

Chernobyl & the marine environment: The radiological impact in context

Scientists at the IAEA's Marine Environment Laboratory in Monaco have played an integral role in post-Chernobyl studies

by
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The Chernobyl nuclear accident in April 1986 had a significant impact on both the terrestrial and marine environments. The total activity of the nuclear debris released was so high (1-2.10¹⁸ becquerel) that the radioactive fallout distributed widely after the accident actually dominated anthropogenic environmental levels in various parts of the world.

Concentrations of anthropogenic radionuclides generally vary from region to region, according to the location and magnitude of the different sources of contamination. The main global contribution to marine radioactivity, as in the terrestrial environment, is still from fallout from nuclear tests in the atmosphere, particularly during the 1950s and 1960s.

However, in some regions, like the Irish and North Seas, the concentrations of anthropogenic radionuclides (e.g. caesium-137 and plutonium-239) in the marine environment have been significantly influenced by discharges (e.g. from European reprocessing plants). On the other hand, the Baltic and Black Seas have been the seas most affected by the Chernobyl accident. In all these latter regions the spatial and temporal trends in the concentrations of anthropogenic radionuclides have been quite dynamic. They are a result of changing source terms and marine processes, including horizontal and vertical transport in seawater, marine sedimentation, re-suspension from sediment and biological uptake, and food-chain transfer.

IAEA-MEL tracer studies

The Chernobyl accident, perhaps surprisingly, was of considerable interest to oceanogra-

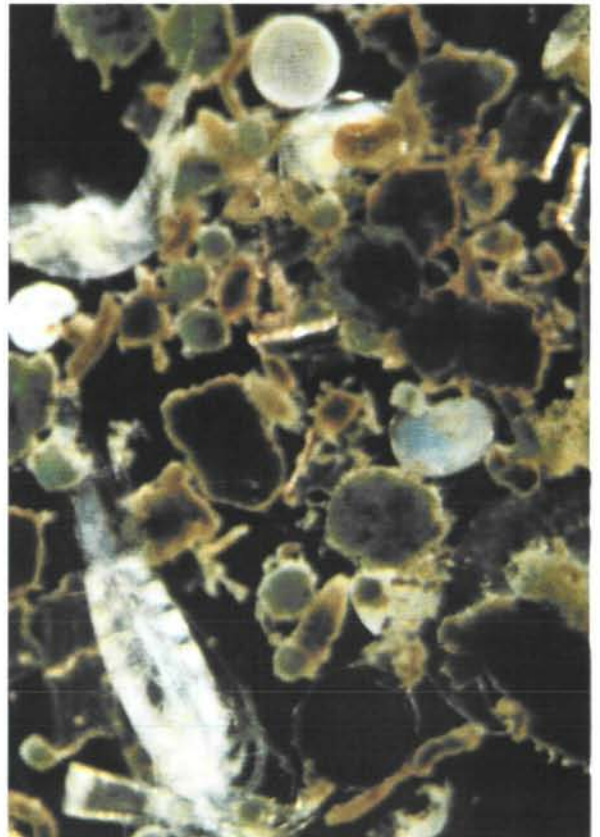
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phers around the world. The accidental release of substantial amounts of radioactivity to the atmosphere essentially initiated a worldwide transient tracer experiment on a scale that would never have been planned deliberately. Shortly after the accident, fission and activation products released by the fire entered marine waters throughout Europe. They became involved in many of the elemental cycles that oceanographers have for decades been trying to characterize using a wide variety of conventional techniques. Suddenly, immediately after the Chernobyl accident, a suite of radioactive tracers became available as a pulse to trace, rather like a coloured dye, the movement of elements through the oceans. IAEA-MEL scientists took part in this exciting and serendipitous experiment through temporal radionuclide monitoring of both the coastal and open ocean ecosystems.

For open sea work, one of the most important innovations in marine monitoring over the past 15 years has been the development of sediment traps to directly measure fluxes of materials associated with sinking particles. Moored sediment traps can be left unattended at any depth in the ocean to collect discrete, time-series samples at pre-determined intervals.

As part of a joint French-IAEA study of open Mediterranean particle flux, in mid-April 1986, IAEA-MEL scientists had, by a happy coincidence of timing, moored their automated time-series sediment trap at a depth of 200 meters in the Ligurian Sea between Monaco and the island of Corsica. In order to study timescale changes in particle flux, each of the six trap collection cups was set to sample sinking particles for consecutive periods of 6.25 days. Following the accident on 26 April, atmospheric measurements made at Monaco by IAEA-MEL indicated that most of the peak Chernobyl fallout entered the Ligurian Sea essentially as a single pulse during the period 4-5 May.

The sediment trap was retrieved on 22 May and the particulate material, along with other



Top: As part of an IAEA-MEL training course in Istanbul in November 1994, scientists collect sediment samples in the Marmara Sea using the University of Istanbul's ship. *Left:* A team of IAEA-MEL scientists deploy a sediment trap such as the ones used during their post-Chernobyl marine studies. *Above:* Typical particles collected in sediment traps include oval, rectangular, and cylindrical fecal pellets produced by zooplankton that are actively feeding on micro-organisms. (Credits: IAEA-MEL).

marine samples, was analyzed for radioactivity by gamma spectrometry. The radioanalyses showed that the primary pulse of particulate radionuclides arrived at 200 meters depth between 8 and 15 May, that is only about seven days after peak radioactivity was delivered to the sea surface. The time lag implied an average sinking speed of the radioactive particles of approximately 30 meters per day. This pulse of radioactivity sinking through 200 meters was particularly evident for the particle-reactive fission products (e.g. zirconium, niobium, and cerium radionuclides) which were either not detectable or present in very low amounts in the last sediment trap sample collected after 15 May.

The rapid descent of these radionuclides on a timescale of a few days indicated that they were not sinking as fallout particles according to Stokesian settling models. Rather, they were incorporated into large aggregates which are known to sink at speeds of tens to hundreds of meters per day.

From earlier laboratory research at IAEA-MEL, it was suspected at the time of the accident that biological activity in surface waters might be responsible for absorbing radionuclides like a sponge and removing them to depth. Primary production of minute plant-like phytoplankton cells creates solid surfaces onto which contaminants like radionuclides are adsorbed. The zooplankton which feed on these radioactive cells subsequently defecate large aggregates (fecal pellets) which can further scavenge radioactivity as they sink.

Therefore, to test this hypothesis, on 6 May 1986 live zooplankton were netted from the waters over the sediment trap and allowed to defecate in special aquaria on board ship. When analyzed, these freshly produced fecal pellets were found to contain radionuclides with similar concentrations and relative distributions as were present in the sinking particles trapped at 200 meters. Microscopic examination of the trap samples confirmed that they were rich (70% by weight) in the same type of fecal pellets as the netted zooplankton had produced. Thus, this accidental "field experiment" was actually the first direct and convincing demonstration of the biological processes by which ocean waters are cleansed of contaminants like radionuclides.

Following the Chernobyl accident, the different radionuclides which entered the surrounding seas were removed to different degrees depending upon their chemical reactivities. For example, in the case of the particle-reactive radionuclides, cerium-141, cerium-144, and plutonium-239+240, from 50% to 75% of the total radionuclide inventories deposited in this region of the Mediterranean had transited through 200 meter

depth by one month after the accident when the sediment trap stopped sampling. In sharp contrast, only 0.2% of the corresponding caesium-137 deposition had passed through 200 meters by that time, an observation which is consistent with the generally non-reactive behaviour of this long-lived nuclide in seawater. For this reason, Chernobyl-derived caesium-137 has proved to be very useful as a water mass movement tracer in the Mediterranean and other seas for several years after the accident.

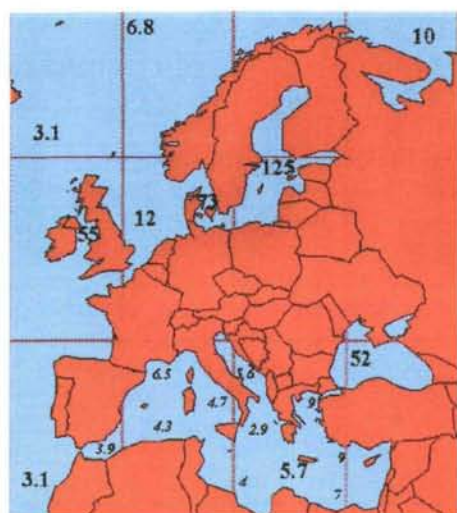
IAEA-MEL was not the only group of marine scientists deploying sediment traps in European waters following the Chernobyl accident; time-series traps were collecting particles at nearly the same time in the North Sea, the Black Sea, and Lake Zurich. Where comparisons could be made, concentrations of Chernobyl-derived radionuclides in the different sinking particles were surprisingly similar, and in most cases biological activity in the upper water column was considered to be the driving force in transporting the radioactivity downward. However, large differences in radionuclide flux were evident due to variations in particle mass flux which is normally site and depth specific. The extreme case was observed in Lake Zurich where roughly 20% of the fallout was removed from the water column in two months due to the sinking of a massive bloom of calcareous algae.

Viewed collectively, the temporal flux data for the Chernobyl radionuclides collected throughout Europe after the accident have proven extremely important for refining general models of contaminant removal and transport in aquatic systems.

Environmental & radiological aspects

Of more than 20 radionuclides which were released in significant quantities during the Chernobyl accident, only a few have been studied extensively in the marine environment. Among the most important have been strontium-90, caesium-134, caesium-137, and plutonium-239+240. Other radionuclides, such as iodine-131, have half-lives that are too short to be harmful or relevant to understanding marine processes, or had very low concentrations (for example, iodine-129).

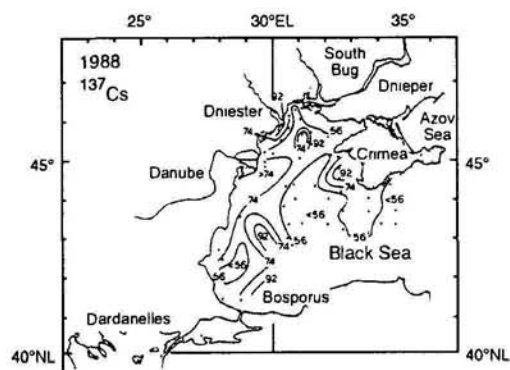
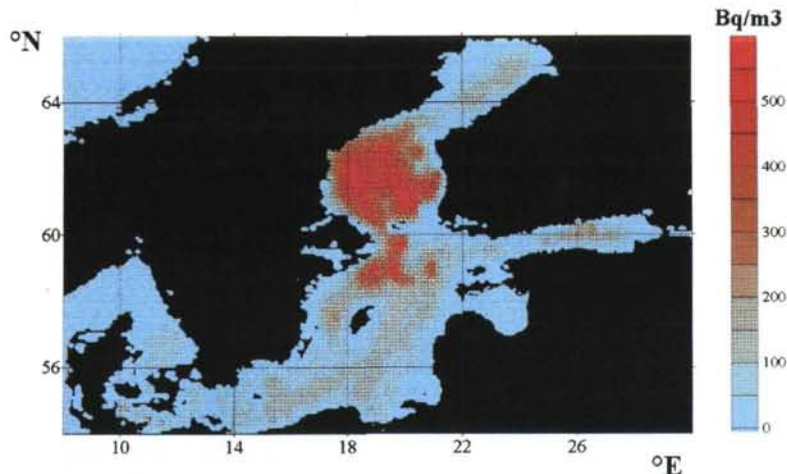
As mentioned previously, considerable differences in marine behaviour were observed. Strontium-90 and caesium-137 are typical representatives of elements which are soluble in seawater and can be used for studies of water dynamics. Their particle reactivity is very low in comparison to, for example, plutonium isotopes, which lie at the other extreme, in a group of



Above left: The average caesium-137 activity of surface seawater (Bq m^{-3}) estimated for the reference year 1990. The Mediterranean regional sea values were extracted from the Commission of the European Community's MARINA-MED report.

Above right: Caesium-137 contours in the surface seawater of the Baltic Sea for the period 1986-88 as extracted from the IAEA-MEL Global Marine Radioactivity Database. The data were mostly supplied by the Bundesamt für Seeschifffahrt und Hydrographie, Hamburg, Germany.

Right: Caesium-137 activity isolines (Bq m^{-3}) in surface waters of the western Black Sea as extracted from the measurements in 1988. Adapted from G. G. Polikarpov *et al.*, *Journal of Environmental Radioactivity* (Vol.13,1 (1991)).



elements having low solubility and high particle reactivity. Plutonium isotopes do not travel long distances from the source because they are deposited into sediment, which therefore contains their main inventory in the ocean.

As the plutonium input to the oceans following the Chernobyl accident was small and localized, this article will concentrate on discussing the impact of Chernobyl radiocaesium on the marine environment. The caesium isotopes were both the most widespread and most abundant of the ones released.

The behaviour of radiocaesium in the oceans has been studied over a long period with reference to its fallout from nuclear bomb tests and in discharges from nuclear reprocessing plants. In particular, discharges from Sellafield in the United Kingdom have been extensively used to study water and sediment dynamics in the Irish, North, and Norwegian Seas. The radiocaesium from weapons test fallout was exclusively caesium-137. Caesium-134 has, however, been present in Sellafield discharges as well as in Chernobyl debris. The ratios of caesium-134 to caesium-137 have been different, however. Caesium from Chernobyl was thus readily distinguishable from other sources by having a differ-

ent caesium-134/caesium-137 activity ratio of about 1:2.

Radiologically, the sea most affected by the Chernobyl accident was the Baltic, since the first radioactive clouds from Chernobyl travelled to the north and caused high deposition over the Scandinavian region. Atmospheric deposition played a dominant role in determining the radioactivity of this sea. The mean caesium-137 concentration in surface waters estimated for the reference year 1990 was highest in the Baltic Sea. (See maps.) Because of the closed nature of this sea and its small exchange of water with the North Sea, the levels of caesium-137 of this sea have remained the highest in Europe.

Caesium-137 contours shown on the map of the Baltic Sea for the period 1986-88 illustrate the enhancement of caesium-137 concentrations in seawater, with clear evidence of the effect of run-off from land, particularly from Sweden. The measured range in 1986 was from a few becquerel (Bq) to 2400 Bq per cubic meter, i.e. two to three orders of magnitude higher than in other European seas.

The next most perturbed sea following the Chernobyl accident was the Black Sea, where the mean caesium-137 concentration in seawater in

1990 was 52 Bq per cubic meter, comparable to that in the Irish Sea. The highest deposited activity was observed in 1986 in its northernmost area, about 500 Bq per cubic meter, i.e. 30 times higher than the pre-accident values. Strontium-90 activity measured in 1988 in surface waters of the western Black Sea was mostly between 10 and 50 Bq per cubic meter. Generally, a similar distribution was observed in 1988 for caesium-137 as well, but the levels were higher for this radionuclide by a factor of two. Strontium-90 and caesium-137 surface concentrations in Aegean Sea waters were much lower, between 5 and 11 Bq per cubic meter. The distribution patterns of strontium-90 and caesium-137 observed in the surface waters of the Black Sea can be explained in terms of two main source functions — namely by a short-term atmospheric deposition which dominated immediately after the accident, and then by a long-term transfer from the Kiev Reservoir and the catchment area of the Dnieper, Dniester, and Danube rivers.

For the Mediterranean Sea, the main Chernobyl contribution then arrived by exchange of waters with the Black Sea, which has essentially acted as a radioactive source. Atmospheric deposition and river inputs were estimated to have played minor roles. The mean caesium-137 concentration in surface water estimated for 1990 was 5.7 Bq per cubic meter. The caesium-137 levels in regional seas as estimated by the Commission of the European Community's MARINA-MED project ranged from 2.9 to 9 Bq per cubic meter, clearly showing a west-east trend towards the highest values in the Aegean Sea.

Technical assistance and training

The IAEA has been active in assisting Member States both to monitor and understand the effects of the Chernobyl accident on their marine systems. Firstly, in the Black Sea region where Chernobyl radioactivity is among the foremost public concerns in relation to health, the IAEA has initiated a major programme of technical co-operation. Activities are directed at strengthening the capabilities of the regional Member States to measure and monitor marine radioactivity, especially alpha-emitters, and involve careful evaluation and selection of equipment and training approaches.

In November 1994, a 2-week regional training course on marine radioactivity monitoring was held in Istanbul. In addition, a complementary Co-ordinated Research Programme (CRP) on isotopic tracers in the Black Sea has been carried out. It is being done with a view to using the Chernobyl pulse of radionuclides and related

nuclear techniques to understand water movement and element cycling in the heavily polluted marine system.

In the Baltic Sea, the Helsinki Commission MORS programme (Monitoring of Radioactive Substances) has received significant support from IAEA-MEL in terms of organization of specially designed analytical quality assurance exercises for marine radioactivity assay. This support will be further increased by the organization, in Finland, of an IAEA training course on marine radioactivity studies in September 1996. Again, the work will feature particular reference to the Chernobyl impact and to the needs of the new Baltic States (Estonia, Latvia, and Lithuania) which have joined the IAEA.

Overview of marine radioactivity

An overview of marine radioactivity perspectives has been provided by the IAEA's recently completed CRP, "Sources of Radioactivity in the Marine Environment and their Relative Contributions to Overall Dose Assessment from Marine Radioactivity (MARDOS)". This study provided new up-to-date estimates of doses to the public from anthropogenic caesium-137 (originating from global fallout, the Chernobyl accident, and authorized discharges) and from natural polonium-210 through consumption of marine food.

It included a study of fishing areas as defined by the United Nations Food and Agriculture Organization (FAO). Analysis of fishing area No.37 (the Mediterranean and Black Seas) has shown that the collective effective dose commitment for caesium-137 in marine food (fish and shellfish) in 1990 was 6 man sievert (Sv) — much smaller than the 700 man Sv derived from polonium-210 ingestion. The highest doses (86 man Sv) in world oceans due to caesium-137 were found in the North Atlantic area (FAO fishing area No. 27, which also includes the Irish, North, Baltic, Norwegian, and Barents Seas). However, they are still negligible in comparison with 2900 man Sv derived from polonium-210 ingestion.

Generally, it can be concluded that the Chernobyl accident has had a measurable impact on the marine environment. Radionuclide levels (mainly caesium-137) were two to three orders of magnitude higher than the pre-Chernobyl levels. However, the doses to the public from ingestion of caesium-137 in marine food have been estimated to be at least an order of magnitude lower than those due to natural polonium-210. □