

WORLDWIDE MARINE RADIOACTIVITY STUDIES ASSESSING THE PICTURE

BY PAVEL P. POVINEC AND ORIHIKO TOGAWA

A growing number of sources of radioactivity from human activities are found in the marine environment. They are known to include global nuclear fallout following atmospheric weapons tests, the Chernobyl accident, discharges of radionuclides from nuclear installations, past dumping of radioactive wastes, nuclear submarine accidents, contributions from nuclear testing sites, loss of radioactive sources, and the burn-up of satellites using radioisotopes as power sources.

Overall, the world's marine environment contains radionuclides that differ from one region to another. Differences are due to dynamic marine environmental processes and the particular source of radionuclides in a region. Scientific assessments of marine radioactivity, therefore, require knowledge of both the source terms and oceanic processes.

Radioactivity now is deposited unevenly over the world's oceans. Global fallout is known to be mainly due to nuclear weapon tests carried out in the 1960s. On the other hand, discharges from nuclear fuel reprocessing plants or past dumping of liquid and solid radioactive wastes generally are confined to more localized areas. Even so,

soluble radionuclides have been transported over long distances by prevailing ocean currents.

To estimate radionuclide inputs from local sources, scientists need to better understand the distribution of radionuclides throughout the world's oceans and seas. The understanding is important for analyzing the results from scientific investigations of localized areas, such as past dumping sites, which then can be reviewed more thoroughly.

As a contribution to fuller understanding of the marine environment, the IAEA's Marine Environment Laboratory (MEL) started a five-year project in 1996 entitled "Research on Worldwide Marine Radioactivity (MARS)". The work is supported by Japan's Science and Technology Agency (STA). This article briefly reviews this project, and describes related research activities and scientific investigations of MEL and its global partners.

RESEARCH INITIATIVES

MARS seeks to provide new data on current radioactivity in the open ocean and to develop a better understanding of the present radionuclide distribution. Data sets

obtained in national and international radioactivity surveys thus can be reviewed comparatively. The project contributes to scientific knowledge of the processes affecting radionuclide distribution and the sources that have introduced radioactivity to the open ocean.

An Experts' Meeting organized in 1996 in Monaco has helped guide the project. Experts at the meeting recommended studies in the North Pacific Ocean and the Atlantic Ocean, with supplementary sampling in the Indian Ocean. They further advocated that the work in the North Pacific Ocean should receive the highest priority as there was a lack of new data, and recommended that efforts should be made to locate the sampling stations at sites where comprehensive and good quality historical data existed. These include, for example, the sites used in the GEOSECS (Geochemical Ocean Sections Study) programme in the mid-1970s or at sites where a suite of measurements could provide

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linkages to extrapolate the results to contemporary ocean-wide surveys.

The project's specific objectives are to:

- examine present distributions of key radionuclides (tritium, carbon-14, strontium-90, caesium-137, and plutonium isotopes) in water, sediment and biota of the world's oceans;
- extrapolate the results from contemporary ocean-wide surveys, such as the tracer component of the WOCE (World Ocean Circulation Experiment Hydrographic Programme), where large sets of tritium data are available and could be used for predictions of strontium-90 and caesium-137 concentrations in the same regions;
- study the development of radionuclide concentrations in water with time using good quality historical data with new data sets collected recently;
- identify the major sources of anthropogenic radionuclides in the world's oceans;
- add all available data on radionuclide distributions in water, sediment and biota into the Global Marine Radioactivity Database, so that temporal and spatial variations of key radionuclides can be investigated.

Coordinated Research Programme (CRP). A CRP on "Worldwide Marine Radioactivity Studies" was launched in 1998 to support the research activities of the project. Participants include experts from Denmark, Germany, India, Italy, Japan, the Republic of Korea, and the United States. Their work will contribute to understanding

the present open ocean distributions of radionuclides in the water column and sediment and to predict the radiological impact. The CRP, which receives extrabudgetary support from Japan, will encourage and support marine radioactivity studies in Member States by methodological assistance and the management of analytical quality. All data obtained through the CRP on radionuclide concentrations in water, sediments, and biota will be available to IAEA Member States in electronic form, both on CD-ROM and over the Internet.

SCIENTIFIC EXPEDITIONS

The MARS project additionally is supported by a number of scientific expeditions to the open ocean.

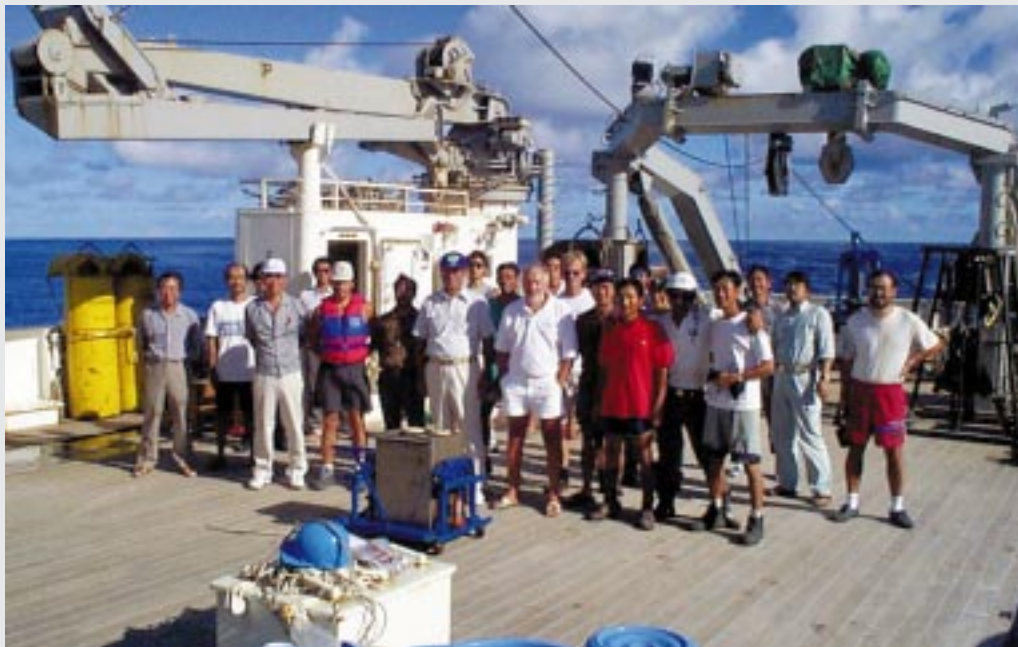
IAEA '97 Pacific Ocean Expedition. This expedition was conducted by MEL in cooperation with nine organizations from five IAEA Member States.* (See box.) It was the first time that an oceanographic expedition to the open ocean was fully organized and accomplished by the IAEA.

The expedition was conducted to the northwest Pacific Ocean from 21 October to 20 November 1997. Its main objective was to sample water at various depths, to sample sediment cores and biota, and to perform oceanographic measurements (salinity, temperature, etc.). The analyses of collected samples and oceanographic measurements performed during the cruise will enable investigation of the

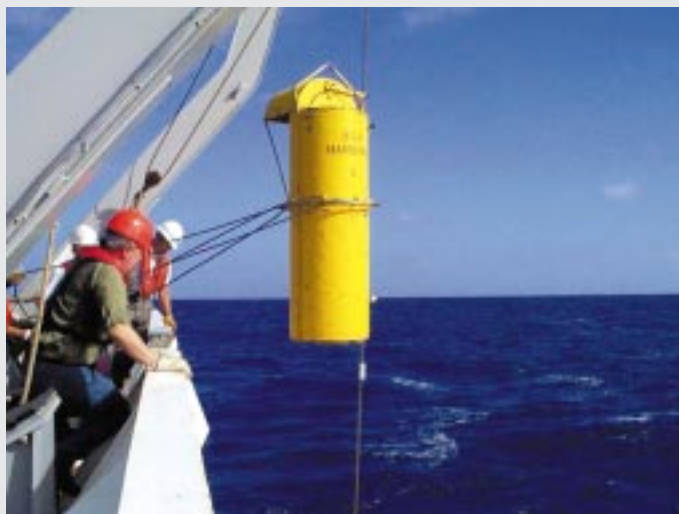
distribution of anthropogenic and natural radionuclides in the marine environment. Additionally, the work will help to study isotopic signals which may have been introduced by the "El Niño" effect. Further, the investigations will look for possible latitudinal effects in the distribution of natural radionuclides in seawater, plankton, and fish.

The expedition was conducted using the research vessel *Bosei Maru* owned by Tokai University. The cruise followed a triangular route from Shimizu (Japan) eastward to Milwaukee Sea-Mountain, then southward to the Marshall Islands, calling at Pohnpei (the Federated States of Micronesia) and back to Shimizu. The sampling work was carried out at 20 stations, including four GEOSECS stations and seven stations close to Bikini and Enewetak Atolls.

Seawater samples were collected at several depths and at different stations using large volume water samplers, submersible pumps and a CTD (conductivity, temperature and depth) system with rosette multi-bottle samplers (CTD/RMS). Sediment and biota were sampled by a box corer and fishing rods/nets, respectively. CTD and general chemistry measurements were also carried out during the expedition. In total, five sets of large-volume water samples, twelve sets of small-volume water samples, fifteen cores of sediment samples, forty-five fish samples of three different species, and plankton samples were collected.



At right, seawater samples are drawn from the Japanese ship Bosei Maru during the IAEA '97 Pacific Ocean Expedition. Scientists from five countries and nine organizations participated. They included the Federal Maritime and Hydrographic Agency, Germany; the Korea Ocean Research and Development Institute, the Republic of Korea; the Physical Research Laboratory, India; University of Linköping, Linköping, Sweden, and five Japanese organizations: the Japan Atomic Energy Research



Institute - Mutsu Establishment; the Japan Chemical Analysis Center; the Japan Marine Science Foundation - Mutsu Marine Laboratory; the Meteorological Research Institute; and Tokai University. Altogether, twenty-two participants, including six IAEA staff members from MEL, took part in the expedition. Some of the participants are shown above with crew members. (Credit: MEL)

Pre-concentration chemistry for several selected radionuclides (strontium-90, caesium-137, plutonium-239, 240, and americium-241) was carried out on board to reduce the volume of large-volume water samples by a factor of

about twenty. This enabled shipment by surface freight from Shimizu to Monaco and to laboratories of the participating Member States for subsequent analysis.

Concentrations of tritium, carbon-14, technetium-99,

iodine-129, and plutonium (ICP-MS) will be determined in seawater samples collected by using a CTD/RMS system. For sediment and biota samples, on the other hand, measurements will be made not only of these

anthropogenic radionuclides but also natural radionuclides (polonium-210, radium-226, thorium-230, uranium-234, and uranium-238).

Preliminary results obtained by CTD and general chemistry measurements were presented in a cruise report that MEL issued soon after the expedition. The full analyses of collected samples is being done by scientists at MEL, the participating organizations, and other laboratories of IAEA Member States (Canada, Germany, India, Japan, New Zealand, and the United States). Final results of all analytical work is planned for publication in 1999.

Japanese-Korean-Russian joint expeditions. Within the framework of an agreement between the Governments of Japan, the Republic of Korea, and the Russian Federation, MEL was invited to participate in joint expeditions to former radioactive waste dumping areas in the northwest Pacific Ocean and its marginal seas. The objective was to investigate the radiological contamination of the marine environment due to radioactive wastes dumped in the past by the former USSR, the Russian Federation, the Republic of Korea, and Japan.

The expedition was carried out on board the Russian research vessel *Okean* in two stages: the first from 22 March to 11 April 1994, visiting dumpsites in the Sea of Japan/East Sea, and the second from 15 August to 15 September 1995, visiting dumpsites in the Sea of Okhotsk, the northwest Pacific Ocean and the Sea of Japan/East Sea.

Seawater from surface and bottom waters and seabed sediments were sampled at fourteen stations in the areas where radioactive wastes of more than 700 TBq had been dumped and at an additional six stations outside these areas. In two areas where larger amounts of radioactive wastes had been dumped, intermediate water was also collected at several depths. In addition, sampling work was done at two stations in an area close to Sakhalin Island in the Sea of Okhotsk, where a strontium-90 source of 13 PBq was accidentally lost.

MEL scientists carried out measurements of tritium, carbon-14, strontium-90, iodine-129, caesium-137, plutonium-238, plutonium-239, 240 and americium-241 in collected samples. Results showed that the concentrations of tritium, strontium-90, caesium-137, plutonium-238, and plutonium-239, 240 in seawater and sediment of the dumping areas were low and not significantly different from those found in corresponding background areas. The levels also compared well with those generally found in the northwest Pacific Ocean and its marginal seas.

Inventories of strontium-90, caesium-127, and plutonium-239, 240, which were estimated from vertical profiles in seawater and sediment, showed a surplus compared with the cumulative deposition densities of global fallout. The estimated inventories, however, were consistent with previous observations in these regions. Results of radionuclide inventories and isotopic activity ratios supported the

conclusion of the predominance of global fallout at the sampling areas.

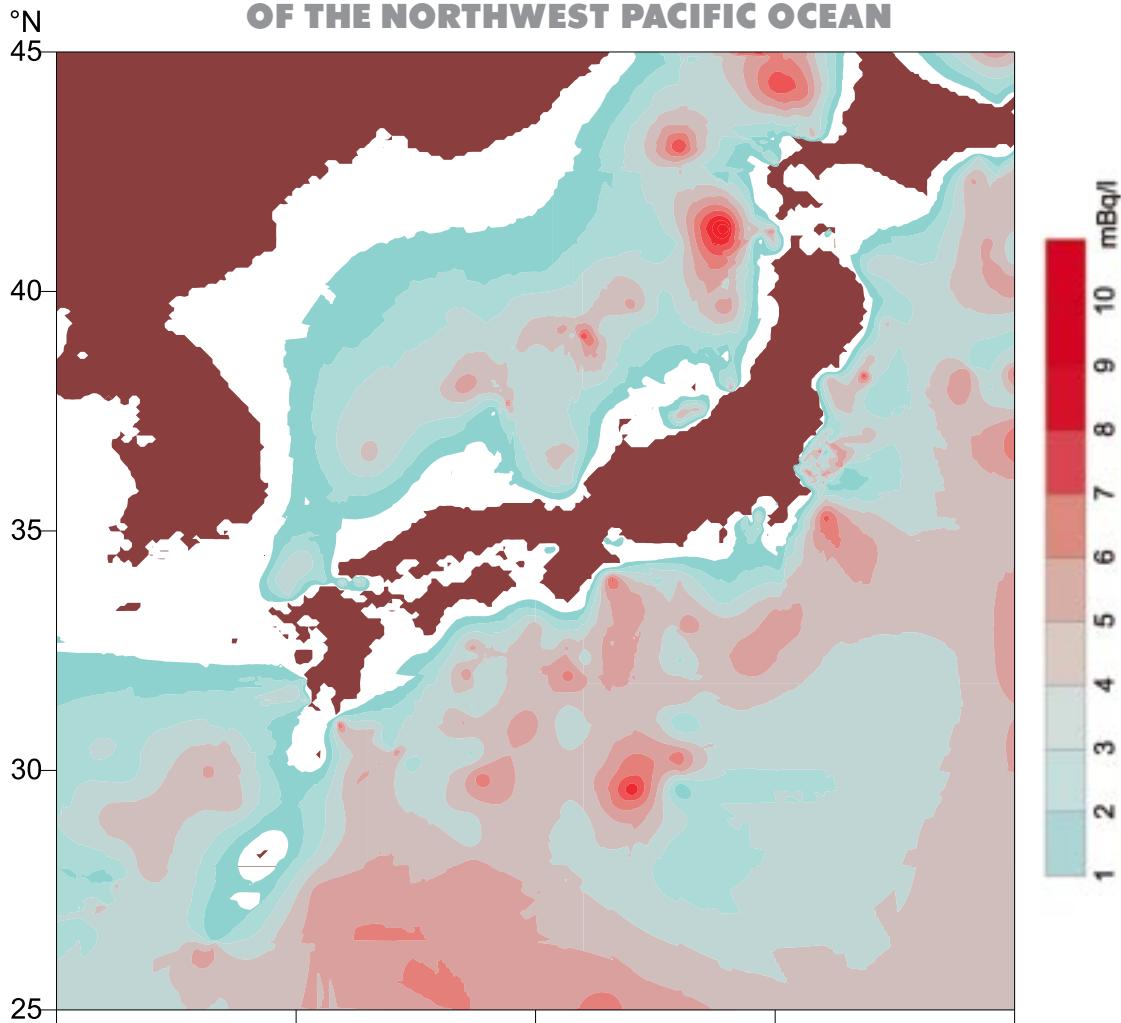
Indian Ocean expeditions. MEL scientists participated in two scientific expeditions to the Indian Ocean.

The first one was conducted in collaboration with the National Agency for New Technology, Energy and Environment (ENEA), Marine Environment Research Center, La Spezia, Italy. The objective was to study the distribution of anthropogenic radionuclides in surface seawater in the Indian Ocean. The expedition took place from 8 March to 9 April 1998 on board the Italian research vessel *Italica* on its way back to Italy from the Antarctica.

Seawater was collected at forty-one sampling stations, approximately every five degrees in longitude. The transect extended from Lyttelton in southeast New Zealand to Ravenna in Italy, crossing Cook Strait, the Tasman Sea, Bass Strait, the Great Australian Bight, the Indian Ocean, the Gulf of Aden, the Red Sea, the Suez Channel and the Mediterranean Sea. Seawater was sampled at an average depth of four meters using the pumping system installed on the vessel. On-board pre-concentration chemistry for several selected radionuclides (strontium-90, caesium-137, plutonium-239, 240 and americium-241) was carried out using the same method employed during the IAEA '97 Pacific Ocean Expedition.

Additional small-volume seawater samples to be processed later at MEL were sampled for tritium and

CONCENTRATION OF CAESIUM-137 IN SURFACE SEAWATER OF THE NORTHWEST PACIFIC OCEAN



iodine-129 analysis as well as for archiving. The analysis of these samples will be carried out at MEL and ENEA, with results available in 1999.

The second expedition to the Indian Ocean was undertaken at the invitation of India's Department of Ocean and Development (DOD). The Physical Research Laboratory (PRL) in Ahmedabad organized the expedition, which took place from 19 March to 20 April 1998 using the DOD research vessel *Sagar Sampada*. The main objective was to determine concentrations and

distributions (horizontal and vertical) of anthropogenic radionuclides in water and sediment.

Sampling work was carried out at six stations, including five GEOSECS stations. At each station, seawater samples were collected at six different layers using a CTD/RMS system. Several selected radionuclides (strontium-90, caesium-137, plutonium-239, 240 and americium-241) were pre-concentrated on board. Sampling of additional seawater was done for the determination at MEL of tritium, technetium-99,

iodine-129, and plutonium (ICP-MS), as well as for archiving. Sediment cores and plankton samples also were collected at a number of stations.

Analyses of all collected samples will be carried out at MEL and PRL, with results available in 1999. Further sampling expeditions to the

Map: IAEA marine scientists at MEL in Monaco have participated in scientific expeditions yielding valuable information about levels of radioactivity in the world's oceans. (Credit: MEL)

Atlantic Ocean and the South Indian Ocean are scheduled to take place in 1998 and 1999.

MARINE RADIOACTIVITY DATABASE

In the framework of the research project, MEL has developed GLOMARD (the Global Marine Radioactivity Database) to provide Member States with data on radionuclide concentrations in the marine environment. This information will help them in radiological assessments related to radioactive waste dumping and nuclear testing, and in emergency response to radiological accidents at sea. It is planned to make GLOMARD available in the near future over the IAEA's Internet site.

The database contains approximately 40 megabytes of data, representing 100,000 measurements of radioactivity in seawater, sediment, biota and suspended matter sampled from the world's oceans.

It contains information on the sources of the data; the laboratories performing radionuclide analysis; the type of samples (seawater, sediment, biota) and associated details (such as volume and weight); the sample treatment, analytical methods, and measuring instruments; and the analyzed results (such as radionuclide concentrations, uncertainties, temperature, salinity, etc.)

The current version of the GLOMARD allows the input, maintenance and extraction of data for the production of various kinds of maps using external computer programs. Extracted data are processed by

these programs to produce contour maps representing radionuclide distributions in studied areas.

The new version of GLOMARD will be connected to the Geographical Information System. This will allow the production of more precise two- and three-dimensional maps. On them, radioactivity data will be correlated with data on bathymetry, temperature or salinity. It will also be possible to produce time-series contamination maps.

RADIONUCLIDE DISTRIBUTION

Applying such mapping capabilities, the distributions of radionuclides in surface seawater and surface layer sediment of the northwest Pacific Ocean were investigated using data sets installed in GLOMARD. (*See map.*) The data is from Japanese institutions, mainly from the Japan Chemical Analysis Center (JCAC). Evaluation was carried out for the vertical profiles of radionuclide concentrations, radionuclide inventories, and their isotopic activity ratios in both seawater columns and sediment cores.

Generally the concentrations of strontium-90, caesium-137, and plutonium-239, 240 in marine samples collected around Japan are very low and show a tendency to decrease year by year. The profiles of strontium-90 and caesium-137 in seawater columns show a gradual decrease in concentration with increasing depth, contrasting with those of plutonium-239, 240. The plutonium profiles show a subsurface maximum at

around a depth of 700 meters, reflecting specific scavenging processes in the water column.

The concentrations of radionuclides in sediment cores tend to decrease with increasing depth. The inventories of strontium-90, caesium-137, and plutonium-239, 240 in seawater and sediment were estimated for several periods from the beginning of the 1980s up to the present. Isotopic activity ratios in sediment cores are quite different from those in the seawater columns, reflecting the difference in the removal of radionuclides from seawater to bottom sediment.

The inventories of strontium-90, caesium-137, and plutonium-239, 240 in the water column in the Sea of Japan/East Sea dumping site are 3.0, 5.8, and 0.10 kBq/m², respectively. In comparison, the respective inventories in the water column in the northwest Pacific Ocean are 1.0, 2.0, and 0.11 kBq/m², respectively. As can be seen, the strontium-90 and caesium-137 inventories in water at the Sea of Japan/East Sea dumping site are significantly higher than those observed in the northwest Pacific Ocean.

The total strontium-90 and caesium-137 inventories in water and sediments are in good agreement with expected global fallout deposition densities. On the other hand, those for plutonium-239, 240 and americium-241 in the northwest Pacific Ocean show a surplus, possibly additional input from close-in fallout transported from the Central Pacific Ocean.

As a result of surveys in the Sea of Japan/East Sea and the northwest Pacific Ocean, there is no definitive evidence of the effect of radioactive waste dumping. However, unique oceanographic features in the behavior of radionuclides have been found.

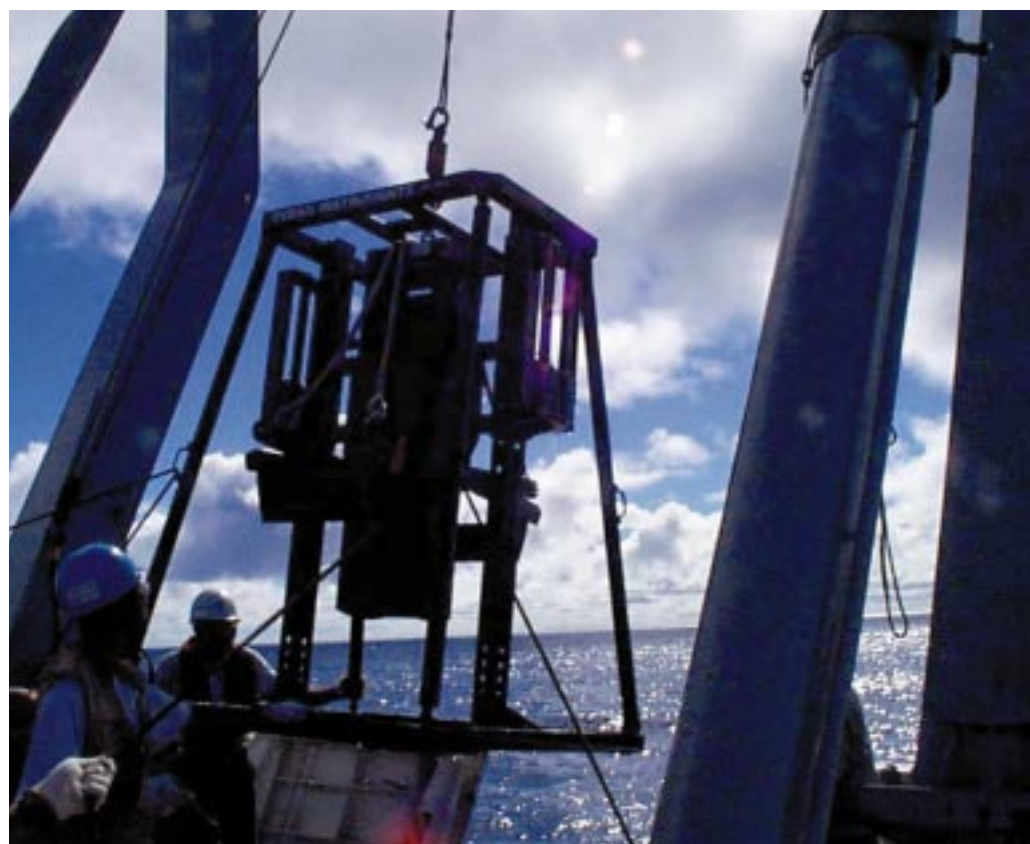
Collective effective dose equivalents (CEDE) were estimated for the Japanese population by the marine food pathway from liquid radioactive wastes dumped in the Sea of Japan/East Sea. A total of 443 TBq of liquid wastes was dumped off Vladivostok.

For the radionuclide composition of the wastes, it was assumed that the ratios of cobalt-60, strontium-90, and caesium-134 to caesium-137 were 0.01, 0.3 and 0.01, respectively. The doses due to long-lived radionuclides had maximum values four to five years after the disposals.

The total CEDE from all radionuclides had a maximum in 1990 of 0.8 man Sv. About 90% of the dose comes from caesium-137, most of which is due to consumption of fish. The collective effective dose equivalent commitment is estimated to be 11 man Sv.

Dose estimates due to global fallout (strontium-90, caesium-137, and plutonium-239, 240) and the natural radionuclide (polonium-210) were also made for comparison with doses from liquid radioactive wastes. CEDEs were estimated from the annual intake of marine products caught in 1990.

The collective dose of 17,000 man Sv from polonium-210 predominates, compared with that of 16 man



Sv from strontium-90, caesium-137, and plutonium-239, 240. The total CEDE from the liquid radioactive wastes is about 5% of that from global fallout.

OUTLOOK

Further investigations are required to more fully assess radioactivity in the marine environment. Research and analysis at MEL will focus on the sources that have introduced radionuclides into the world's oceans and seas; the distribution of anthropogenic and natural radionuclides in the marine environment; and the contribution of anthropogenic and natural radionuclides to doses that the world population receives from marine radioactivity through ingestion of seafoods. The studies will help to clarify the

respective contributions from different sources of radioactivity, with results supporting decision-making in various fields.

An important forum will be the International Symposium on Marine Pollution in Monaco in October 1998, which the IAEA is sponsoring jointly with a number of bodies. (See box, pages 4 & 5, for an overview of Symposium objectives and topics.) Leading national and international scientists in the field will examine a wide range of issues, and help to set priorities for the coming years. □

Photo: Marine scientists deploy a box corer for collecting samples of sediment on the seabed.

(Credit: MEL)