UNDERGROUND WATER RADIONUCLIDE CONCENTRATIONS FROM MURUROA AND FANGATAUFA ATOLLS

S. MULSOW, S., M. COQUERY, C. DOVLETE, J. GASTAUD, Y. IKEUCHI, M. K. PHAM, P. POVINEC
Marine Environment Laboratory, International Atomic Energy Agency, Monaco

The fate of radionuclide concentration distribution patterns resulting from underground nuclear weapon testing, depends on the different geochemical and structural characteristics of the areas where the tests took place. The different origin and characteristics of each of the facies found in Mururoa and Fangataufa Atolls that underwent nuclear testing, may play an important role in the radionuclide concentration fate; confinement and/or remobilization. To obtain basic information on the current status of the radionuclide concentrations in underground waters as a result of nuclear tests, the IAEA within the framework of the IAEA Study [1], in conjunction with the Commissariat de l'Energie Atomique de France, undertook a monitoring survey of interstitial waters (dissolved and particulate) from 7 monitoring wells and two cavities located at Mururoa Atoll and two monitoring wells at Fangataufa Atoll (Table 1).

Table 1. Characteristics of underground cavities and monitoring wells

Atoll	well name	location	facies	casing	tubing	depth m
Mururoa	Aristee	rim	Basalt	?	single	650 m
	Ceto	rim	Basalt	?	single	490 m
	Geo 5	rim	carbonate	80 m	polytube	230 m
	Geo 8	rim	carbonate	83 m	polytube	276 m
	Geo 10	rim	carbonate	120 m	polytube	309 m
	Isurus 10	lagoon	carbonate	68 m	polytube	265 m
	Tazard 14	lagoon	transition	72 m	polytube	245 m
	Pieuvre 37	lagoon	transition	80 m	polytube	300 m
	Murene 16	lagoon	transition	78 m	polytube	230 m
Fangataufa	Fuseau 30	lagoon	transition	62 m	polytube	193 m
	Mitre 27	lagoon	transition	77 m	polytube	238.5 m

The sampling protocol for each of the sites involved three steps; 1) the retrieval of the sampling tube (underwater or in the rim), one in the case of cavities (Aristee and Ceto) and more than one tube in the case of wells (Polytube). Underwater wells required the assistance of divers. 2) Upon retrieval, the sampling tube was connected to a peristaltic pump. 3) Once a dead volume (initial water contained in the tube) was drawn off, a filter (0.45 μm), followed by a Plexiglas (vol. 1 l) cell containing pH and Eh electrodes (Ag/AgCl ref.) was placed in line. Later on, and to ensure stability of the pH and Eh measurements, a magnetic bar and stirrer were added to the measuring system. This setting allowed continuous measurements of pH, Eh during sampling time. The exit tube from the cell was then connected to a sample-receiving container. Thus, from each station, a filtered water sample and a particle fraction (0.45 μm pore size) were collected. After every dead volume was pumped off, a 1 l sample was taken to measure ^{3}H. On each sample fraction, the analyses of selected elements were performed (Fe, Mg, Mn, Al, Ca, K, Sr, Si, Na, Cl) by Atomic Absorption Spectrometry (AAS) [1]. The same analyses were carried out for the water fraction using ICP/AES for the metals with the addition of Cl, SO_4 (ion chromatography) and alkalinity by titration [3]. Additionally, concentrations were determined for radionuclides of interest, such as ^{137}Cs, $^{239,240}Pu$, ^{238}Pu ^{241}Am, ^{90}Sr in both water and particles; ^{155}Eu, ^{125}Sb and ^{60}Co in particles. The chemical separation for each radionuclide was carried out following IAEA-MEL procedures. Briefly, from acidified water samples, transuranic ^{137}Cs and ^{90}Sr were sequentially co-precipitated. ^{137}Cs was measured by gamma spectrometry. Transuranics were radiochemically separated, electrodeposited on stainless steel discs and counted by alpha spectrometry. ^{90}Sr was determined by ^{90}Y ingrowth and measured by beta counting.

The characterization of the water samples using the parameters measured *in situ* showed that after 1 dead volume of the underground water was pumped, the geochemical characteristics were rather similar. HTO was constant throughout the sampling time at all the stations. Although some of the parameters, particularly pH and Eh, seemed to be related to tidal changes, this was not a general feature in all the stations sampled.

In the dissolved fraction of Mururoa waters, ^{3}H, ^{137}Cs and ^{90}Sr were detected in all wells except for ^{137}Cs at Murene (Table 2). The highest values for the three radionuclides were found at Aristee, Ceto, Geo 8, Geo 10 and Pieuvre 37. The only unexpected findings were Geo 8 and Geo 10. These two stations are

Table 2. Radionuclide concentrations in water and particulates

Well	Water [mBq/l]				Particulates [mBq/g]					
	^{3}H	^{90}Sr	^{137}Cs	$^{239,240}Pu$	$^{239,240}Pu$	^{238}Pu	^{241}Am	^{60}Co	^{125}Sb	^{137}Cs
Aristee	(6.1± 0.31) x10^{9}	(3.19 ± 0.29) x10^{5}	1.0 x10^{5}	< 0.008	235 ± 18	32 ± 3	44.4 ± 4.5	690 ± 80	7600 ± 200	4770 ± 90
Ceto	(2.17 ± 0.11) x10^{10}	(2.54 ± 0.15) x10^{5}	(11.2 ± 0.8) x10^{3}	0.020 ± 0.004	38.1 ± 3.2	3.9 ± 0.6	17.2 ± 1.7	160 ± 20	310 ± 50	4260 ± 80
Geo 5	(1.83 ± 0.36) x10^{4}	8.4 ± 1.3	20.3 ± 3.0	< 0.03	< 0.4	< 0.2	< 0.6	<20	<40	<20
Geo 8	(1.4 ± 0.07) x10^{6}	(1.6 ± 0.1) x 10^{4}	(1.1 ± 0.1) x10^{3}	< 0.01	< 0.7	< 0.8	< 1	<20	<60	<20
Geo 10	(7.47 ± 0.38) x10^{6}	(1.6 ± 0.1) x 10^{4}	1.1 x10^{3}	< 0.03	< 0.2	< 0.2	< 0.7	<12	<35	<24
Isurus 10	(4.82 ± 0.27) x10^{5}	64 ± 4.1	11.9 ± 0.2	< 0.004	<1	< 1	< 2	<46	<70	<23
Tazard 14	(6.88 ± 0.35) x10^{6}	53 ± 3.4	78.3 ± 7.0	< 0.005	< 0.8	< 0.5	< 1	<100	<260	<100
Pieuvre 37	(8.45 ± 0.42) x10^{6}	(1.2 ± 0.06) x 10^{4}	9.7 x10^{3}	< 0.007	< 3	< 3	< 4	<45	<90	<45
Murene 16	(1.00 ± 0.05) x10^{7}	103 ± 7	< 2	< 0.007	< 1	< 0.7	< 0.8	<24	<70	<48
Fuseau 30	(3.44 ± 0.18) x10^{6}	337 ± 21	127 ± 9	< 0.009	23.3 ± 2.0	8.9 ± 0.8	0.54 ± 0.11	<1	<3	<1
Mitre 27	(1.02 ± 0.32) x10^{4}	2.2 ± 1.2	19.7 ± 2.0	< 0.05	24.3 ± 2.0	9.3 ± 0.8	1.15 ± 0.38	<20	<50	<20

monitoring wells (called "instrument wells"). These findings suggest that dissolved radionuclides may have mobilized from a source point reaching Geo 8 and Geo 10 wells. The lowest values were found in underground water collected from Geo 5. This finding is in accordance with the fact that Geo 5 is often regarded as a "Mururoa background" station by the CEA. Isurus 10, Tazard 14 and Murene 16, showed intermediate values for these radionuclides. Of the transuranic elements measured, only $^{239,240}Pu$ and ^{241}Am were found at Ceto, but in quantities too close to detection limits. Thus at the wells sampled, transuranics in dissolved phase were only found in low concentrations. In order to define underlying patterns for the monitoring wells sampled at Mururoa, a cluster analysis was carried out. Aristee and Ceto show marked differences to the other stations as expected. They have the highest values of all the radionuclides measured. Among the other stations, Geo 5 has the lowest radionuclide signature. Geo 8 is similar to Geo 5 with high ^{137}Cs values which are not to be found at either Geo 10 or Pieuvre 37.

At Fangataufa, Fuseau 30 values consistently one and two orders of magnitudes higher than at Mitre 27 were found for the radionuclides detected in dissolved phase. These two monitoring wells are categorized as nuclear experimental tests. Fuseau 30 has a different radionuclide signature to Mitre 27.

Only 5 stations yielded enough particulate material to carry out both elemental analysis and radionuclide characterizations. Fe and Si had concentrations up to two orders of magnitude higher than any other element measured in the particulate fraction. Geo 5, Isurus 10 and Fuseau 30, had deficiencies in Ca, Si, Al, Mg and Sr compared to the volcanic cavities. Ceto particles are apparently enriched in Mg and Al. A cluster analysis was applied to this data set and clearly distinguishes Aristee and Ceto as sharing unique elemental compositions. The same statistical technique was used to ordinate these five stations using the available data on radionuclide concentrations found in those same particles (Table 2). The resulting cluster shows two different sets of stations sharing similar radionuclide signatures. One formed by Geo 5 and Isurus 10 and the other by Aristee and Ceto. Fuseau 30 has its own radionuclide signature that is not shared by any other station.

High concentrations of particles found in Fuseau 30 and Mitre 27 samples are responsible for the elevated concentrations of transuranics observed in these wells, although their concentrations in the dissolved phase were below detection limit.

The authors are indebted to Drs. D. Smith and E. Warnecke who took part in the sampling campaign, as well as the CEA for logistical support.

REFERENCES

[1] IAEA, Radiological Situation at the Atolls of Mururoa and Fangataufa, IAEA, Vienna (1998).

[2] UNEP, Manual for the geochemical analyses of marine sediments and suspended particulate matter. Reference Methods for Marine Pollution Studies, Monaco (1995) No. 63, 74pp.

[3] PARSONS, T., Y. MAITA AND C. LALLI. A manual of chemical and biological methods for seawater analysis. Pergamon Press, New York (1984) 174pp..