RADIONUCLIDE MIGRATION THROUGH THE GEOSPHERE

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After it abandoned atmospheric nuclear testing in 1974, the French Government carried out, between 1975 and 1996, a further 147 nuclear experiments beneath the atolls of Mururoa and Fangataufa. Of these experiments, ten were safety trials (designed to simulate accident conditions) with little or no nuclear yield.

No very high yield nuclear devices were exploded underground at Mururoa and Fangataufa. None exceeded 150 kilotons, and the total energy release from all underground tests was reported by the French Government to be 3.2 megatons. *(See table, page 32.)* Throughout the world, there have been more than 1800 underground nuclear tests with a total yield of 90 megatons, thirty times that reported from French underground testing.

In atmospheric nuclear tests most of the radioactive material is dispersed into the upper atmosphere and is eventually deposited globally as fallout. In contrast, the residual radioactive material from underground tests is largely confined within the host rock although there is potential for release of radionuclides into the local environment. In assessing the radiological consequences of underground testing, it is necessary to estimate the future releases of radionuclides from the geosphere into the accessible environment over periods of time that typically extend to 10,000 years or more.

Estimation of radionuclide release rates from the geosphere was one of the most complex and challenging tasks within the overall study of the radiological situation at Mururoa and Fangataufa atolls. Although the geological environment of the atolls is in many ways unique, the methodology used in the study is applicable to the assessment of the migration of radionuclides from other underground test sites.

ATOLL GEOLOGY

Viewed from the air, the atolls appear to be thin rings of coral only a few metres above sea level. In reality they are massive volcanic seamounts rising about four kilometers from the sea floor and capped with several hundred metres of carbonate rock (derived from coral accretions).

Volcanic eruptions that led to the formation of the atolls occurred about 11 million years ago. Fuelled by a hotspot in the Earth's crust, volcanoes grew in size, reaching the ocean surface and continuing for some time as subaerial volcanoes. When volcanic activity ceased, the structures slowly sank due to their enormous weight and the isostatic condition of the underlying Pacific Plate. During glaciation periods, the sea level dropped by up to 120 meters, killing the corals and exposing the carbonate to the effects of erosion and leaching by rainwater. Most probably, the karsts (interconnected areas of high permeability), which occur in the carbonate rocks, originated during these glaciation periods.

Today the volcanic rocks are capped by up to 450 meters of carbonate rock, the lowest parts of which have been altered to dolomite by diagenetic exchange processes with the surrounding seawater.

The volcanic base of the atoll consists of an extensive network of essentially linear fissures of various sizes. Conductive pathways originated by a variety of mechanisms, e.g. large-scale magmatic intrusions, cooling contraction processes and more extensive fractures (dykes and sills) produced by magmatic injections that took place after cooling of the initial volcanic rock mass. This network of

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fractures is the main conduit for water (and radionuclide) transport.

The surfaces of initially large conduits in basalt become chemically altered by the flowing water and over long time may become partially sealed by alteration products such as clays and calcite. These alteration products typically have a high surface area and excellent retentive properties for some radionuclides, especially the actinide elements. They therefore play an important role in the retardation of radionuclide transport through the geosphere.

NUCLEAR TESTS & LOCATIONS

An underground nuclear explosion melts and vaporizes the volcanic rock of the atolls in the immediate vicinity of the detonation to form a roughly spherical cavity and a lens-shaped pool of molten volcanic rock at the bottom of the cavity which cools to form a glass-like lava.

The mass of material vaporized in a nuclear explosion is about 80 tonnes per kiloton yield. The quantity of lava formed varies between 500 and 1000 tonnes per kiloton yield, depending on the nature of the rock and its moisture content.

The French authorities did not provide the study team with the exact location of each of the 147 underground tests. They did, however, provide a plan view of each atoll showing the test zones, the number of tests, maximum test yield and total test yield in each zone.

All nuclear tests were conducted in the volcanic rock at depths between 500 and 1100 meters. Of the ten safety trials, all were in one zone at Mururoa and seven were in the carbonate rocks at depths below 280 meters. Three of the safety trials in the carbonate rocks had a small nuclear yield.

Not all the tests were equally effective in retaining the radionuclides produced by the nuclear explosion. For calculation purposes, the discrete source terms were grouped into seven categories:

Category 1. A total of 121 normal tests for which there was adequate thickness of essentially undamaged volcanic rock above the top of each test chimney to achieve good confinement.

Category 2. Four tests with apparently sufficient thickness of cover but inadequacies nevertheless in the volcanic cover.

Category 3. Twelve tests in which the cavity-chimney reached the top of the volcanic rock.

Category 4. Three safety trials in which there was a (very small) fission yield, at least 280 meters deep in the carbonate rock.

Category 5. Four safety trials, at least 280 meters deep in the carbonate rock, in which there was no fission yield.

Category 6. Three safety trials in the volcanic rock that had no fission yield.

Category 7. Two waste shafts drilled deep into the volcanic rock into each of which waste containing 3.7 kilograms of plutonium was disposed.

In practice, it was found that most of the release to the biosphere could be attributable to four categories: these were categories 2, 3, 4, and 5.

RADIONUCLIDE INVENTORY

The first step in the estimation of migration rate of radionuclides is determination of the underground radionuclide inventory. French scientists provided upper limits for the yields in each test area. These were independently checked using independent seismic data for each test from the Rarotonga seismic station in the Cook Islands. There was excellent agreement between the French data and the IAEA estimates, strongly suggesting that the French upper limits were in fact very close to the actual values.

The radionuclide inventory was estimated from the yields by making some reasonable assumptions about the proportion of energy from fission of plutonium-239, uranium-235, uranium-238 and from fusion of hydrogen isotopes. These estimates were also in good agreement with information provided by the French authorities. *(See table, next page.)*

HYDROGEOLOGY OF THE ATOLLS

The volcanic and carbonate zones of the atoll are both saturated with water. Essentially, groundwater circulation in the atoll is governed by buoyancy forces due to the geothermal flux heating the system from below. Cold and denser ocean waters penetrate at depth from the flanks of the atoll, flow toward the central warmer regions, gradually heat up, become lighter and move upward towards the lagoon. The high permeabilities within the carbonate formations allow large, almost hori-

zontal centripetal fluxes of cold water.

After a nuclear explosion, the hydrology in the vicinity of the cavity-chimney is altered from the natural, pre-test condition due to the increase in permeability and heating of water and rock. This results in an increase in upward flow of water from the cavity-chimney towards the accessible environment.

Once thermal equilibrium is established, the temperature in the cavity-chimney is about 25° to 50° C above the ambient temperature. This temperature rise is essentially independent of the yield of the test. The temperature in the cavity-chimney will slowly decrease over a few hundred years through the combined effects of convection and conduction.

Regarding the groundwater flow under Mururoa atoll, velocities are highest in the vicinity of each test and in the more permeable carbonate rocks. Flow in the carbonate rock is also affected by tidal fluctuations which effectively mix water in the carbonate zone (especially the karsts) and thereby affect the rate of release of radionuclides to both the lagoon and directly to the ocean.

INVENTORY OF SELECTED LONG-LIVED RADIONUCLIDES AT MURUROA AND FANGATAUFA ATOLLS

	Study Data (TBq)		
Radionuclide	*Mururoa*	*Fangataufa*	*Total*
Tritium	232 000	48 000	280 000
Carbon-14	25	2.6	28
Chlorine-36	1.3	0.4	1.7
Calcium-41	1.0	0.3	1.3
Nickel-59	2.9	0.9	3.8
Nickel-63	340	110	450
Selenium-79	0.008	0.003	0.011
Krypton-85	670	380	1000
Strontium-90	7300	3500	10 800
Zirconium-93	0.23	0.09	0.32
Technetium-99	1.9	0.6	2.5
Palladium-107	0.18	0.03	0.21
Iodine-129	0.0047	0.0014	0.0061
Caesium-135	0.20	0.07	0.27
Caesium-137	10 700	4100	14 800
Europium-152	230	100	330
Neptunium-237	0.22	0.03	0.25
Plutonium-238	185	15	200
Plutonium-239	1030	70	1100
Plutonium-240	280	20	300
Plutonium-241	6200	620	6800
Americium-241	350	30	380

COMPARISON OF ESTIMATES OF YIELDS FROM UNDERGROUND NUCLEAR TESTS

		Total Energy (kilotons TNT equivalent)	
Location	*Number of tests*	*Study Estimate*	*French Value*
Mururoa rim	83	970	<1060
Mururoa lagoon	54	1443	<1450
Fangataufa rim	2	39	<20
Fangataufa lagoon	8	731	<750
Total	147	3183	<3280

MIGRATION OF RADIONUCLIDES

Before radionuclides can migrate through the geosphere, they must be present in the water phase. Owing to the high pressures at depth, volatile or gaseous radionuclides (such as tritium, noble gases, and iodine) initially present in the gas phase will dissolve in the water.

Most radionuclides, however, are either trapped in the lava or sorbed on the rubble. Experimental evidence shows that release of radionuclides from lava and rubble occurs by different mechanisms. The leaching of lava is a slow, rate-limited process whereas the release from the rubble is assumed to be an equilibrium-controlled process between groundwater and radionuclides sorbed on solid surfaces.

In the Study, a dual porosity model was used to estimate transport rates of radionuclides from the test cavity to the carbonate formation. Calculations were carried out for 32 radionuclides, with partcular attention focused on the four key radionuclides — plutonium-239, caesium-137, strontium-90, and tritium.

As part of the analysis, an independent sampling programme of underground waters was undertaken. It was done to verify the results of more extensive measurements carried out by French scientists and to compare the Study team's predictions with

measured concentrations of radionuclides. Samples of water were taken from within the cavity-chimney of two tests and at nine other locations within the carbonate formations.

There was good agreement between the French data and the Study's results. The plutonium concentrations were found to be either close to the detection limit or not detectable, even within the cavity-chimneys. It was concluded that plutonium is effectively retained within the glassy lava formed as a result of the nuclear explosion, although the Study conservatively retained the initial assumption that 5% was deposited on the rubble.

From the underground sampling results, estimates were made of the inventories of radionuclides within particular zones of the carbonate formations. Radionuclides in the carbonate formation can be released to the biosphere either by groundwater flow upwards into the lagoons or by flow along the karstic layers and into the ocean at a depth of about 300 meters.

Although the carbonate formations are quite permeable, they are also large water reservoirs, and the average residence time is much longer than the half-life of some radionuclides, such as tritium, strontium-90, and caesium-137.

Two models were assessed to describe the release to the biosphere, a single porosity model and a mixing model.

In the mixing model, the release rate to the lagoons or ocean is proportional to the inventory in the carbonates. For transfer into the lagoon, the proportionality constant can be calculated from estimates of the current inventory of tritium in the carbonate formations and measurements of the elevation of tritium in the lagoons. This corresponds to a release rate of about 0.12% of the inventory per year. For release into the ocean, the release rate has been estimated to be about 5% per year based on limited data of tritium lateral dispersion in the carbonates.

Using the mixing model for release from the carbonates and the dual porosity model for transportation through the volcanic formations, the inventories in the carbonate and release rates to the biosphere were estimated for several hundred years for tritium, strontium-90, and caesium-137, and for over 100,000 years for plutonium-239.

The most important conclusion from these predictions is that the future release rates of tritium, strontium-90, and caesium-137 to the lagoons are unlikely to be higher than current release rates. The predicted maximum release rates into the ocean at depth have already occurred.

For plutonium-239, the peak release rates from underground sources are predicted to occur from 5000 to 10,000 years in the future but are expected to be lower than the current release rates into the lagoons, due to leaching of plutonium-bearing sediments.

The release rates to the lagoons and the ocean as predicted from the models described above were the input for modelling of marine dispersion of radionuclides and, ultimately, dose estimation from exposure to these radionuclides. *(See related articles, pages 34 and 38.)*

CONCLUSIONS

The main conclusions from this assessment of radionuclide migration from underground sources at Mururoa and Fangataufa atolls are as follows:

- The Study's independent estimates of energy release and radionuclide inventory from underground tests were in good agreement with French reported values.
- Nuclear testing results in an increase in permeability of the rock surrounding a nuclear test.
- There is a natural upward flow of groundwater in the volcanic and carbonate rocks, which is highest in the immediate vicinity of a nuclear test.
- Underground sampling and analysis of water in two test cavity-chimneys indicates a very low concentration of plutonium. Most of the plutonium (>99%) is effectively retained within the solidified glassy lava formed after the nuclear explosion.
- There are tidal effects in the carbonate zones that could result in some release of radionuclides directly into the ocean at depth.
- Most of the radionuclides released in the short term comes from relatively few tests where there is no volcanic cover above the cavity-chimney or where the cover is defective.
- Only a very small fraction of the radionuclide inventory escapes the geosphere into the accessible environment. The highest release rates (in terms of activity) are due to tritium but these are of no radiological significance. The release rates of other radionuclides are generally not detectable in the environment having regard for the large dilutions in the lagoons and the ocean. ❐