° N)
	METEOR 33 36°00'N 04°00'W FEB. 1974 1300 m	CH 121/1363 36°07.8'N 02°46.8'W MAY 1975 1794 m	METEOR 33 36°16'N 02°01'W FEB. 1974 1850 m	METEOR 33 37°00'N 00°01'E FEB. 1974 2660 m	METEOR 33 37°30'N 02°00'E FEB. 1974 2700 m	CH 121/1361 41° 12.4'N 5°52.3'E MAY 1975 2612 m	AII 49/1489 39° 58'N 06° 42.6'E MAY 1969 2725 m	CH 121/1359 35°56.4'N 17°55.8'E MAY 1975 4076 m	
^{137}Cs	85 (77%)	91 (82%)	88 (79%)	118 (106%)	104 (94%)	97 (87%)	127 (114%)	232 (209%)	111
^{90}Sr		58 (77%)				65 (84%)	90 (117%)	153 (199%)	77
$^{239,240}Pu$		1.07 58%				1.50 (81%)	1.88 (101%)	2.63 (141%)	1.86
^{241}Am		0.16 (42%)				0.17 (46%)	—	0.36 (97%)	0.37

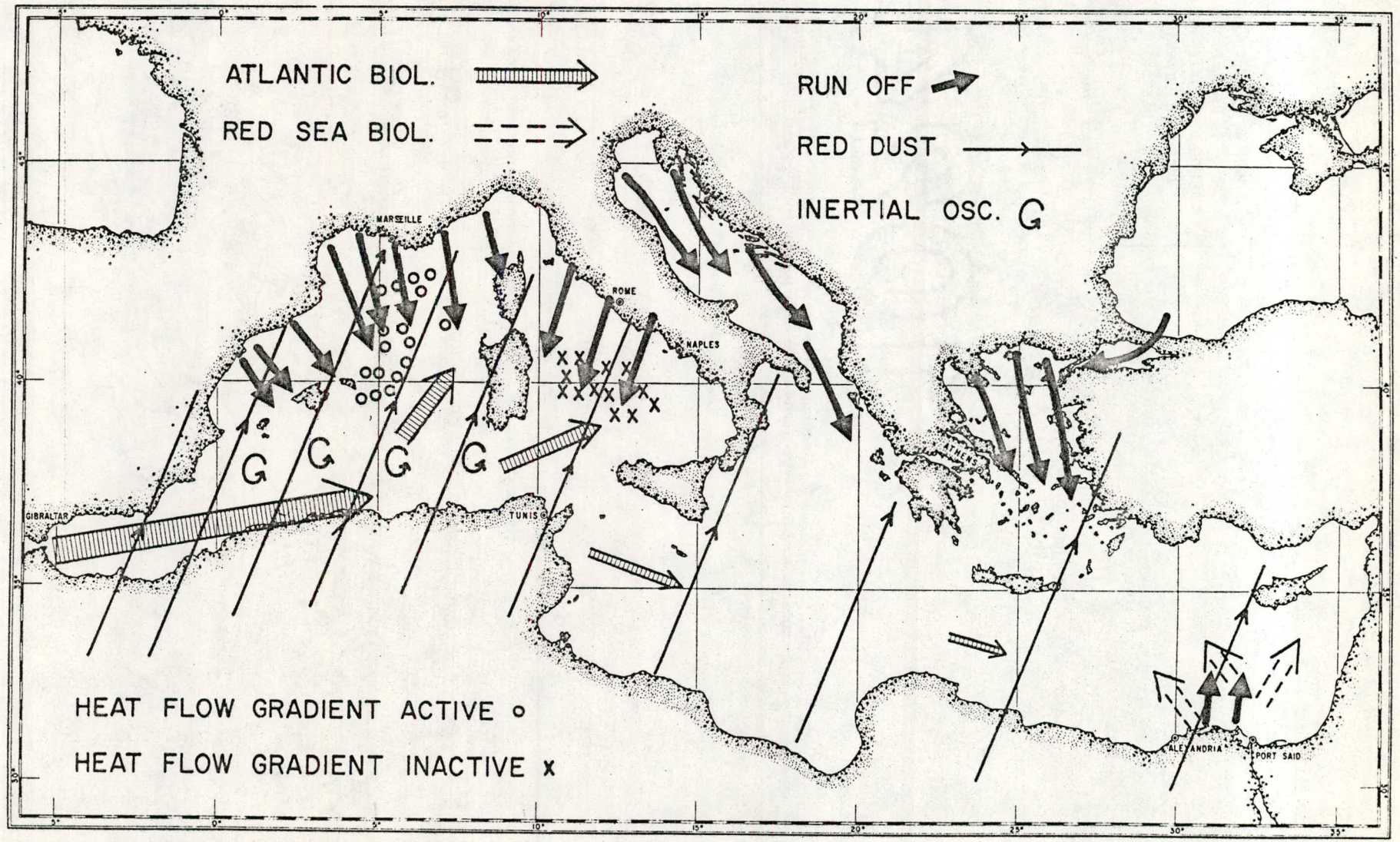
RADIONUCLIDE INVENTORIES IN MEDITERRANEAN SEDIMENTS

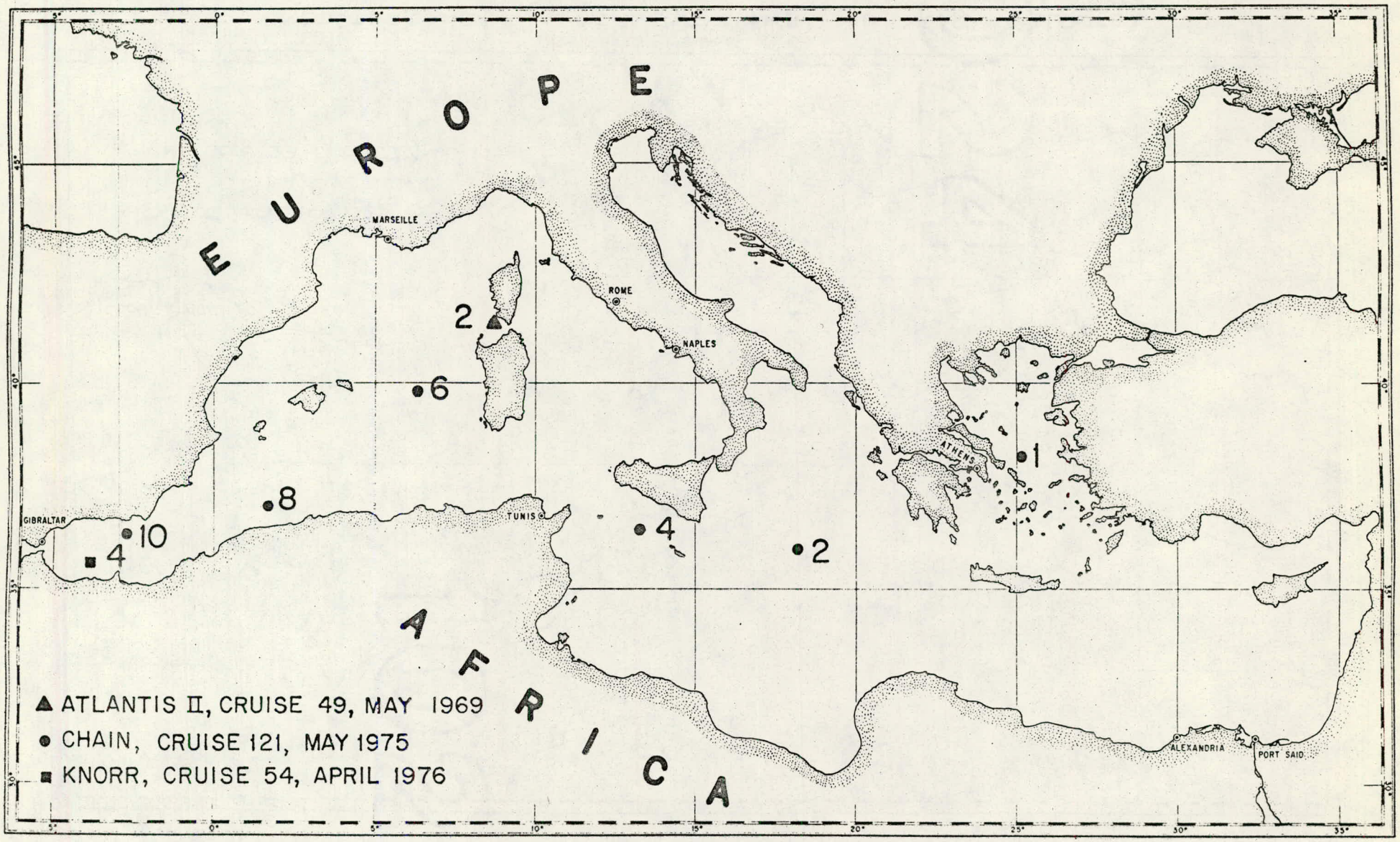
mCi/km^2 (FIGURE IN BRACKETS IS PERCENTAGE FALLOUT DELIVERY)

	ALBORAN SEA	ALGERIAN BASIN	BALEARIC SEA	IONIAN SEA	S. IONIAN SEA	AEGEAN SEA	AVERAGE MEDITERRANEAN DELIVERY (37° N)
	CH121/10 36°06.9'N 2° 48.1'W MAY 1975 1771 m	CH121/8 36°54.5'N 1°36.5'E MAY 1975 2740 m	CH121/6 39°44.2'N 6°21.2'E MAY 1975 2829 m	CH121/4 36°22.1'N 13°46.4'E MAY 1975 448 m	CH121/2 35°51.8'N 18°01.7'E MAY 1975 4043 m	CH121/1 38°16.4'N 24°57.2'E MAY 1975 725 m	
^{137}Cs	4.1 (4%)	6.3 (6%)	2.3 (2%)	7.1 (6%)	3.3 (3%)	3.2 (3%)	111
$^{239,240}Pu$	0.18 (10%)	0.15 (8%)	0.06 (3%)	0.18 (10%)	0.07 (4%)	0.06 (3%)	1.86
^{241}Am	0.21 (57%)	0.12 (32%)	—	0.23 (62%)	0.06 (16%)	0.06 (16%)	0.37
^{55}Fe	16.7 (75%)	24.8 (111%)	—	19.4 (87%)	5.5 (25%)	—	22.3

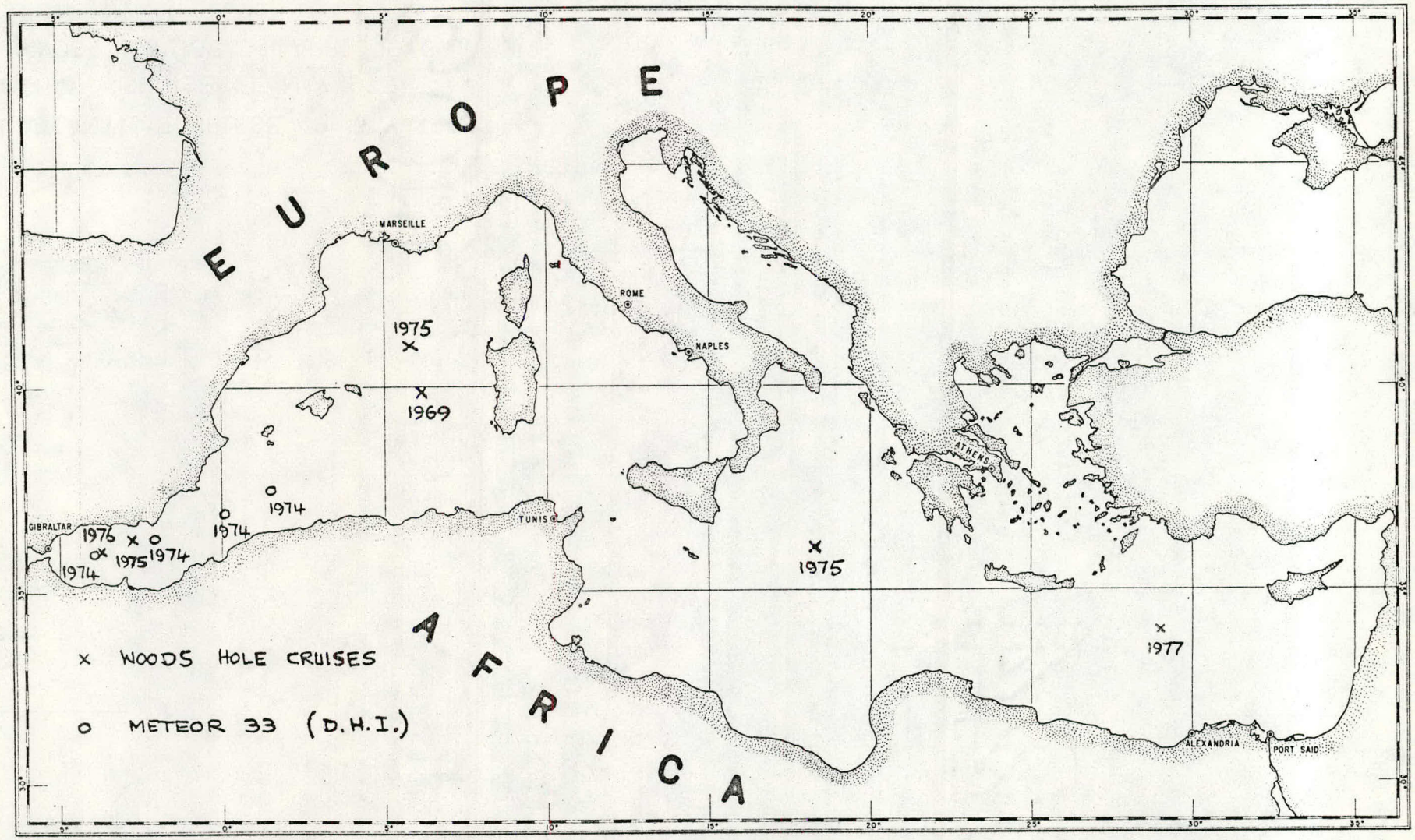

SALINITE ET CIRCULATION
DANS
LA VEINE D'EAU INTERMEDIAIRE
D'ORIGINE LEVANTINE
EN ETE
200 m
(WÜST 1961)
5°W
0°
5°
10°
15°
20°
25°
30°
35°E
N
45°
40°
35°
30°


CIRCULATION SUPERFICIELLE
EN ETE
5°W
0°
5°
10°
15°
20°
25°
30°
35°E
N
45°
40°
35°
30°

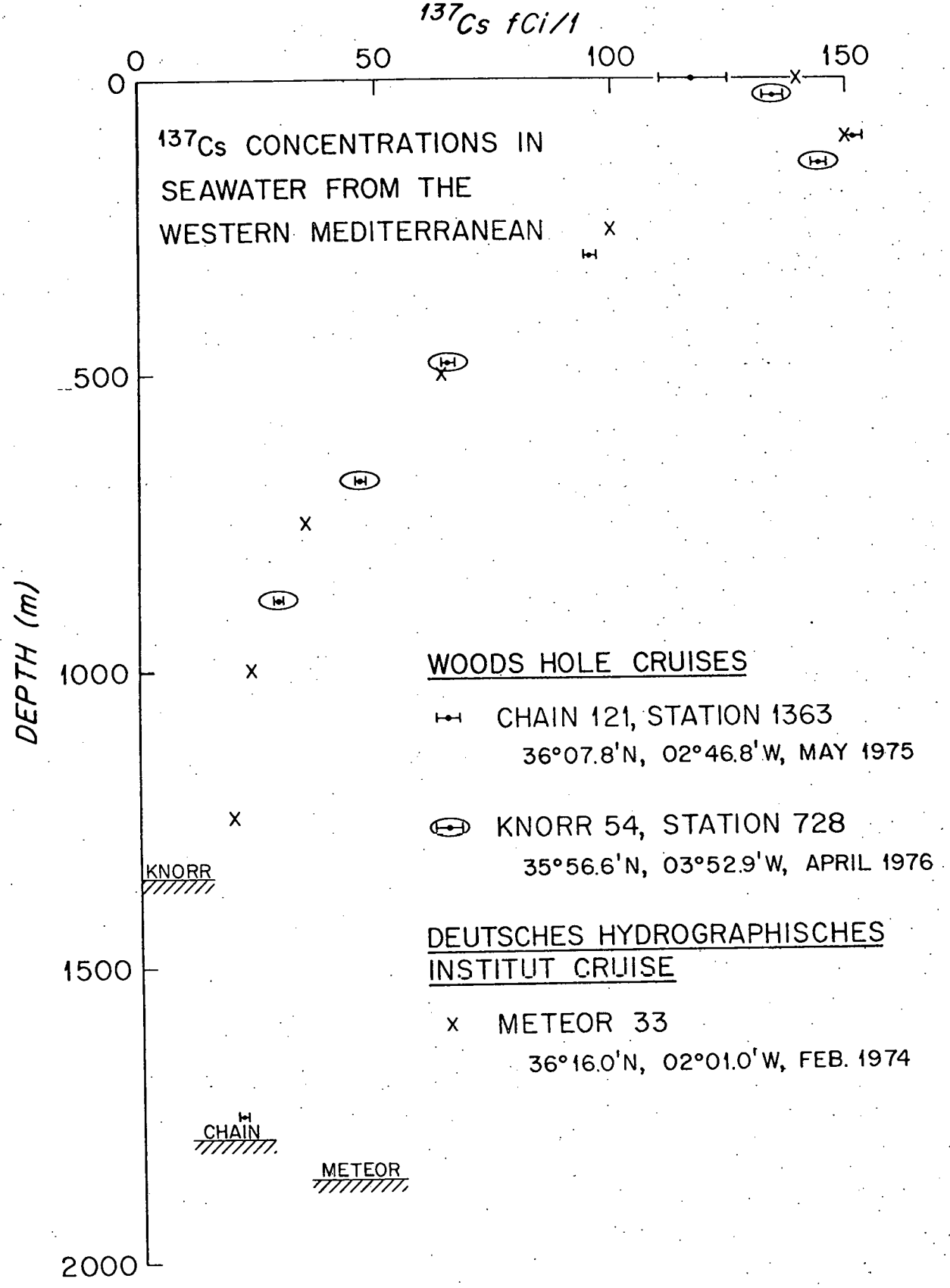

ATLANTIC BIOL.
RED SEA BIOL.
RUN OFF
RED DUST
INERTIAL OSC. G
MARSEILLE
ROME
NAPLES
ATHENS
GIBRALTAR
TUNIS
ALEXANDRIA
PORT SAID
G
G
G
G
HEAT FLOW GRADIENT ACTIVE o
HEAT FLOW GRADIENT INACTIVE x

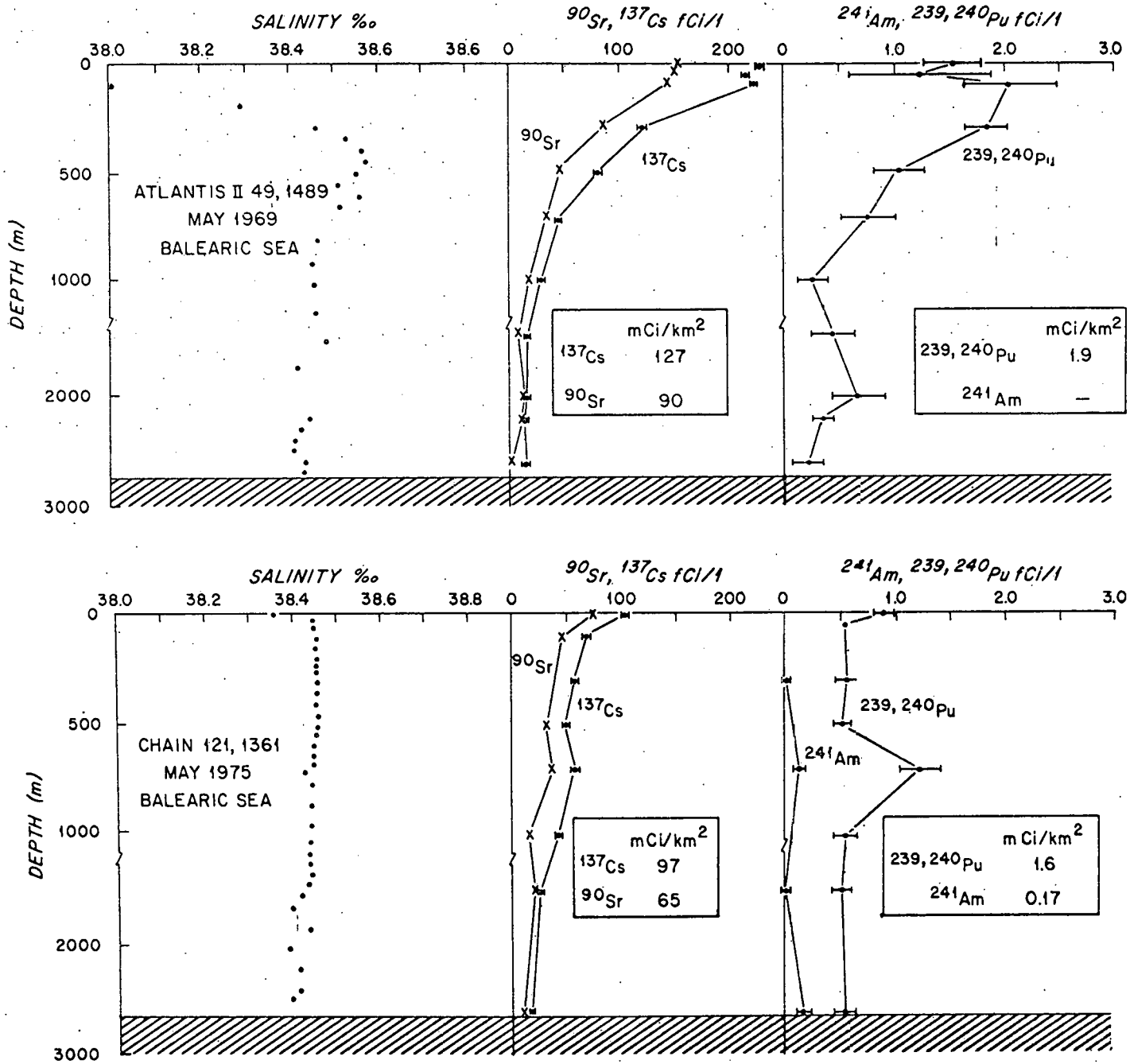


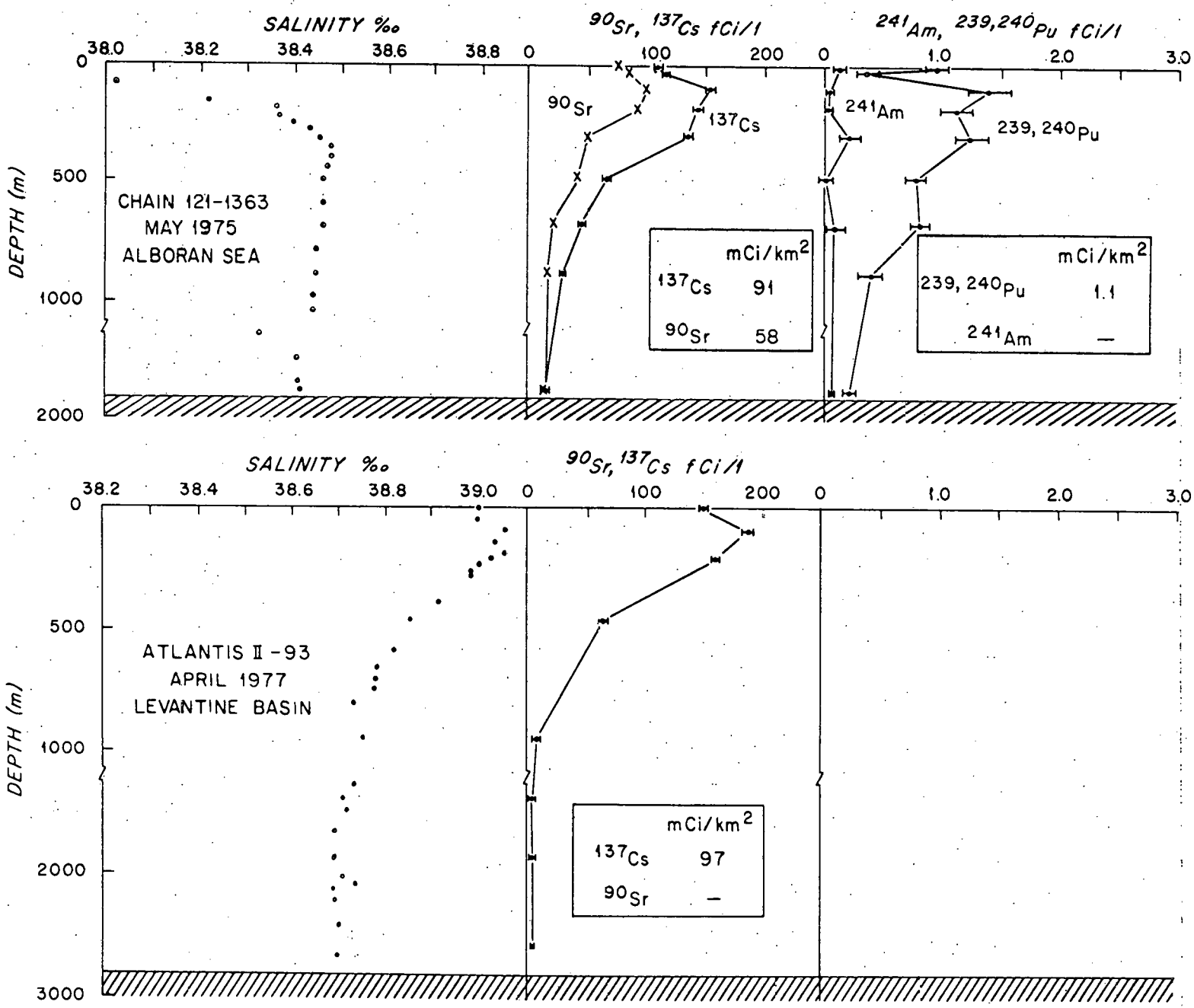
MEDITERRANEAN SEDIMENT CORE LOCATIONS AND NUMBERS

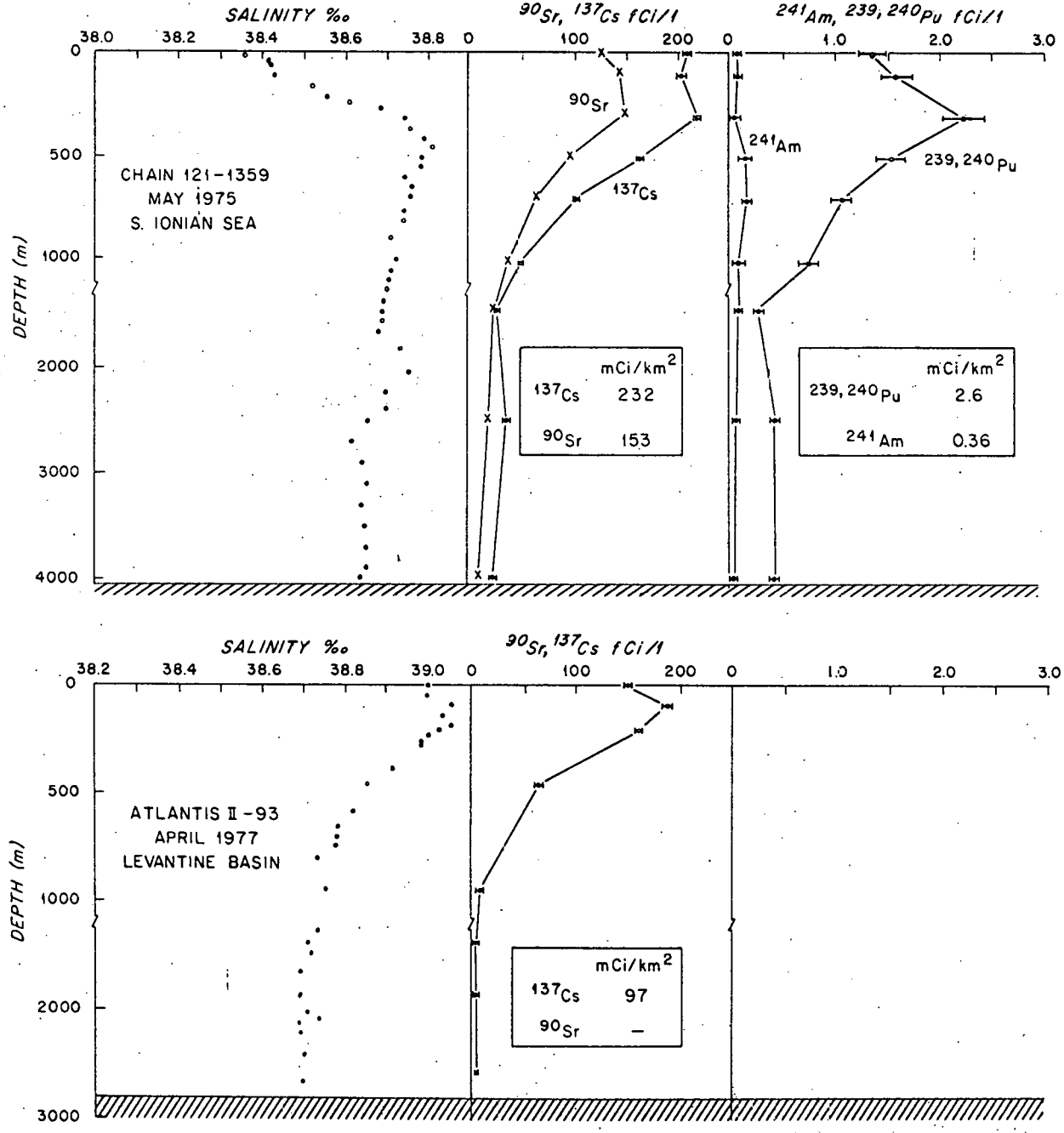


MEDITERRANEAN WATER STATIONS AND YEAR COLLECTED


^{137}Cs fCi/l
0
50
100
150
0
^{137}Cs CONCENTRATIONS IN SEAWATER FROM THE WESTERN MEDITERRANEAN
500
1000
1500
2000
DEPTH (m)
KNORR
CHAIN
METEOR
WOODS HOLE CRUISES
CHAIN 121, STATION 1363
36°07.8'N, 02°46.8'W, MAY 1975
KNORR 54, STATION 728
35°56.6'N, 03°52.9'W, APRIL 1976
DEUTSCHES HYDROGRAPHISCHES INSTITUT CRUISE
METEOR 33
36°16.0'N, 02°01.0'W, FEB. 1974

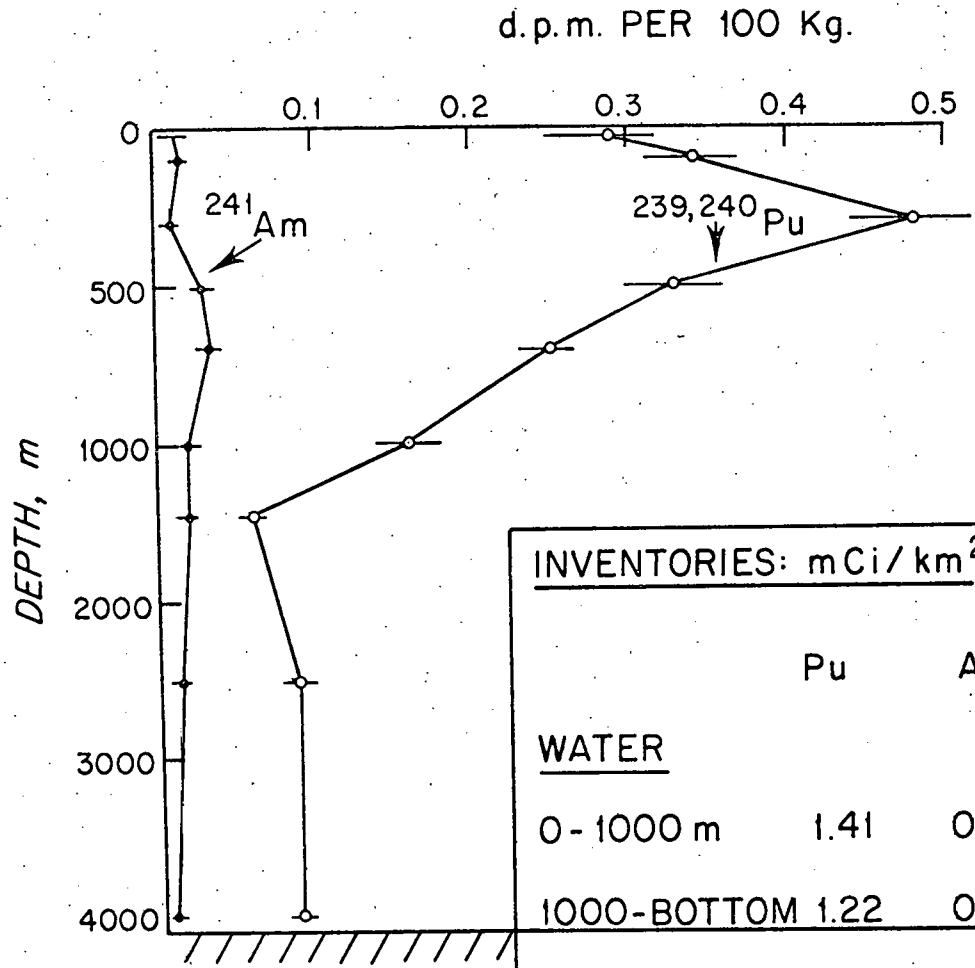

SALINITY ‰
38.0
38.2
38.4
38.6
38.6
^{90}Sr, ^{137}Cs fCi/l
0
100
200
^{241}Am, $^{239,240}Pu$ fCi/l
0
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
3000
ATLANTIS II 49, 1489
MAY 1969
BALEARIC SEA
^{90}Sr
^{137}Cs
$^{239,240}Pu$
mCi/km^2
^{137}Cs 127
^{90}Sr 90
mCi/km^2
$^{239,240}Pu$ 1.9
^{241}Am —
SALINITY ‰
38.0
38.2
38.4
38.6
38.8
^{90}Sr, ^{137}Cs fCi/l
0
100
200
^{241}Am, $^{239,240}Pu$ fCi/l
0
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
3000
CHAIN 121, 1361
MAY 1975
BALEARIC SEA
^{90}Sr
^{137}Cs
$^{239,240}Pu$
^{241}Am
mCi/km^2
^{137}Cs 97
^{90}Sr 65
mCi/km^2
$^{239,240}Pu$ 1.6
^{241}Am 0.17


SALINITY ‰
38.0
38.2
38.4
38.6
38.8
^{90}Sr, ^{137}Cs fCi/l
0
100
200
^{241}Am, $^{239,240}Pu$ fCi/l
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
^{90}Sr
^{137}Cs
^{241}Am
$^{239,240}Pu$
CHAIN 121-1363
MAY 1975
ALBORAN SEA
mCi/km^2
^{137}Cs 91
^{90}Sr 58
mCi/km^2
$^{239,240}Pu$ 1.1
^{241}Am —
SALINITY ‰
38.2
38.4
38.6
38.8
39.0
^{90}Sr, ^{137}Cs fCi/l
0
100
200
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
3000
ATLANTIS II -93
APRIL 1977
LEVANTINE BASIN
mCi/km^2
^{137}Cs 97
^{90}Sr —


SALINITY ‰
38.0
38.2
38.4
38.6
38.8
^{90}Sr, ^{137}Cs fCi/l
0
100
200
^{241}Am, $^{239,240}Pu$ fCi/l
0
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
3000
4000
CHAIN 121-1359
MAY 1975
S. IONIAN SEA
^{90}Sr
^{137}Cs
^{241}Am
$^{239,240}Pu$
mCi/km^2
^{137}Cs 232
^{90}Sr 153
mCi/km^2
$^{239,240}Pu$ 2.6
^{241}Am 0.36
SALINITY ‰
38.2
38.4
38.6
38.8
39.0
^{90}Sr, ^{137}Cs fCi/l
0
100
200
0
1.0
2.0
3.0
DEPTH (m)
0
500
1000
2000
3000
ATLANTIS II-93
APRIL 1977
LEVANTINE BASIN
mCi/km^2
^{137}Cs 97
^{90}Sr —

MEDITERRANEAN: CHAIN 121, STATION 1359, 5/7/75

Pu AND Am DISTRIBUTIONS AND INVENTORIES



Am/Pu x 100
4 ± 2
5 ± 2
2 ± 2
9 ± 4
14 ± 3
13 ± 7
32 ± 12
14 ± 6
11 ± 7
81 ± 6
0.13

INVENTORIES: mCi/km²

	Pu	Am
WATER		
0 - 1000 m	1.41	0.107
1000-BOTTOM	1.22	0.253
SEDIMENT:	0.07	0.06
TOTAL:	2.7	0.36

RADIONUCLIDE INVENTORIES IN MEDITERRANEAN WATER COLUMNS

mCi/km^2 (FIGURE IN BRACKETS IS PERCENTAGE FALLOUT DELIVERY)

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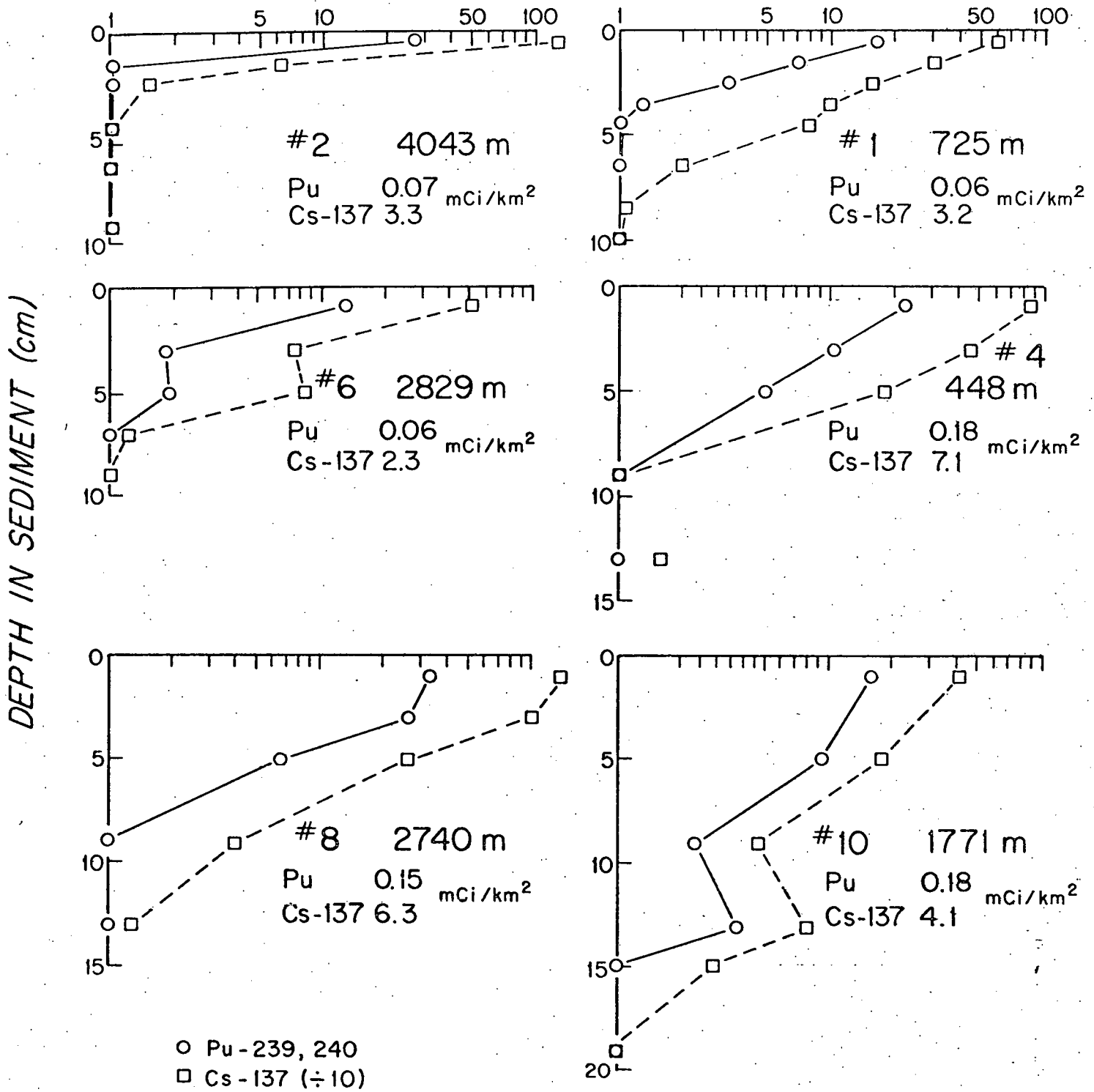
^{137}Cs	85	91	88	118	104	97	127	232	111
	(77%)	(82%)	(79%)	(·06%)	(94%)	(87%)	(114%)	(209%)	
^{90}Sr		58				65	90	153	77
		(77%)				(84%)	(117%)	(199%)	
$^{239,240}Pu$		1.07				1.50	1.86	2.63	1.86
		58%				(81%)	(101%)	(141%)	
^{241}Am		0.16				0.17	—	0.36	0.37
		(42%)				(46%)		(97%)	

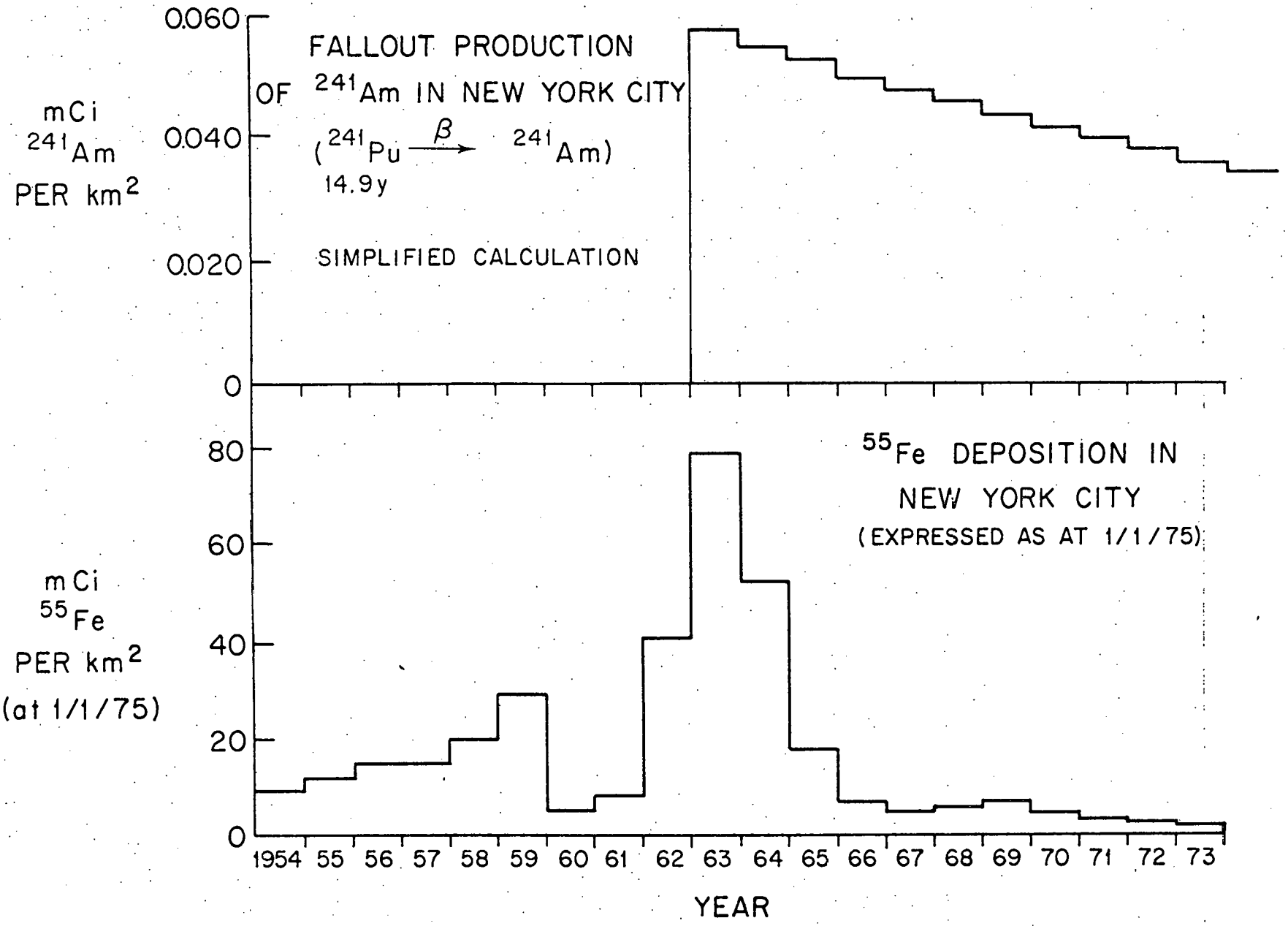


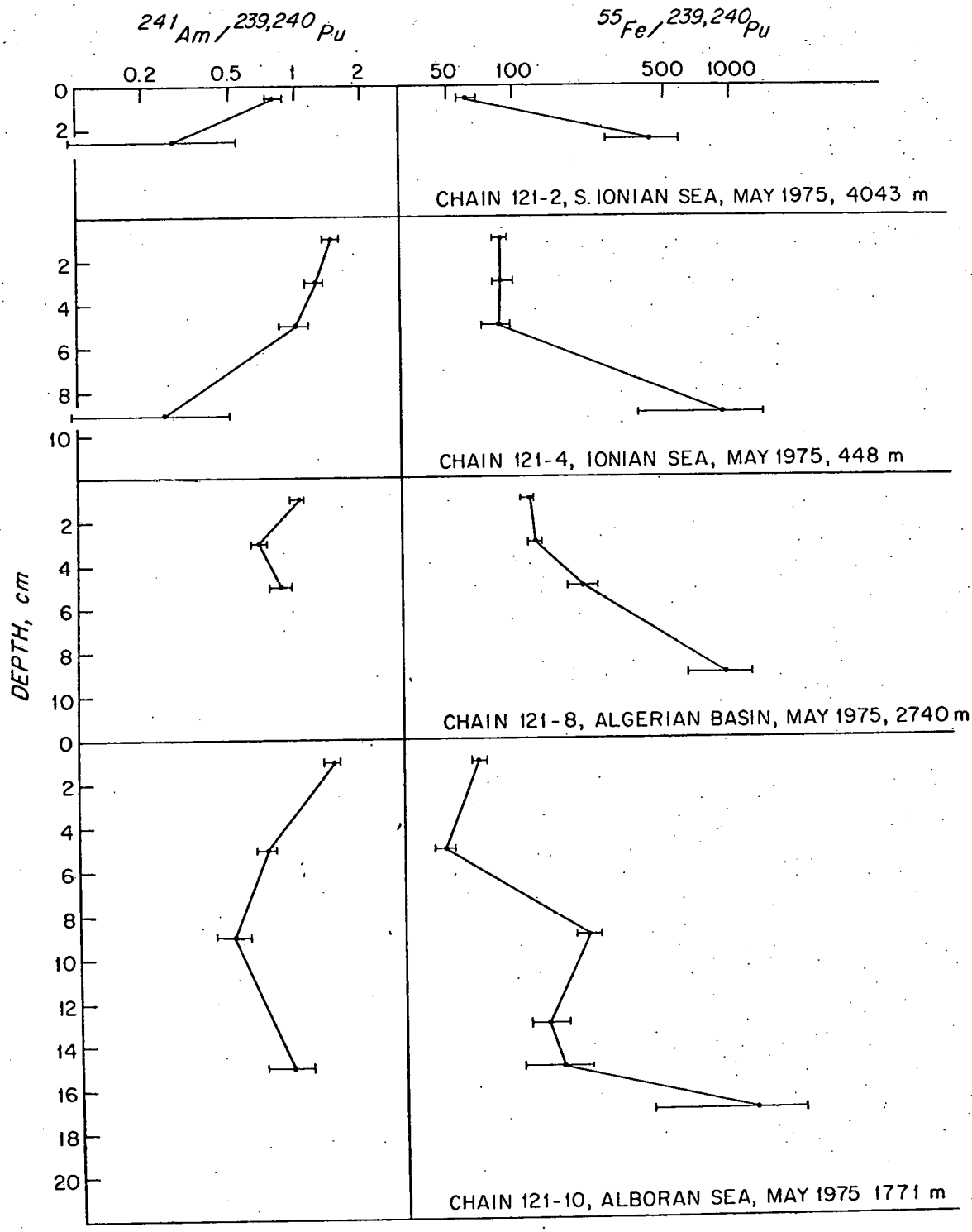
Pu, Cs-137 DISTRIBUTIONS IN MEDITERRANEAN SEDIMENT CORES

RADIONUCLIDE INVENTORIES IN MEDITERRANEAN SEDIMENTS

mCi/km^2 (FIGURE IN BRACKETS IS PERCENTAGE FALLOUT DELIVERY)

	ALBORAN SEA CH121/10 36°06.9'N 2° 48.1'W MAY 1975 1771 m	ALGERIAN BASIN CH121/8 36°54.5'N 1°36.5'E MAY 1975 2740 m	BALEARIC SEA CH121/6 39°44.2'N 6°21.2'E MAY 1975 2829 m	IONIAN SEA CH121/4 36°22.1'N 13°44.4'E MAY 1975 448 m	S. IONIAN SEA CH121/2 35°51.8'N 18°01.7'E MAY 1975 4043 m	AEGEAN SEA CH121/1 38°16.4'N 24°57.2'E MAY 1975 725 m	AVERAGE MEDITERRANEAN DELIVERY (37° N)
^{137}Cs	4.1	6.3	2.3	7.1	3.3	3.2	111
	(4%)	(6%)	(2%)	(6%)	(3%)	(3%)	
$^{239,240}Pu$	0.18	0.15	0.06	0.18	0.07	0.06	1.86
	(10%)	(8%)	(3%)	(10%)	(4%)	(3%)	
^{241}Am	0.21	0.12	—	0.23	0.06	0.06	0.37
	(57%)	(32%)		(62%)	(16%)	(16%)	
^{55}Fe	16.7	24.8	—	19.4	5.5	—	22.3
	(75%)	(111%)		(87%)	(25%)		


FALLOUT PRODUCTION
OF ^{241}Am IN NEW YORK CITY
(^{241}Pu $\xrightarrow[14.9y]{\beta}$ ^{241}Am)
SIMPLIFIED CALCULATION
mCi
^{241}Am
PER km^2
0.060
0.040
0.020
0
^{55}Fe DEPOSITION IN
NEW YORK CITY
(EXPRESSED AS AT 1/1/75)
mCi
^{55}Fe
PER km^2
(at 1/1/75)
80
60
40
20
0
1954 55 56 57 58 59 60 61 62 63 64 65 66 67 68 69 70 71 72 73
YEAR



241Am/239,240Pu and 55Fe/239,240Pu RATIO PROFILES IN MEDITERRANEAN SEDIMENTS