Artificial Radionuclides Database in the Pacific Ocean: HAM Database

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The database “Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas”, or HAM database, has been created. The database includes ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ concentration data from the seawater of the Pacific Ocean and its marginal seas with some measurements from the sea surface to the bottom. The data in the HAM database were collected from about 90 literature citations, which include published papers; annual reports by the Hydrographic Department, Maritime Safety Agency, Japan; and unpublished data provided by individuals. The data of concentrations of ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ have been accumulating since 1957–1998. The present HAM database includes 7737 records for ^{137}Cs concentration data, 3972 records for ^{90}Sr concentration data, and 2666 records for $^{239,240}\text{Pu}$ concentration data. The spatial variation of sampling stations in the HAM database is heterogeneous, namely, more than 80% of the data for each radionuclide is from the Pacific Ocean and the Sea of Japan, while a relatively small portion of data is from the South Pacific. This HAM database will allow us to use these radionuclides as significant chemical tracers for oceanographic study as well as the assessment of environmental affects of anthropogenic radionuclides for these 5 decades. Furthermore, these radionuclides can be used to verify the oceanic general circulation models in the time scale of several decades.

KEYWORDS: database, artificial radionuclides, marine environment, ^{90}Sr , ^{137}Cs , plutonium

DOMAINS: isotopes in the environment, environmental monitoring, oceans chemistry

INTRODUCTION

Chemical tracers are useful tools to understand the physical and biogeochemical processes in the ocean. Tritium, CFCs, and others have been used as transient tracers of physical processes, such as mixing and advection, in the ocean. The distributions of chemical tracers have been used for validation of global oceanic circulation models.

Artificial radionuclides (^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$) that have been injected into the ocean by global fallout, released from the nuclear processing plants and others, might be also significant transient tracers. Although the concentrations of these radionuclides in seawater have been determined for the assessment

of environmental affects of radionuclides for these 5 decades, they have not been used for oceanographic purposes. One of the exceptions are the data obtained from GEOchemical Ocean SECTIONS Study (GEOSECS) in the 1970s[1].

We think a large number of the concentrations of these radionuclides in seawater, which were published in variety of research fields and governmental reports, might be useful for oceanographic studies as well as the study of artificial radionuclides in the marine environment. We, then, constructed the database “Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas”, or HAM database. The database includes ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ concentration data from the seawater in the Pacific Ocean and its marginal seas with some measurements from the sea surface to the bottom. The data of concentrations of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ have been accumulating since 1957–1998. The Marine Environment Laboratory of the IAEA has also created and maintained a database on marine radioactivity, the Global Marine Radioactivity Database (GLOMARD)[2]. GLOMARD consists of radionuclide concentration data for seawater, sediment, biota, and suspended matter. During the first phase of GLOMARD, the work concentrated on the Sea of Japan and the Barents and Kara Seas in the arctic in connection with the investigation of radioactive waste dumping sites and their impact on the marine environment. Some of the data in the GLOMARD and our HAM database may overlap, especially those from the Sea of Japan and western North Pacific. The HAM database, however, focuses on the radionuclides concentration in seawater in the Pacific Ocean and its marginal seas, so further data exchange between the two databases will make the both databases more powerful. In this paper, we present the HAM database in detail and discuss its features.

OBJECTIVES

One of the major objectives of the HAM database is to reconstruct the history of artificial radionuclides in the oceans. Since our interest is mainly in the Pacific Ocean and its marginal seas, our effort to collect data sources was concentrated in these sea areas. We expect that the artificial radionuclides are good transient tracers of oceanographic studies in the Pacific Ocean and its marginal seas. ^{137}Cs , which is a soluble radionuclide, is a good tracer for water mass movement. $^{239,240}\text{Pu}$, which is a typical particle reactive radionuclide, could also be used for biogeochemical studies of trace metals, including radionuclides, in the ocean. The HAM database can be applied to validate the results of the 3D simulations using ocean global circulation models.

Some results of the studies using the HAM database are already published. A typical example is a study on the formation of deep waters in the Sea of Japan. It provided new information that the formation of deep waters occurred in the 1990s based on the analysis of the ^{137}Cs concentration data in the database (Fig. 2 in [3]). The temporal variation of the ^{137}Cs concentration in the surface water in the North Pacific during these 4 decades was examined by Hirose and Aoyama (Figs. 1, 2, 3, 6, and 12 in [4]). Aoyama and Hirose also examined the temporal variation of the ^{137}Cs inventory in the North Pacific, then, increases of the ^{137}Cs inventory at the lower latitude of 10–20° N in the North Pacific were found in the 1970s and 1980s. It is concluded that surface and subsurface southward transports were considered as the source of this increasing ^{137}Cs inventory (Figs. 3 and 4 in [5]). In the studies of temporal variation of ^{137}Cs and $^{239,240}\text{Pu}$ in the Pacific Ocean using an oceanic general circulation model, the HAM database was also used to verify the results of the model calculation by Tsumune et al.[6,7].

DATABASE CONSTRUCTION

Sources of the Data in this Database

We collected about 90 literature citations to construct the HAM database during these 7 years. As shown in Table 1, the sources of data are as follows:

1. Geochemical Research Department, Meteorological Research Institute, JAPAN (hereafter GRD/MRI) data from the scientific papers in the Pacific Ocean since 1960[8,9,10,11,12,13,14,15,16,17,18,19,20,21,22].
2. Health and Safety Laboratory, now Environmental Measurement Laboratory, USA (hereafter HASL/EML) reports[23,24,25].
3. Scientific papers, reports, and books 1960–1995[26,27,28,29,30,31,32,33,34,35,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52,53,54,55,56,57,58,59,60].
4. The annual reports of the Hydrographic Department/Maritime Safety Agency/Japan during the period from 1963–2000[61,62,63,64,65,66,67,68,69,70,71,72,73,74,75,76,77,78,79,80,81,82,83,84,85,86,87,88,89,90,91,92,93].
5. Unpublished data obtained from the authors[94,95,96,97,98,99,100].

The numbers of data of each radionuclide from each source are also shown in Table 1.

This database includes 7737 records for ^{137}Cs concentration data, 3972 records for ^{90}Sr concentration data, and 2666 records for $^{239,240}\text{Pu}$ concentration data. For the ^{137}Cs concentration data, the major contributors are Folsom's work in the 1960s and the 1970s and the long-term work by the Hydrographic Department, Maritime Safety Agency, Japan during 1963–2000. About 3000 of the total 7737 data of the ^{137}Cs concentration are available from HASL/EML reports[23,24,25]. The Hydrographic Department, Maritime Safety Agency, Japan contributed about 1000 data of the ^{137}Cs concentration during 1963–1998. For the ^{90}Sr concentration data, the major contributors are Livingston's work in the GEOSECS project and the long-term work by the Hydrographic Department, Maritime Safety Agency during 1963–1998. About 700 of the total 3972 data of the ^{90}Sr concentration are available from Livingston et al.[39]. The Hydrographic Department, Maritime Safety Agency contributed about 1000 data of ^{90}Sr concentration during 1963–1998. For the $^{239,240}\text{Pu}$ data, the major contributors are Livingston et al.[39], Wong et al.[60], and Noshkin et al.[56]. The Hydrographic Department, Maritime Safety Agency also contributed about 500 data of the $^{239,240}\text{Pu}$ concentration during 1989–2000.

Handling of Original Data in the HAM Database

All of the data in the publications/reports are treated as follows:

- a. Input original data in original unit with data originator name.
- b. The values in the original unit are converted to the values in the SI unit, Bq m^{-3} .
- c. Create Microsoft Access file. Microsoft Access is a desktop database software that operates in a user desktop environment such as Microsoft Windows 98, Windows Me, or Windows XP.
- d. Create PostgreSQL file. PostgreSQL is an object-relational database management system (ORDBMS) based on POSTGRES, Version 4.2, developed at the University of California at Berkeley Computer Science Department. PostgreSQL is an open-source descendant of this original Berkeley code. It supports SQL92 and SQL99 and offers many modern features.
- e. Web database access for data search.

Throughout the handling of the data, the data originator name, the name of the reference literature, and original values with original unit were kept. When counting errors were reported in the original literature, those were also kept throughout the handling. To identify the original literature, a unique code, of which field name in the database is MRIREFNO, is given for each literature and added to each record. For each profile, a unique number, of which field name is profileID, is also given. A code to identify that the data are in a certain profile, of which field name is PS, is given as P. A code S at the field PS is given for the concentration data of the surface water only without deeper layer sampling. Keeping these original and created data throughout the process allows us to maintain the database more easily. Users of the database can also see the original values and unit together with the values in SI by the database creator.

TABLE 1

Data Sources and Number of the Data Archived in the Database for Each Radionuclide

MRIREFNO	¹³⁷ Cs	⁹⁰ Sr	Pu	Ref.	MRIREFNO	¹³⁷ Cs	⁹⁰ Sr	Pu	Ref.
Aoyama1995	67	0	39	[8]	msa1992	64	68	24	[85]
Aoyama2001	34	0	0	[9]	msa1993	64	64	13	[86]
Bourlat1992	3	0	0	[26]	msa1994	64	63	13	[87]
Bourlat1995	0	0	22	[27]	msa1995	150	152	102	[88]
Bourlat1996	54	54	51	[28]	msa1996	113	114	69	[89]
Broecker1966	49	43	0	[29]	msa1997	96	98	55	[90]
Broecker1968	123	78	0	[30]	msa1998	89	90	63	[91]
Folsom1960a	12	0	0	[31]	msa1999	86	87	63	[92]
Folsom1960b	4	0	0	[32]	msa2000	101	101	63	[93]
Folsom1968	1141	0	0	[23]	Ikeuchi1999	30	30	30	[37]
Folsom1970	712	0	0	[24]	Imai1973	0	0	9	[38]
Folsom1975	62	0	0	[33]	Livingston1985	79	709	552	[39]
Folsom1979	1188	0	0	[25]	Livingston1987	70	0	12	[40]
Fowler1983a	0	0	5	[34]	Livingston2000	15	0	14	[96]
Fowler1983b	0	0	8	[94]	Miyake1960	0	9	0	[14]
Fowler1991	0	0	20	[35]	Miyake1961	6	6	0	[15]
Hirose1992	3	0	4	[10]	Miyake1962	9	9	0	[16]
Hirose1999	23	23	23	[11]	Miyake1963	3	3	0	[17]
Hirose2000	11	0	110	[95]	Miyake1968	0	0	15	[18]
Hirose2001	12	0	52	[12]	Miyake1978	0	0	37	[19]
Hirose2002	11	0	56	[13]	Miyake1988	126	0	46	[20]
Hong1999	21	0	22	[36]	Miyao1998	8	0	31	[21]
msa1963	11	11	0	[61]	Nagaya1964	12	12	0	[41]
msa1965	51	48	0	[62]	Nagaya1965	32	31	0	[43]
msa1966	40	40	0	[63]	Nagaya1970	53	59	0	[44]
msa1967	51	55	0	[64]	Nagaya1976	34	99	0	[45]
msa1968	49	53	0	[65]	Nagaya1981	77	77	0	[46]
msa1969	49	50	0	[66]	Nagaya1984	94	97	73	[47]
msa1970	56	59	0	[67]	Nagaya1987	104	70	78	[48]
msa1971	47	51	0	[68]	Nagaya1993	46	0	24	[49]
msa1976	114	115	0	[69]	Nakanishi1990	0	0	117	[50]
msa1977	49	49	0	[70]	Nakanishi1995	0	0	55	[51]
msa1978	38	38	0	[71]	Noshkin1974	4	0	2	[97]
msa1979	37	37	0	[72]	Noshkin1976	5	0	5	[52]
msa1980	51	53	0	[73]	Noshkin1978	21	0	24	[53]
msa1981	60	61	0	[74]	Noshkin1979	0	0	23	[54]
msa1982	70	75	0	[75]	Noshkin1981	0	0	5	[55]
msa1983	43	43	0	[76]	Noshkin1987	9	0	0	[98]
msa1984	72	76	0	[77]	Noshkin1999	196	39	266	[56]
msa1985	57	62	0	[78]	Noshkin2000	13	0	8	[99]
msa1986	73	78	0	[79]	Pillay1964	1	1	2	[57]
msa1987	67	65	0	[80]	Delfanti2000	99	34	88	[58]
msa1988	80	78	0	[81]	Saruhashi1975	231	176	0	[22]
msa1989	72	72	24	[82]	Shirasawa1968	83	85	0	[59]
msa1990	66	66	24	[83]	Wong1971	1	0	3	[100]
msa1991	56	56	12	[84]	Wong1992	131	0	210	[60]
Total						7737	3972	2666	

FEATURES OF THE HAM DATABASE

Geographical Distribution

The total numbers of the data in the HAM database for the three major radionuclides in each sea area are summarized in Table 2. In the database, sea area code is given in each data. The geographical distributions for each decade from the 1950s to the 1990s are shown in Figs. 1 to 5, respectively. Since our effort was concentrated in the Pacific Ocean and its marginal seas, more than 80% of the data for each radionuclide are for the Pacific Ocean and the Sea of Japan in the HAM database as shown in Table 2. In the 1950s, a few ^{137}Cs and ^{90}Sr concentration data at the western North Pacific near Japan were available as shown in Fig. 1. The $^{239,240}\text{Pu}$ concentration data did not exist in this decade in HAM database. The number of ^{137}Cs concentration data increased abruptly in the North Pacific in the 1960s, especially in 1966–1968 as shown in Fig. 6. The station locations of ^{137}Cs concentration data covered well over the Pacific Ocean, while ^{90}Sr concentration data and $^{239,240}\text{Pu}$ concentration data showed still less and sparsely, respectively, in the 1960s (Fig. 2). In the 1970s (Fig. 3), the number of ^{137}Cs concentration data decreased, however, the station locations of ^{137}Cs concentration data covered well over the Pacific Ocean. The station locations of ^{90}Sr concentration data and $^{239,240}\text{Pu}$ concentration data covered the North Pacific Ocean. The geographical distributions of ^{137}Cs concentration data and ^{90}Sr concentration data in the 1980s were concentrated at the western North Pacific, while that of $^{239,240}\text{Pu}$ concentration data covered over the North Pacific (Fig. 4). No data area at the central and the eastern North Pacific for the three radionuclides was seen in the 1990s (Fig. 5).

TABLE 2
Total Number of the Data in Oceans and Marginal Seas

Sea Area	Area Code	^{137}Cs	^{90}Sr	Pu
Eastern North Pacific	NEP	2530	373	598
Western North Pacific	NWP	2620	1650	1009
Eastern South Pacific	SEP	510	115	63
Western South Pacific	SWP	127	12	30
Sea of Japan	SOJ	1276	1224	541
Sea of Okhotsk	SOO	21	16	35
East China Sea	ECS	48	41	3
South China Sea	SCS	16	0	1
North Atlantic	NA	238	287	171
South Atlantic	SA	135	190	109
Indian Ocean	IO	106	26	14
Not assigned yet	XXX	110	38	92
Total		7737	3972	2666

Temporal Variation of the Numbers for the Three Radionuclides

The temporal variation of the numbers of the data for the three radionuclides in each year is shown in Fig. 6. ^{137}Cs and ^{90}Sr concentration data were available since 1957, while $^{239,240}\text{Pu}$ data were available since 1964. A maximum number of 1268 for the ^{137}Cs concentration data was in 1967, while the numbers of ^{90}Sr and $^{239,240}\text{Pu}$ concentration data were 83 and 15 in the same year, respectively. This tendency to have so much ^{137}Cs concentration data and less ^{90}Sr and plutonium concentration data was observed during the

FIGURE 1. Geographical distribution of ^{137}Cs and ^{90}Sr concentration data in the 1950s in HAM database. (a) ^{137}Cs ; (b) ^{90}Sr .

1960s. The numbers of the data for the three radionuclides became the same due to the GEOSECS project, in which the analysis for the three radionuclides was done simultaneously in the early 1970s. The numbers of the data for each radionuclide were around 100 per year throughout the 1980s and the 1990s.

The Number of the Data for the Surface Water and the Profiles

The number of the data for surface water and profiles in each of the oceans and marginal seas are summarized in Table 3 for each radionuclide. For the ^{137}Cs concentration data, 55.7% of the total amount of 7737 data is for surface data, then the remaining 44.3% is for the profiles. The numbers of the profile for the ^{137}Cs is about 430. For the ^{90}Sr concentration data, 42.7% of the total amount of 3972 is for the surface and 57.3% is for the profiles. The numbers of the profile for the ^{90}Sr is about 320. For the $^{239,240}\text{Pu}$ concentration data, 23.6% of the total amount of 2666 is for the surface and 76.4% is for the profile. The numbers of the profile for the $^{239,240}\text{Pu}$ is about 240.

Counting Error of the Data

The number of the data associated with counting error for the ^{137}Cs concentration data is 7164, which is 92.6% of the total amount of 7737 in this database. For the ^{90}Sr concentration data and the $^{239,240}\text{Pu}$ concentration data, 98.0% of the total amount of 3972 and 96.0% of the total amount of 2666 in this database are reported with counting error.

FIGURE 2. Geographical distribution of ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$ concentration data in the 1960s in HAM database. (a) ^{137}Cs ; (b) ^{90}Sr ; (c) $^{239,240}\text{Pu}$.

FIGURE 3. Geographical distribution of ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$ concentration data in the 1970s in HAM database. (a) ^{137}Cs ; (b) ^{90}Sr ; (c) $^{239,240}\text{Pu}$.

FIGURE 4. Geographical distribution of ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$ concentration data in the 1980s in HAM database. (a) ^{137}Cs ; (b) ^{90}Sr ; (c) $^{239,240}\text{Pu}$.

FIGURE 5. Geographical distribution of ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$ concentration data in the 1990s in HAM database. (a) ^{137}Cs ; (b) ^{90}Sr ; (c) $^{239,240}\text{Pu}$.

FIGURE 6. Number of concentration data for each radionuclides during the period from 1957–1998. Open bar: ^{137}Cs ; solid bar: ^{90}Sr ; hatched bar: $^{239,240}\text{Pu}$.

TABLE 3
The Number of the Data for Surface Water and Profiles in Oceans and Marginal Seas

Sea Area	Area Code	^{137}Cs		^{90}Sr		Pu	
		Surface	Profiles	Surface	Profiles	Surface	Profiles
Eastern North Pacific	NEP	1410	1120	56	317	96	502
Western North Pacific	NWP	1444	1176	772	878	368	641
Eastern South Pacific	SEP	368	142	23	92	39	24
Western South Pacific	SWP	118	9	12	0	26	4
Sea of Japan	SOJ	738	538	715	509	56	485
Sea of Okhotsk	SOO	5	16	0	16	3	32
East China Sea	ECS	44	4	41	0	3	0
South China Sea	SCS	16	0	0	0	1	0
North Atlantic	NA	22	216	17	270	16	155
South Atlantic	SA	5	130	5	185	3	106
Indian Ocean	IO	106	0	26	0	14	0
Not assigned yet	XXX	36	74	30	8	4	88
Subtotal		4312	3425	1697	2275	629	2037
		(55.7%)	(44.3%)	(42.7%)	(57.3%)	(23.6%)	(76.4%)
Total		7737		3972		2666

FIGURE 7. Number of data for each error class for each radionuclide in the database. (a) ^{137}Cs ; (b) ^{90}Sr ; (c) $^{239,240}\text{Pu}$.

For the ^{137}Cs concentration data, the magnitude of the counting error varied within 30% in general as shown in Fig. 7(a). Of the total amount of 7164, 208 (2.9%) showed larger counting error exceeding 100%. Those larger counting errors were seen for the data in deeper layers[24,25,29,30, 36,39,47,48,75,78,79,81,83,85,91] where the ^{137}Cs concentrations were generally very low. For the ^{90}Sr concentration data, the magnitude of the counting error varied within 30% in general as shown in Fig. 7(b). Of the total amount of 3892, 205 (5.3%) show larger counting error, which is exceeding 100%. Those larger counting errors were seen for the data in deeper layers[29,30,39,45,47,48, 73,74,77,78,79,84,85] as well as the ^{137}Cs concentration data. For the $^{239,240}\text{Pu}$ concentration data, the magnitude of the counting error varied between 6 and 30% in general as shown in Fig. 7(c). Of the total amount of 2562, 167 (6.5%) show larger counting error, which is exceeding 100%. Those larger counting errors were seen both in shallower layers and deeper layers[47,48,54,60].

DATABASE DISTRIBUTION AND CITATION OF THIS DATABASE

This HAM database is freely available on request. When results or illustrations obtained using the HAM database are presented in papers, their authors should cite this paper together with a version of the HAM database used.

The description in this paper on the HAM database is based on the version U as of 29 Dec. 2001.

CONCLUSIONS

The database regarding “Historical Artificial Radionuclides in the Pacific Ocean and its Marginal Seas”, HAM database, has been created. The data in the HAM database have been collected from about 90

literature citations, which include published papers/reports; annual reports of the Hydrographic Department, Maritime Safety Agency, Japan; and unpublished data provided by individuals. The data of the concentrations of ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ have been accumulating since 1957–1998. The present database includes 7737 records for ^{137}Cs concentration data, 3972 records for ^{90}Sr concentration data, and 2666 records for $^{239,240}\text{Pu}$ concentration data. The spatial variation of sampling stations in the HAM database is heterogeneous, namely, more than 80% of the data for each radionuclide is from the Pacific Ocean and the Sea of Japan, while a relatively small amount of data is from the South Pacific. This HAM database will allow us to use these radionuclides as significant chemical tracers for oceanographic studies as well as the assessment of environmental effects of anthropogenic radionuclides for these 5 decades. Furthermore, these radionuclides can be used to verify the oceanic general circulation models in the time scale of several decades.

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REFERENCES

1. Bowen, V.T., Noshkin, V.E., Livingston, H.D., and Volchok, H.L. (1980) Fallout radionuclides in the Pacific Ocean: vertical and horizontal distributions, largely from GEOSECS stations. *Earth Planet. Sci. Lett.* **49**, 411–434.
2. International Atomic Energy Agency (2000) Global Marine Radioactivity Database (GLOMARD), IAEA-TECDOC-1146, PP51. Vienna, Austria.
3. Miyao, T., Hirose, K., Aoyama, M., and Igarashi, Y. (2000) Trace of the deep water formation in the Japan Sea deduced from historical ^{137}Cs data. *Geophys. Res. Lett.* **27**, 3731–3834.
4. Hirose, K. and Aoyama, M. (2003) Analysis of ^{137}Cs and $^{239,240}\text{Pu}$ concentrations in waters of the Pacific. *Deep-Sea Res. II* **50**, 2675–2700.
5. Aoyama, M. and Hirose, K. (2003) Temporal variation of ^{137}Cs water column inventory in the North Pacific since the 1960s. *J. Environ. Radioact.* **69**(1–2), 107–117.
6. Tsumune, D., Aoyama, M., and Hirose, K. (2003) Behavior of ^{137}Cs concentrations in the North Pacific in an ocean general circulation model. *J. Geophys. Res.* **108**, doi:10.1029/2002JC001434.
7. Tsumune, D., Aoyama, M., and Hirose, K. (2003) Numerical simulation of ^{137}Cs and $^{239,240}\text{Pu}$ concentrations by an ocean general circulation model. *J. Environ. Radioact.* **69**(1–2), 61–84.
8. Aoyama, M. and Hirose, K. (1995) The temporal and spatial variation of ^{137}Cs concentration in the western North Pacific and its marginal seas during the period from 1979 to 1988. *J. Environ. Radioact.* **29**, 57–74.
9. Aoyama, M., Hirose, K., Miyao, T., Igarashi, Y., and Povinec, P.P. (2001) ^{137}Cs activity in surface water in the western North Pacific. *J. Radioanal. Nucl. Chem.* **248**, 789–793.
10. Hirose, K., Sugimura, Y., and Aoyama, M. (1992) Plutonium and ^{137}Cs in the western North Pacific: estimation of residence time of plutonium in surface waters. *Appl. Radiat. Isot.* **43**, 349–359.
11. Hirose, K., Amano, H., Baxter, M.S., Chaykovskaya, E., Chumichev, V.B., Hong, G.H., Isogai, K., Kim, C.K., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K., Pettersson, H.B.L., Povinec, P.P., Seto, Y., Tkalin, A., Togawa, O., and Veletova, N.K. (1999) Anthropogenic radionuclides in seawater in the East Sea/Japan Sea: results of the first-stage Japanese-Korean-Russian expedition. *J. Environ. Radioact.* **43**, 1–13.
12. Hirose, K., Aoyama, M., Miyao, T., and Igarashi, Y. (2001) Plutonium in seawaters on the western North Pacific. *J. Radioanal. Nucl. Chem.* **248**, 771–776.

13. Hirose, K., Miyao, T., Aoyama, M., and Igarashi, Y. (2002) Plutonium isotopes in the Sea of Japan. *J. Radioanal. Nucl. Chem.* **252**, 293–299.
14. Miyake, Y., Saruhashi, K., and Katsuragi, Y. (1960) Strontium 90 in western North Pacific surface waters. *Pap. Meteorol. Geophys.* **XI**, 188–191.
15. Miyake, Y., Saruhashi, K., Katsuragi, Y., and Kanazawa, T. (1961) Cesium 137 and Strontium 90 in sea water. *J. Radiat. Res.* **2-1**, 25–28.
16. Miyake, Y. (1962) Penetration of ^{90}Sr and ^{137}Cs in deep layers of the Pacific and vertical diffusion rate of deep water. *J. Radiat. Res.* **3-3**, 141–147.
17. Miyake, Y. (1963) Artificial radioactivity in the Pacific Ocean. In *Radioactive Tracers in Oceanography*. I.U.G.G. Monograph. International Union of Geodesy and Geophysics. pp. 21–30.
18. Miyake, Y. and Sugimura, Y. (1968) Plutonium content in the western North Pacific waters. *Pap. Meteorol. Geophys.* **19**, 481–485.
19. Miyake, Y. and Sugimura, Y. (1978) The Plutonium Content of Pacific Ocean Waters. IAEA-SM-199/22. pp. 91–105.
20. Miyake, Y., Saruhashi, K., Sugimura, Y., Kanazawa, T., and Hirose, K. (1988) Contents of ^{137}Cs , plutonium and americium isotopes in the southern ocean waters. *Pap. Meteorol. Geophys.* **39**, 95–113.
21. Miyao, T., Hirose, K., Aoyama, M., and Igarashi, Y. (1998) Temporal variation of ^{137}Cs and $^{239,240}\text{Pu}$ in the Sea of Japan. *J. Environ. Radioact.* **40**, 239–250.
22. Saruhashi, K., Katsuragi, Y., Kanazawa, T., Sugimura, Y., and Miyake, Y. (1975) ^{90}Sr and ^{137}Cs in the Pacific waters. *Rec. Oceanogr. Works Jpn.* **13**, 1–15.
23. Folsom, T.R., Mohanrao, G.J., Pillai, K.C., and Sreekumaran, C. (1968) Distributions of Cs137 in the Pacific. HASL-197, I-95 ~ I-203.
24. Folsom, T.R., Sreekumaran, C., Hansen, N., Moore, J.M., and Grismore, R. (1970) Some Concentrations of Cs137 at Moderate Depths in the Pacific 1965–1968. HASL-217.
25. Folsom, T.R. (1979) Summary of Cs-137 Concentrations Measured at Scripps Institution in N. Pacific Surface Waters. EML-356.
26. Bourlat, Y. and Martin, G. (1992) Precise determination of the concentration of radiocaesium in the water of Mururoa Lagoon. *J. Environ. Radioact.* **17**, 13.
27. Bourlat, Y., Millies-Lacroix, J.C., and Nazard, R. (1995) Determination of plutonium radioactivity in Mururoa Lagoon water. *J. Radioanal. Nucl. Chem.* **197**, 387–408.
28. Bourlat, Y., Millies-Lacroix, J.-C., Petit, G.L., and Bourguignon, J. (1996) ^{90}Sr , ^{137}Cs and $^{239+240}\text{Pu}$ in world ocean water samples collected from 1992 to 1994. In *Radionuclides in the Oceans Inputs and Inventories*. Guéguéniat, P., Germain, P., and Métivier, H., Eds. Institut de Protection et de Sureté Nucleaire, Cherbourg, France. pp. 75–93.
29. Broecker, W.S., Bonebakker, E.R., and Rocco, G.G. (1966) The vertical distribution of cesium137 and strontium90 in the oceans, 2. *J. Geophys. Res.* **71**, 1999–2003.
30. Broecker, W.S. and Simpson, H.J. (1968) A Summary of Lamont Sr90 and Cs137 Measurements on Ocean Water Samples. HASL-197, I-9 ~ I-104.
31. Folsom, T.R., Mohanrao, G.J., and Winchell, P. (1960a) Fallout caesium in surface sea water off the California coast (1959–60) by gamma-ray. *Nature* **187**, 480–482.
32. Folsom, T.R. and Mohanrao, G.J. (1960b) Measurement of fallout cesium in the Pacific Ocean and in terrestrial effluents likely to alter coastal waters. *J. Radiat. Res.* **1-2**, 150–154.
33. Folsom, T.R., Hansen, N., Tatumi, T., and Hodge, V.F. (1975) Recent improvements in methods for concentrating and analyzing radiocesium in sea water. *J. Radiat. Res.* **16**, 19–27.
34. Fowler, S.W., Ballestra, S., Rosa, J.L., and Fukai, R. (1983a) Vertical transport of particulate-associated plutonium and americium in the upper water column of the Northeast Pacific. *Deep-Sea Res.* **30**, 1221–1233.
35. Fowler, S.W., Small, L.F., Rosa, J.L., Lopez, J.J., and Teyssie, J.L. (1991) Interannual variation in transuranic flux at the vertex time-series station in the northeast Pacific and its relationship to biological activity. In *Radionuclides in the Study of Marine Processes*. Kershaw, P.J. and Woodhead, D.S., Eds. Kluwer, Dordrecht. pp. 286–298.
36. Hong, G.H., Kim, S.H., Lee, S.H., Chung, C.S., Tkalin, A.V., Chaykovskaya, E.L., and Hamilton, T.F. (1999) Artificial radionuclides in the East Sea (Sea of Japan) proper and Peter the Great Bay. *Mar. Pollut. Bull.* **38**, 933–943.
37. Ikeuchi, Y., Amano, H., Aoyama, M., Bereznev, V.I., Chaykovskaya, E., Chumichev, V.B., Chung, C.S., Gastaud, J., Hirose, K., Hong, G.H., Kim, C.K., Kim, S.H., Miyao, T., Morimoto, T., Nikitin, A., Oda, K., Pettersson, H.B.L., Povinec, P.P., Tkalin, A., Togawa, O., and Veletova, N.K. (1999) Anthropogenic radionuclides in seawater of the Far Eastern seas. *Sci. Total Environ.* **237/238**, 203–212.
38. Imai, J. and Sakanoue, M. (1973) Content of plutonium, thorium and protactinium in seawater and recent coral in the North Pacific. *J. Oceanogr. Soc. Jpn.* **29**, 76–82.
39. Livingston, H.D., Bowen, V.T., Casso, S.A., Volchok, H.L., Noshkin, V.E., Wong, K.M., and Beasley, T.M. (1985) Fallout Nuclides in Atlantic and Pacific Water Columns: GEOSECS Data. Woods Hole Oceanographic Institute Tech. Rep., WHOI-85-19.
40. Livingston, H.D., Mann, D.R., Casso, S.A., Schneider, D.L., Surprenant, L.D., and Bowen, V.T. (1987) Particle and solution phase depth distributions of transuranics and ^{55}Fe in the North Pacific. *J. Environ. Radioact.* **5**, 1–24.
41. Nagaya, Y., Shiozaki, M., and Seto, Y. (1964a) Radioactive contamination of the Indo-Antarctic Ocean water in each earlier period in 1961 and 1962. *J. Radiat. Res.* **5-3-4**, 206–214.

42. Nagaya, Y., Shiozaki, M., and Seto, Y. (1964b) On the Radiological Survey of Harbours during 1960–1962. msa1964.
43. Nagaya, Y., Shiozaki, M., and Seto, Y. (1965) Some fallout radionuclides in deep waters around Japan. *J. Radiat. Res.* **6-1**, 23–31.
44. Nagaya, Y. and Nakamura, K. (1970) A study on the vertical transport of ^{90}Sr and ^{137}Cs in the surface waters of the seas around Japan. *J. Radiat. Res.* **11-1**, 32–43.
45. Nagaya, Y. and Nakamura, K. (1976) ^{90}Sr and ^{137}Cs contents in the surface waters of the adjacent seas of Japan and the North Pacific during 1969 to 1973. *J. Oceanogr. Soc. Jpn.* **32**, 228–234.
46. Nagaya, Y. and Nakamura, K. (1981) Artificial radionuclides in the western Northwest Pacific (I): ^{90}Sr and ^{137}Cs in the deep waters. *J. Oceanogr. Soc. Jpn.* **37**, 135–144.
47. Nagaya, Y. and Nakamura, K. (1984) $^{239,240}\text{Pu}$, ^{137}Cs and ^{90}Sr in the central North Pacific. *J. Oceanogr. Soc. Jpn.* **40**, 410–424.
48. Nagaya, Y. and Nakamura, K. (1987) Artificial radionuclides in the western Northwest Pacific(II): ^{137}Cs and $^{239,240}\text{Pu}$ inventories in water and sediment columns observed from 1980 to 1986. *J. Oceanogr. Soc. Jpn.* **43**, 345–355.
49. Nagaya, Y. and Nakamura, K. (1993) Distributions and mass-balance of $^{239,240}\text{Pu}$ and ^{137}Cs in the northern North Pacific. In *Deep Ocean Circulation, Physical and Chemical Aspects*. Teramoto, T., Ed. Elsevier Oceanography Series, Elsevier, Amsterdam.
50. Nakanishi, T., Satoh, M., Takai, M., Ishikawa, A., Murata, M., Dairyoh, M., and Higuchi, S. (1990) Successive determinations of ^{210}Pb , ^{210}Po , ^{226}Ra , ^{228}Ra and selected actinides in seawater and sea sediment. *J. Radioanal. Nucl. Chem.* **138**, 321–230.
51. Nakanishi, T., Shiba, Y., Muramatsu, M., and Haque, M.A. (1995) Estimation of mineral aerosol fluxes to the Pacific by using environmental plutonium as a tracer. In *Biogeochemical Processes and Ocean Flux in the Western Pacific*. Sakai, H. and Nozaki, Y., Eds. Terra Scientific Publishing Company, Tokyo. pp. 15–30.
52. Noshkin, V.E., Eagle, R.J., and Wong, K.M. (1976) Plutonium levels in Kwajalein Lagoon. *Nature* **262**, 745–748.
53. Noshkin, V.E., Wong, K.M., Jokela, T.A., Eagle, R.J., and Brunk, J.L. (1978) Radionuclides in the Marine Environment Near the Farallon Island. UCRL-52381. pp. 16.
54. Noshkin, V.E., Wong, K.M., and Eagle, R.J. (1979) Plutonium concentrations in fish and seawater from Kwajalein Atoll. *Health Phys.* **37**, 549–556.
55. Noshkin, V.E., Brunk, J.L., Jokela, T.A., and Wong, K.M. (1981) ^{238}Pu concentrations in the marine environment at San Clemente Island. *Health Phys.* **40**, 643–659.
56. Noshkin, V.E. (1999) Concentrations of Radionuclides in Some Seawater and Sediment Samples from the Equatorial Pacific with Emphasis on Samples from the Western Pacific Ocean. Preliminary report prepared for the 1st Coordinated Research Project on Worldwide Marine Radioactivity Studies.
57. Pillai, K.C., Smith, R.C., and Folsom, T.R. (1964) Plutonium in the marine environment. *Nature* **203**, 568–571.
58. Delfanti, R., Carlo, P., Stefano, S., and Ruggero, L. (2000) IAEA CRP "Worldwide Marine Radioactivity". Final Report Research contract - Agreement N. ITA-26803.
59. Shirasawa, T.H. and Schuert, E.A. (1968) Fallout Radioactivity in the North Pacific Ocean: Data Compilation of Sr-90 and Cs-137 Concentrations in Seawater. HASL-197 I-66 ~ I-94.
60. Wong, K.M., Jokela, T.A., Eagle, R.J., Brunk, J.L., and Noshkin, V.E. (1992) Radionuclide concentrations, fluxes, and residence times at Santa Monica and San Pedro Basins. *Prog. Oceanogr.* **30**, 353–391.
61. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1965) Annual Reports of Radioactivity Survey for 1963 [in Japanese]. msa1963.
62. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1967) Annual Reports of Radioactivity Survey for 1965 [in Japanese]. msa1965.
63. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1968) Annual Reports of Radioactivity Survey for 1966 [in Japanese]. msa1966.
64. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1969) Annual Reports of Radioactivity Survey for 1967 [in Japanese]. msa1967.
65. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1970) Annual Reports of Radioactivity Survey for 1968 [in Japanese]. msa1968.
66. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1971) Annual Reports of Radioactivity Survey for 1969 [in Japanese]. msa1969.
67. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1972) Annual Reports of Radioactivity Survey for 1970 [in Japanese]. msa1970.
68. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1973) Annual Reports of Radioactivity Survey for 1971 [in Japanese]. msa1971.
69. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1976) Annual Reports of Radioactivity Survey for 1972–1974 [in Japanese]. msa1976.
70. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1977) Annual Reports of Radioactivity Survey for 1975 [in Japanese]. msa1977.
71. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1978) Annual Reports of Radioactivity Survey for 1976 [in Japanese]. msa1978.

72. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1979) Annual Reports of Radioactivity Survey for 1977 [in Japanese]. msa1979.
73. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1980) Annual Reports of Radioactivity Survey for 1978 [in Japanese]. msa1980.
74. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1981) Annual Reports of Radioactivity Survey for 1979 [in Japanese]. msa1981.
75. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1982) Annual Reports of Radioactivity Survey for 1980 [in Japanese]. msa1982.
76. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1983) Annual Reports of Radioactivity Survey for 1981 [in Japanese]. msa1983.
77. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1984) Annual Reports of Radioactivity Survey for 1982 [in Japanese]. msa1984.
78. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1985) Annual Reports of Radioactivity Survey for 1983 [in Japanese]. msa1985.
79. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1986) Annual Reports of Radioactivity Survey for 1984 [in Japanese]. msa1986.
80. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1987) Annual Reports of Radioactivity Survey for 1985 [in Japanese]. msa1987.
81. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1988) Annual Reports of Radioactivity Survey for 1986 [in Japanese]. msa1988.
82. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1989) Annual Reports of Radioactivity Survey for 1987 [in Japanese]. msa1989.
83. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1990) Annual Reports of Radioactivity Survey for 1988 [in Japanese]. msa1990.
84. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1991) Annual Reports of Radioactivity Survey for 1989 [in Japanese]. msa1991.
85. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1992) Annual Reports of Radioactivity Survey for 1990 [in Japanese]. msa1992.
86. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1993) Annual Reports of Radioactivity Survey for 1991 [in Japanese]. msa1993.
87. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1994) Annual Reports of Radioactivity Survey for 1992 [in Japanese]. msa1994.
88. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1995) Annual Reports of Radioactivity Survey for 1993 [in Japanese]. msa1995.
89. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1996) Annual Reports of Radioactivity Survey for 1994 [in Japanese]. msa1996.
90. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1997) Annual Reports of Radioactivity Survey for 1995 [in Japanese]. msa1997.
91. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1998) Annual Reports of Radioactivity Survey for 1996 [in Japanese]. msa1998.
92. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (1999) Annual Reports of Radioactivity Survey for 1997 [in Japanese]. msa1999.
93. Hydrographic Department, Maritime Safety Agency, Ministry of Transport (2000) Annual Reports of Radioactivity Survey for 1998 [in Japanese]. msa2000.
94. Fowler, S.W. (1983b) Unpublished.
95. Hirose, K. (2000c) Personal communication. Unpublished data.
96. Livingston, H.D. (2000) Personal communication.
97. Noshkin, V.E. (1974) Personal communication.
98. Noshkin, V.E. (1987) Personal communication.
99. Noshkin, V.E. (2000) Personal communication.
100. Wong, K.M. (1971) Personal communication.

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