ANALYSIS OF RADIOACTIVITY IN THE ENVIRONMENT OF MURUROA & FANGATAUFA ATOLLS: THE IAEA LABORATORIES IN OPERATION

SCIENTIFIC TEAMWORK

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Shortly after the International Advisory Committee was set up to supervise the Study of the Radiological Situation at the Atolls of Mururoa and Fangataufa in French Polynesia, a number of scientific questions arose. Among them was the question of data collection and analysis. France had conducted 193 experiments on nuclear weapons between July 1966 and January 1996 at the atolls, and an independent sampling and analysis programme would be needed. The programme would serve a number of purposes — to evaluate the credibility of the available French data, and to establish whether French monitoring provided an adequate estimate of the concentrations and inventories of the relevant radionuclides in the terrestrial and marine environment so that sound dose assessments could be made.

The involvement of the IAEA's Laboratories in Seibersdorf, Austria, and in Monaco (MEL) was requested because of their extensive experience in monitoring environmental radioactivity in terrestrial and marine environments.

Both the terrestrial and marine sampling campaigns of the Study were conducted in 1996. To assess the logistic requirements for missions to such a remote area, a technical team from the laboratories visited the atolls in March 1996. It inspected potential sampling sites and laboratory equipment and facilities, and met with members of the French Liaison Office and local staff. Thereafter, the sampling and monitoring programmes were designed and discussed by the Mururoa Study's Task Group A, chaired by Dr. A. McEwan of the National Radiological Laboratory, New Zealand. The group included experts from Denmark, Japan, Fiji, Australia, United States, Austria, and the United Kingdom.

The campaigns were carried out 1 July to 2 August 1996. The terrestrial and marine groups were augmented to include a number of international experts. The analysis of radionuclides in the collected samples was conducted partly by the IAEA's laboratories and partly by a network of independent international laboratories.

THE TERRESTRIAL CAMPAIGN

The sampling programme was designed to obtain representative environmental samples. With the exception of some samples collected in the Colette region of the Mururoa atoll, where hot particles were investigated, care was taken in the preparation and processing of the samples to ensure that all radionuclides were distributed homogeneously in the final sub-samples. Moreover, each sample was split into three parts. One was measured by collaborating international laboratories, one was sent to the French, and a third was archived at Seibersdorf. The French protocols were reviewed by Seibersdorf scientific staff and other international experts. They were found to be adequate and closely followed in all sampling procedures in order to ensure reasonable comparability of the results with French data. The samples collected were then analyzed by the Seibersdorf Laboratory and by members of the IAEA's international network of Analytical Laboratories for Measuring Environmental Radioactivity (ALMERA). From this network of 53 laboratories, eleven were selected to carry out analyses of Mururoa and Fangataufa samples.

Locations and samples. Samples were collected from fifteen selected sites as close as possible to those sampled during French environmental

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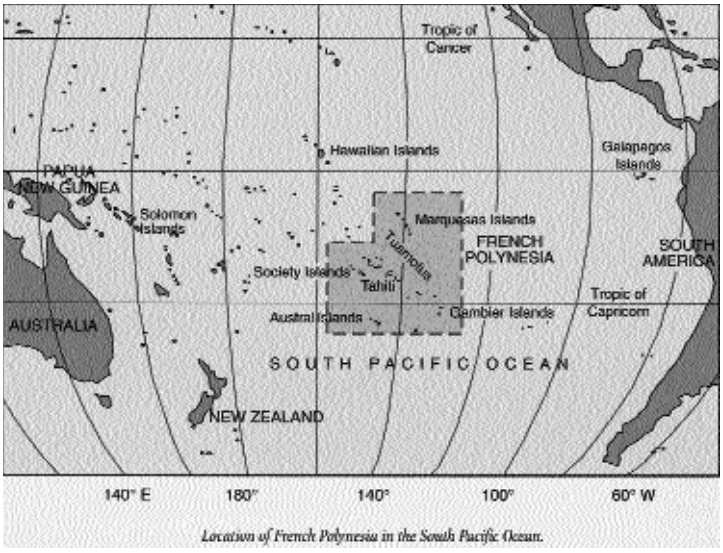
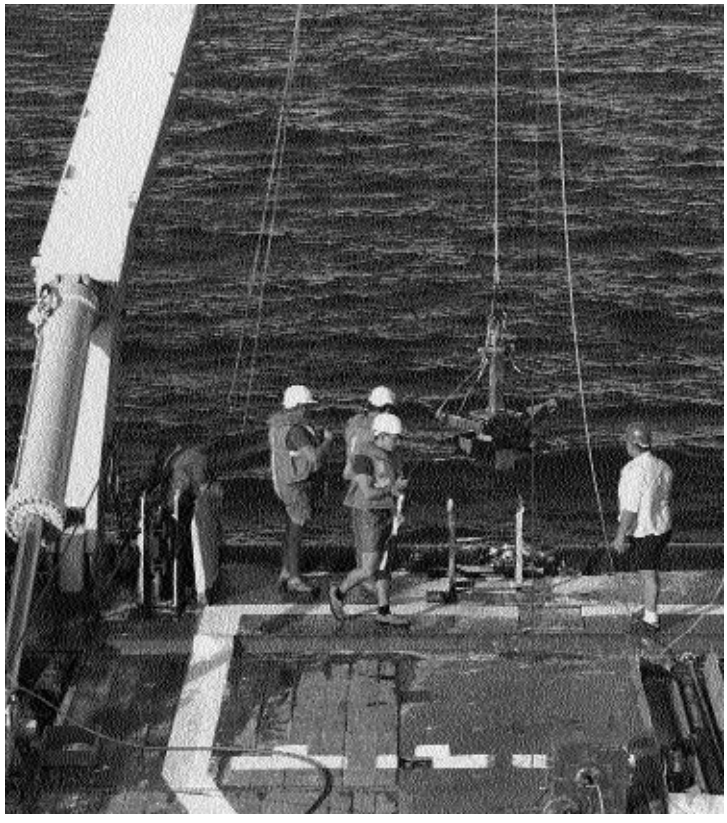


Location of French Polynesia in the South Pacific Ocean.



Map: Location of Mururoa and Fangataufa Atolls in the South Pacific. Study teams collected nearly 300 samples for analysis. Photos: From top right, members of the sampling team drill into a coral bedrock core in Fangataufa; rock samples are collected in Mururoa; water is sampled in Mururoa lagoon; a seabed gamma spectrometer is deployed in Mururoa lagoon; and sediment samples are collected in the lagoon on Fangataufa.

(Credits: IAEA Seibersdorf Laboratories/IAEA-MEL)

monitoring campaigns over the years. In addition some samples were collected on Tureia, the nearest inhabited atoll.

Several types of samples, amounting to a total of 299, were collected. After screening all samples for their activity content, 198 were analyzed. Air filter samples, utilized to collect aerosols or resuspended radioactive particles, were analyzed to evaluate the potential exposure due to inhalation. The filters were collected daily and measured in Mururoa for gross alpha and beta activity. They were then sent to the Seibersdorf Laboratories where they were measured for gamma activity and then ashed for alpha and beta activity determinations.

Samples of top soil and depth profiles were collected wherever possible to estimate the total surface contamination and to evaluate the transfer of radionuclides through soil.

In areas without soil, loose corals and sand samples were collected. Sand and loose rocks on the beach are subject to disturbance and mixing by waves and storms. Therefore random sampling, followed by mixing to give one composite sample representing the area as a whole, was considered as the most appropriate procedure to obtain unbiased information.

Cores of coral rocks were taken to investigate the downward migration of radionuclides originally deposited on the surface. The vertical concentration profile was also an important parameter for the calculation of the overall radionuclide inventory from *in situ* gamma spectrometry.

No vegetation or fruit are grown on Mururoa and Fangataufa, except for a few specimens in very small private gardens that utilize soil shipped from other parts of French Polynesia. Nevertheless, it was considered desirable to obtain some, although indirect, information on radionuclide transfer characteristics from soil to plants that might be grown by an hypothetical residing population. Therefore in the absence of vegetables, tree and bush leaves were sampled and analyzed as a substitute. More specific data were obtained for coconuts. Coconuts palms were introduced in Mururoa about one hundred years ago. As they play a very important role in the diet of the Polynesians, coconuts were collected in several parts of Mururoa, in Tureia and from a few isolated locations of Fangataufa. Coconuts were chosen by their ripeness, in order to have samples of both coconut water and flesh (copra).

At various locations of the two atolls, 106 *in situ* gamma spectrometric measurements with high purity germanium detectors were performed. They were done to collect information on surface and subsurface inventories of the radionuclides plutonium-239, americium-241, cobalt-60, caesium-137, and europium-155.

Gamma spectrometric analyses were carried out on 192 samples, while alpha and beta emitting radionuclides were analyzed in 178 samples. The measurements were performed in duplicate and triplicate as part of the validation and quality control measures. This resulted in a total of 941 individual determinations for alpha emitting radionuclides (plutonium-238, plutonium-239, 240 and americium-241), 78 determinations for strontium-90, and 2520 results for the different gamma-emitting radionuclides.

The entire process of sampling, preparation, storage and distribution was carried out under stringent quality assurance measures.

RESULTS OF THE TERRESTRIAL CAMPAIGN

From the very beginning it was recognized that a campaign lasting only a few weeks could only provide a relatively limited sampling. In turn, this would inevitably lead to some differences in radionuclide concentrations between the spot samples taken during the campaign and the much larger data sets accumulated by French monitoring campaigns over several years. Based on the IAEA's past analytical experiences, some differences were expected for various reasons, and therefore overlaps in the range of values could be regarded as a good measure of agreement.

Results of the Mururoa Study were compared with the French results in terms of activity ranges. Summary views of the ranges observed for plutonium-239, 240 and caesium-137 show that for the majority of the combinations of locations and sample types, the ranges are overlapping and hence in satisfactory agreement. Nevertheless a few disagreements were observed. Although they are of minor significance in terms of radiological hazard, it is worthwhile to discuss them briefly.

In the location Irene (western end of the airfield), the measured aerosol activity concentrations for plutonium isotopes were 2 mBq/m^3 while the French reported a value of 0.11 mBq/m^3 , measured at the eastern end of the airfield (Kathie). This can be attributed to the fact that the French had used some contaminated debris from the Colette area in the construction of the airport. Therefore some particle resuspension, leading to occasional high activity in some air samples, was not unexpected.

In the Faucon area, three top soil samples showed plutonium-239,240 concentrations between 1200 and 1600 Bq/kg, i.e. considerably higher than the French values which ranged from 0.6 to 360 Bq/kg. This difference can be attributed to the small-scale heterogeneity of the radionuclide distribution in this area.

Near the area where safety trials were conducted (Colette region), hot spots and hot particles containing plutonium-239 and americium-241 were identified. *(See related article, page 43.)* The measured surface concentrations of plutonium-239 and americium-241 were found to be about two to six times higher than the 1987 French values. They fell in the range of 1 to 3 MBq/m^2 for plutonium-239, and 20 and 70 kBq/m^2 for americium-241.

The difference was attributed to the different measurement techniques used by the IAEA international team and the French scientists. The international team, which conducted a more limited series of measurements, employed *in situ* gamma spectrometry, which is more time consuming, and used a high resolution germanium detector placed at a fixed distance from the ground (one meter), taking advantage of the different gamma energies of plutonium-239 and americium-241. Therefore the activity levels obtained by the IAEA team can be considered more representative of the situation at the time.

These measurements also showed that the activity ratio of plutonium-239,240 to americium-241 clustered around the two values of 45 and 60. This probably reflected the different age and purity of the plutonium used in the safety trials. However, it must be pointed out that the external dose rates calculated from these *in situ* gamma spectrometry measurements amounted only to a few hundred microSv per year.

In the location Kilo on the Fangataufa atoll, the international team measured the highest concentrations of caesium-137 and strontium-90 in coconut flesh and water. These activity levels can be considered irrelevant from the standpoint of radiological hazards: they are far lower (by a factor of three) than the natural activity levels of potassium-40 found in all the samples of coconut water.

On the inhabited atoll of Tureia, the team measured activity levels of plutonium-239, 240, caesium-137 and strontium-90 in soil, vegetation and coconut samples. In general, they were all found to be low and of no radiological concern.

In conclusion, the activity concentrations measured by the Study's terrestrial sampling campaign were found consistent with the French data. As such, they could be considered a comprehensive and reliable account of the levels of artificial radioactive materials in the terrestrial environment of the atolls.

THE MARINE CAMPAIGN

The terms of reference of the Marine Working Group coordinated by MEL included a review of the data provided by the French authorities on radionuclide distributions in the littoral and sub-littoral environments at the atolls. The twofold objective was to conduct sufficient and new independent monitoring work at and around the atolls in order to validate existing French data. At the same time, the monitoring would provide a representative and high-quality data set on current radionuclide concentrations in the marine environment.

The aquatic sampling expedition to Mururoa and Fangataufa was organized from 1-27 July 1996. Work was arranged to collect relevant and comparable samples of water, biota, and sediment from the lagoons of the atolls, as well as from the ocean surrounding the atolls. The results obtained, combined with earlier IAEA intercomparison data evaluated in 1994, were used for the validation of the far more numerous data from French monitoring campaigns.

Five researchers from laboratories in IAEA Member States and four from MEL participated in the marine sampling campaign. The team operated in parallel on five

vessels simultaneously. The pre-treatment of samples was carried out in four independently managed laboratories in Mururoa.

As in the terrestrial campaign, all samples were collected in triplicate. Over 300 samples were collected (from lagoon water, ocean water, sediment pore water, sediment, corals and biota). Some 13,000 litres of water and one tonne of solid samples were collected, processed, packaged and transported to Monaco for distribution to ten analytical laboratories in eight countries.

Underwater gamma spectrometry was used for *in-situ* measurements of sediment gamma activity. The survey's purpose was to identify the most contaminated areas and to guide subsequent sample collection. It was carried out inside the lagoons of Mururoa and Fangataufa Atolls. Since the rugged topography of the bottom does not allow towing of any benthic devices, the measurements were done on a discrete network of points. These were mostly constituted in transects crossing the areas of highest contamination that were identified by the French.

The team's survey evaluated gamma count rates due to cobalt-60 and caesium-137, whose distributions in lagoon sediments, due to different origins and geochemistry, are not necessarily the same as for plutonium. Previous French investigations showed, however, that they correlate with the plutonium distribution patterns.

Following the survey, samples were taken from the sites identified as being local contamination maxima. Later calibration and correlation analyses were carried out to calculate cobalt-60 and caesium-137 inventories from *in-situ* measurements, as well as to estimate cobalt-60 and plutonium-239,240 inventories in lagoon sediments.

The highly sophisticated underwater gamma spectrometry proved to be an efficient technique for the purpose of on-site identification of areas with highest contamination of the seabed. The process otherwise would have required a time-consuming sequence of sampling, sample preparation, and counting.

A network of marine laboratories carried out the analytical work. In addition to IAEA-MEL, the network included six internationally renowned laboratories providing high-quality data in Australia, Denmark, Germany, New Zealand, the UK, and the United States. All of them had participated in regular intercomparison exercises organized by MEL, and in their particular work they had shown good analytical competence in specially organized proficiency tests.

RESULTS OF THE MARINE CAMPAIGN

The marine campaign corroborated extensive data already available and provided additional scientific information. The activity concentrations of radionuclides in the aquatic environment were found to be generally low and comparable with reported French data.

Residual strontium-90 and caesium-137 were measurable in the environment of both lagoons, but concentrations were very low. Typical activities were: in lagoon waters, about 2 Bq/m^3 (only marginally above the level prevailing generally in the South Pacific due to global fallout); and in fish from the lagoons, less than 0.3 Bq/kg for caesium-137 and much lower for strontium-90 (in comparison, the activity of naturally occurring radioactive potassium-40 in fish is of the order of 100 Bq/kg).

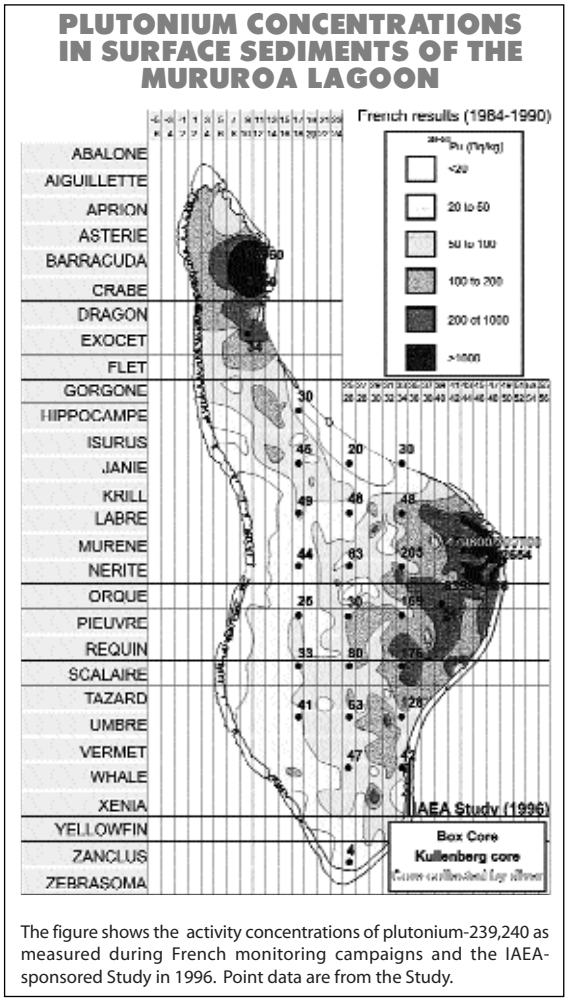
For hypothetical residents of Mururoa Atoll, the most significant contributor to doses would be plutonium-239,240. Plutonium levels were around 0.3 Bq/m^3 in the lagoon waters, 0.01 Bq/kg in fish, 0.08 Bq/kg in crustacea and 0.8 Bq/kg in molluscs from the lagoon (about 100 times the concentrations of these radionuclides in the ocean and comparable ocean biota arising from global fallout). However, in radiological terms, these activity concentrations are all very low and have no radiological significance.

Analysts found evidence of a time trend in the concentrations of tritium, strontium-90, caesium-137 and plutonium-239,240 in lagoon water. Confinement was not equally effective for all 137 underground nuclear tests, and it is apparent from the elevated tritium concentrations that have persisted for some years that some leakage of tritium has been occurring into both lagoons. This was confirmed by data from the French Liaison Office on levels of tritium and its distribution in the carbonate rock of both atolls.

Measurements made in 1996 indicate that tritium levels in the lagoons may be beginning

to fall (this decline may, however, be reversed for the lagoon at Mururoa as more tritium from underground nuclear tests migrates to the lagoon water). The strontium-90 levels at present may be showing a slight increase. Levels of caesium-137 and plutonium-239,240 have been falling for some years, faster than would be expected through radioactive decay alone.

The dominant radionuclide observed in lagoon sediments was plutonium-239,240. Its surface concentrations ranged from about a few tens of Bq/kg of sediment in the central part of the lagoons to a few thousands of Bq/kg in Denise and Dindon on Mururoa and Fregate on Fangataufa hot spots that were left after barge tests. The highest plutonium-239,240 concentrations were observed in Colette bank sediment (up to about 500,000 Bq/kg) that was contaminated by safety trials.



The figure shows the activity concentrations of plutonium-239,240 as measured during French monitoring campaigns and the IAEA-sponsored Study in 1996. Point data are from the Study.

OVERALL CONCLUSIONS

Overall, the Mururoa Study's sampling campaigns found that the terrestrial and aquatic environments of Mururoa and Fangataufa atolls that are accessible to people contain residual radioactive material attributable to the nuclear tests. However, these are at generally low concentrations which are of no radiological significance.

There are, however, some features of note whose radiological implications may be of interest:

■ Generally, there was reasonably good agreement between the Study's results and the French data. A decline in radionuclide concentrations in the lagoons was observed in comparison with the French data. Radionuclide concentrations in biota were low and consistent with previous IAEA and French data.

■ Particles containing plutonium and small amounts of americium resulting from atmospheric safety trials remain in the area of the trial sites — the motus of Colette, Ariel and Vesta on Mururoa Atoll.

■ Elevated levels of caesium-137 (about 1kBq/kg) were found over small areas totalling several hectares on the Kilo-Empereur rim of Fangataufa.

■ Several kilograms of plutonium resulting from atmospheric nuclear tests at the atolls remain in sediments under the lagoon of each atoll. Some of the plutonium in the sediments of the Mururoa Atoll lagoon came from the atmospheric safety trials. The total inventory of plutonium-238 and 239,240 in both lagoons was estimated at about 30 TBq. Inventories of americium-241, caesium-137, europium-155 and cobalt- 60 were below 1TBq (each).

■ The concentrations of tritium, strontium-90, caesium-137 and plutonium-239,240 in each lagoon were found to be higher than in the open ocean, as the result of leaching of sediment (the last three radionuclides) and contributions from underground sources (first two radionuclides). The estimated radionuclide release rates from both lagoons to the open ocean were (in TBq/year): 6 for tritium, 0.03 for strontium-90, 0.01 for caesium-137 and 0.01 for plutonium 238 and plutonium-239,240.

■ Distribution coefficients and concentration factors applied for both lagoons were found to be within ranges of IAEA-recommended values.❐