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

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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.

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	91	88	118	104	97	127	232	111
	(77%)	(82%)	(79%)	(106%)	(94%)	(87%)	(114%)	(209%)	
^{90}Sr		58				65	90	153	77
		(77%)				(84%)	(117%)	(199%)	
$^{239,240}Pu$		1.07				1.50	1.88	2.63	1.86
		58%				(81%)	(101%)	(141%)	
^{241}Am		0.16				0.17	—	0.36	0.37
		(42%)				(46%)		(97%)	

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°44.4'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	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%)		

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Paper presented by H.D. Livingston (USA)

Discussion

R. CHESSELET: 1. Could you compare with North Atlantic?
2. To what content is Mediterranean specific in terms of the geochemistry of ^{55}Fe, Pu, Am, regarding the North Atlantic situation?

H.D. LIVINGSTON: 1. When comparisons are made between water and sediment distributions of ^{55}Fe, $^{239,240}Pu$ and ^{241}Am in the North Atlantic Ocean and the Mediterranean Sea, the data support the conclusion that biogeochemical processes affecting $^{239,240}Pu$ and ^{241}Am distributions differ substantially. $^{239,240}Pu$ behaves much more as if in true "solution" in the Mediterranean and is distributed mainly by the water circulation. Hence, compared with North Atlantic sediments taken at comparable depths and times, Mediterranean sediments show $^{239,240}Pu$ inventories which are smaller fractions of fallout delivery to the sea surface. On the other hand, Mediterranean water is depleted and sediments enriched in ^{241}Am relative to comparable situations in the North Atlantic. It appears that ^{241}Am, when produced in the water column by ^{241}Pu decay, is rapidly transferred to Mediterranean sediments at rates considerably faster than those which apply in the North Atlantic.
2. It is not possible to argue from our data whether the geochemistry of ^{55}Fe is significantly different in the Mediterranean and North Atlantic. Comparison of ^{55}Fe inventories (expressed in terms of fraction of expected

fallout delivery) in Mediterranean sediments with those measured in North Atlantic sediments collected at comparable times and depths, shows that the data for the Mediterranean sediments are not distinguishable from those for North Atlantic sediments. However, for sediments collected at depths shallower than 3000 m, the inventories range around the amounts predicted to have been delivered in fallout. It is, therefore, possible to argue that sinking rates could differ for ^{55}Fe. However, the samples from which this body of data was derived, were collected too long after the input of fallout from atmospheric nuclear weapons testing to be able to distinguish different rates of delivery of ^{55}Fe to sediments.

R. FUKAI: Comment.
I think only large difference between your data and our data for caesium--137 is found in the Ionian Basin. Whereas your inventory of caesium-137 in this area is approximately 200% of fallout delivery, our inventory shows about 95%. I believe this difference is real and due mainly to the different locations of the stations.

H.D. LIVINGSTON: What are the coordinates and collection date for your Ionian Sea station?

After this last question, other comments follow, which were not recorded.