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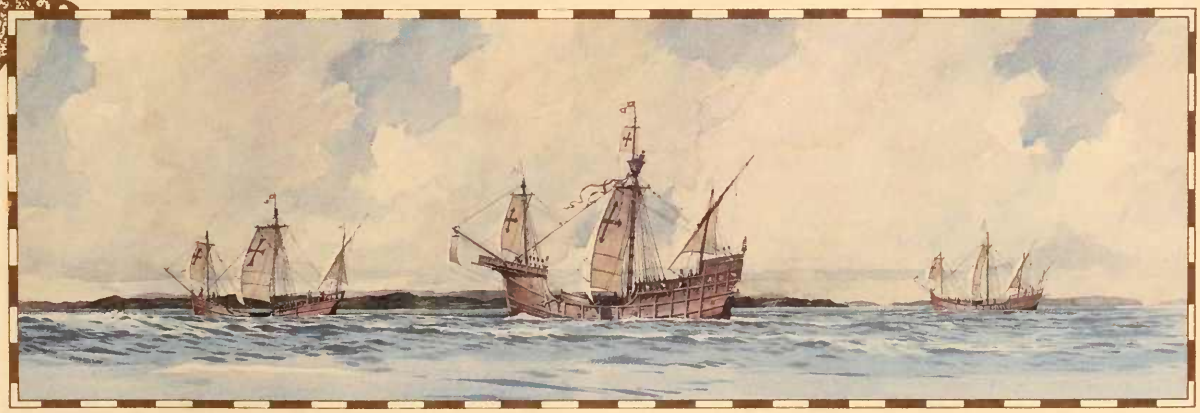
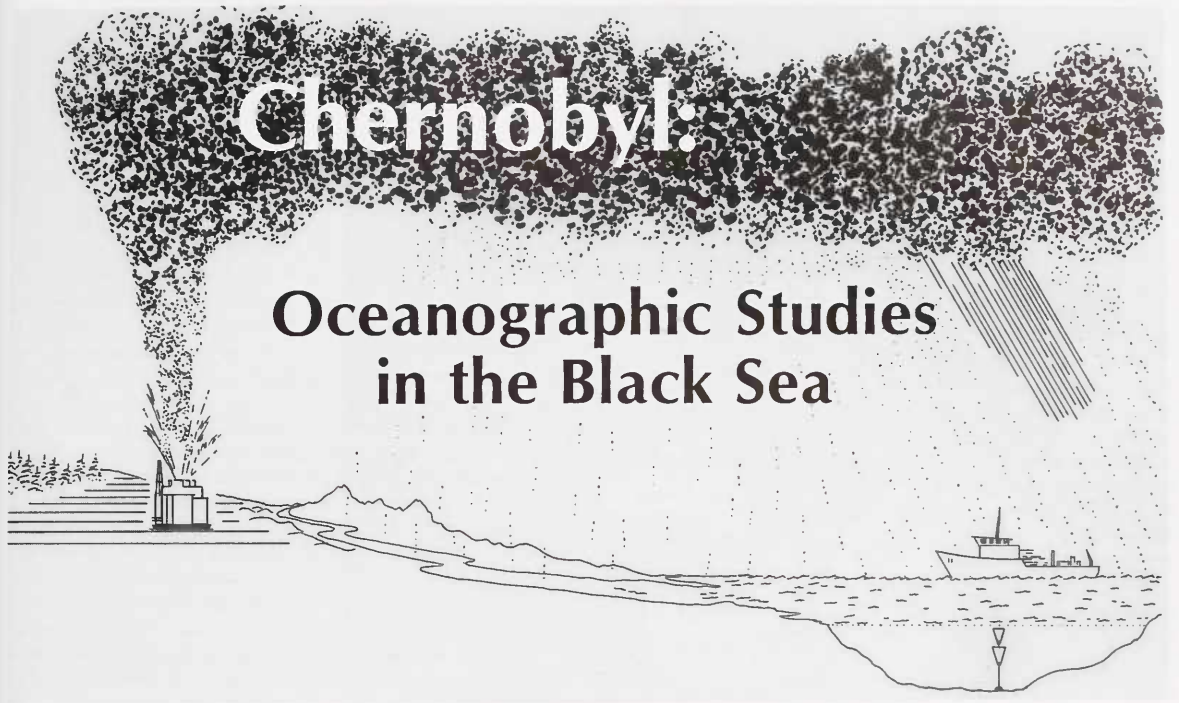
Columbus' Landfall Still a Mystery



New Calculations Point Again to San Salvador, Not Samana Cay



Chernobyl:

Oceanographic Studies in the Black Sea

—Studies on the radioactive fallout from Chernobyl provide information about the fates of nuclear discharges to the environment, and about the circulation of the nearest body of salt water—the Black Sea.

by Ken O. Buesseler

During the early morning hours of April 26, 1986, the Unit 4 reactor at the Chernobyl nuclear power station in the Soviet Union became the source of the largest release of radioactive material to the atmosphere from an industrial accident. Now, more than a year later, many of the questions concerning how the accident occurred, and the nature of some of the immediate and long-term environmental consequences can be answered.

For marine scientists, the Chernobyl accident resulted in the deposition to the oceans of a unique mixture of fallout radionuclides.* These radionuclides can be used to trace a variety of geochemical processes, such as water circulation and ventilation** processes, and the chemical scavenging process† in surface waters.

The Black Sea is the closest body of salt water to the reactor site (Figure 1). It received considerable direct atmospheric fallout from Chernobyl. Additional Chernobyl radionuclides were and are being carried to the Black Sea by the Danube and Dnepr rivers—their drainage basins include many of the high fallout areas of Eastern Europe and the accident site itself.

Aside from its proximity to the accident site, the Black Sea has several unique characteristics. For example, its deep waters are completely devoid of oxygen, and as such, the geochemical processes characteristic of this basin are quite unusual, and not well understood. The research team now studying the Black Sea includes the author and colleagues from the Woods Hole Oceanographic Institution (WHOI), along with West German and Turkish scientists.

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* Radionuclides—for the atoms of a given element, there may be several forms. The atoms have the same atomic number (the same chemical element), but have different atomic weights (different characteristics). These various forms are termed isotopes. A radionuclide is an unstable, radioactivity-emitting isotope.

** Ventilation—the transport of oxygen to the deep ocean as surface water sinks. In the absence of such a process, as in the Black Sea, the deep water becomes anoxic.

† Chemical scavenging—the transport of elements to the deep waters, with subsequent removal from the surface layers, through attachment to sinking particles.

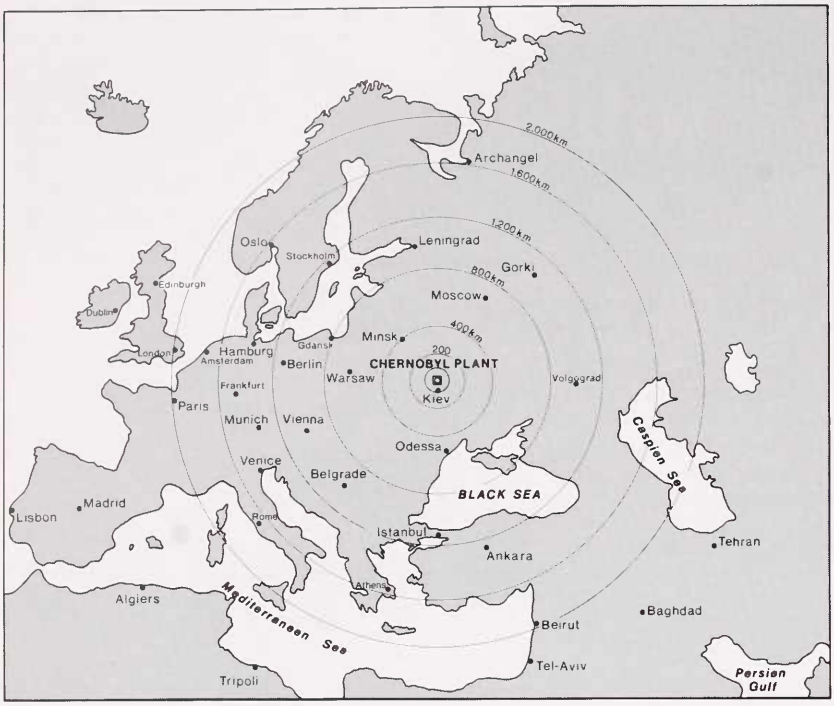

Figure 1. Chernobyl, with major cities and bodies of water.

The Accident

There now appears to be little disagreement that the Chernobyl accident was caused by a series of operator errors that occurred during the 24-hour period prior to the accident. Electrical engineers (not nuclear reactor specialists) at the site were given approval to conduct a test of the turbogenerators.

The Chernobyl test needed to be conducted at low reactor power, and a variety of automatic and emergency controls were disabled to allow the reactor to run at far below its regular operating power. At this low power level, the Chernobyl-type reactor becomes difficult to maintain at constant energy levels, and there is the possibility of rapid power excursions.

The operators mistakenly reduced reactor power too low during their tests, when, in order to increase reactor power, in the early hours of April 26 they attempted to rectify their error by removing most of the control and safety rods from the reactor core. As the result of this and other oversights, when the power began to increase, it did so with a tremendous surge.

Within 4 seconds, the reactor power peaked at approximately 100 times its nominal power rating, resulting in an explosive release of energy equivalent to many tons of TNT. This energy caused some of the fuel rods to rupture, allowing hot fuel particles to contact the cooling water directly, resulting in a massive steam-explosion. The reactor containment structure was breached, and large pieces of flaming debris were ejected from the destroyed reactor building. Exposed to air, the reactor's graphite core caught fire, further vaporizing the more volatile radionuclides in the core, and providing the major pathway for uncontrolled radionuclide releases.

Emergency measures were undertaken, including a heroic effort to contain the fires to Unit 4. Starting on April 28, more than 5,000 tons of boron, dolomite, sand, clay, and lead were dropped on the site from helicopters to quell the graphite fire in the reactor core, and to adsorb and filter some of the aerosol* particles. The aerial measures were successful in slowing down the fire and radionuclide releases, but, despite these efforts, the reactor burned out of control for 9 days until May 6.

Somewhat unexpectedly, the heated smoke plume from the fire carried the largest amounts of radioactivity up to higher altitudes, where it was carried away from the accident site by the prevailing winds. Thus, in the vicinity of the accident, radioactive fallout deposition was much lower than earlier models had predicted. Radioactivity levels at sites further away, however, were surprisingly high. Even with this widespread dispersion of fallout radioactivity, more than 100,000 people living in the surrounding areas were evacuated by bus. This evacuation started on April 27, some 36 hours after the explosion. Because of the extensive radiation contamination of the soils and waters around the vicinity of the reactor, these residents may never return to their homes.

Radionuclide Releases

A fact unrecognized by many is the complexity and variability in both the composition and quantity of this type of radioactive release. This was indeed true

* Aerosol—the suspension of fine solid or liquid particles in a gas (smoke, fog, mist).

for the Chernobyl accident. By the end of the accident, the radioactivity releases totaled more than 50 million curies*, or 3 to 5 percent of the total radionuclide inventory in the reactor core. This release of aerosol particles and gases was made up of more than 30 different radionuclides characteristic of reactor operations, many of which are potentially hazardous to man.

It was both the quantity and the characteristic composition of the fallout radionuclides that alerted the Swedes on April 28, some 2 days after the fire began, that a nuclear reactor accident had occurred. It is these same characteristics that enable oceanographers to use the fallout from this type of incident as a "label" or "tag" for ocean dynamics and circulation studies.

The major pathway for release of radioactivity was through volatilization during the fire. In general, those radionuclides with lower boiling points, such as the isotopes of radioactive iodine (iodine-131) and cesium (cesium-137 and cesium-134), escaped preferentially from the reactor core. Since the intensity and temperature of the fire varied considerably from day to day, the relative ratios among the different radionuclides released also varied. This has important implications for environmental studies, since many areas received a different mix of fallout radionuclides, depending on when the particular source cloud for that region was emitted.

A second mechanism for radionuclide release was through mechanical processes, such as during the initial explosion when many small particles were generated from the reactor core material. Many of these tiny, micron-sized particles (a millionth of a meter in diameter) were carried, along with the gaseous releases, by the smoke plume into the environment.

One way to think of this complex release of radioactivity is by analogy to what I call "Chernobyl soup." If each radionuclide were an ingredient in a soup, every day the amount of that ingredient in the recipe would be changing. In this instance, volatility and mechanical release mechanisms were continually changing the flavor of the "soup" (Figure 2).

Additionally, the composition of the Chernobyl fallout has changed quite rapidly since its release as the result of radioactive decay.** Using the soup analogy, as soup ages, it tastes differently from day to day; radioactive decay similarly produces striking differences over time in the relative abundances of the Chernobyl radionuclides in the fallout. For example, of major immediate health concern were releases of iodine-131, and cesium-137. Iodine-131 is a highly toxic substance that is easily volatilized. Iodine-131 has an 8-day half-life,† so that after one month, the original iodine-131 activity had decreased, as a result of radioactive decay, by more than a factor of 10. Today, more than a year later, essentially no iodine-131 remains. The radionuclide cesium-137, on the other hand, is also easily volatilized, and of similar concern. Because it has a much longer 30-year half-life, most of the Chernobyl cesium-137 remains with us. Even after 100 years, 10 percent of the cesium-137

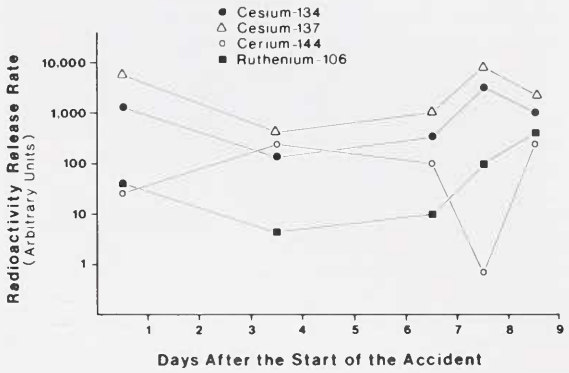

Figure 2. Isotopic release rates for Chernobyl releases versus the day of release. Note that the mixture of escaping radionuclides changed dramatically from day to day.

released by Chernobyl accident will still be present in the environment. Of the major Chernobyl radionuclides of concern, their half-lives range from a few days to many thousands of years.

Two other variables in determining the final impact of Chernobyl at any given site were the winds that carried the radioactive debris, and the rains that were primarily responsible for depositing the fallout on land. Since the radioactive fallout was carried by prevailing winds, variability in the wind patterns resulted in the eventual spread of Chernobyl radioactivity in a variety of directions. Although the fallout plume was initially carried north towards Scandanavia, by April 30, the winds had completely shifted towards the south (Figure 3). In fact, the fallout originating at the accident site after May 1 is the most important component of the Chernobyl releases for our Black Sea studies.

Within any given fallout plume, the actual deposition of Chernobyl aerosol particles and gases occurred predominantly during rain storms, which are often highly localized in nature. Thus, even within a distance of a few 10s of kilometers, the relative levels of radioactivity at ground level varied by a factor of 10 to 100, depending on local wind and rain patterns. Environmental scientists are finding that, with limited sampling, this patchiness in radionuclide deposition levels and radionuclide composition makes it very difficult to predict fallout levels and characteristics over large areas.

To put the Chernobyl releases in perspective, some 10 percent of the total inventory of the volatile cesium-137 in the reactor core escaped. This represents some 1 million curies of cesium-137. This total is significantly smaller than the 36 million curies

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* Curie—a relatively large unit describing the rate of release of radioactivity, defined as 37 billion disintegrations per second.

** Radioactive decay—as unstable atoms emit energy or particles, they lose radioactivity in the transformation to different, more stable, elements.

† Half-life—the rate at which unstable atoms disintegrate or decay, expressed in the time required for half of the atoms to decay to another form. This may range from seconds to billions of years, depending on the isotope.

of cesium-137 released during all of the atmospheric nuclear weapons-testing programs in the 1950s and 1960s. However, in many places in Europe, the cesium-137 inventories from Chernobyl are greater than the preexisting weapons-testing inventories, because of the patchiness in the Chernobyl deposition pattern discussed earlier. Also for comparison, the total release of cesium-137 from the Three-Mile Island incident in the United States in 1979 was only 10 curies. In the Black Sea, we estimate that the Chernobyl cesium-137 fallout input was roughly twice as large as the cesium-137 inventory deposited to this basin from weapons testing fallout.

Black Sea Studies

In general, we, along with many other scientists, are learning about the fates of radioactive discharge to the environment. In particular, we have focused our attention on the Black Sea.

The Black Sea (Figure 4) is quite an interesting basin for chemical oceanographers because of a variety of unusual features. The surface waters are brackish (a salinity of 16 to 18 parts per thousand) because of the strong influence of the major river discharges into the basin. The surface waters also are biologically very productive. In sharp contrast, the Black Sea deep waters are completely devoid of oxygen, and hence most life forms are absent at depth. These deeper waters are more saline (21 to 23 parts per thousand) than the surface waters, and represent about 90 percent of the basin by volume. Being saltier, the deep waters are significantly denser than the surface layer, and hence a strong density gradient occurs somewhere between 50 to 200 meters, depending on the location and season.

In essence, the surface waters are "floating" on top of the deeper waters like "oil and vinegar." Any mixing between the layers is severely limited. It is this lack of mixing that produces the depletion of oxygen at depth, occurring as the organic matter produced in the surface waters sinks and decomposes in the deep waters. A limited source of fresh oxygenated (and highly saline) water to the deep Black Sea is an undercurrent of Mediterranean water that flows into the Black Sea through the narrow and shallow Bosporus Straits. The total water balance is maintained by the surface outflow of less saline surface waters, and by evaporation in the basin.

Because of this unique set of conditions, the Black Sea can be used as a natural laboratory to study a variety of geochemical processes. For instance, in the Black Sea, we can examine to what extent any downward mixing of surface waters occurs, despite the strong density gradient between the surface and deep layers. Many of the so-called particle-reactive elements, attached to sinking particles, are likely to cross this oxic/anoxic

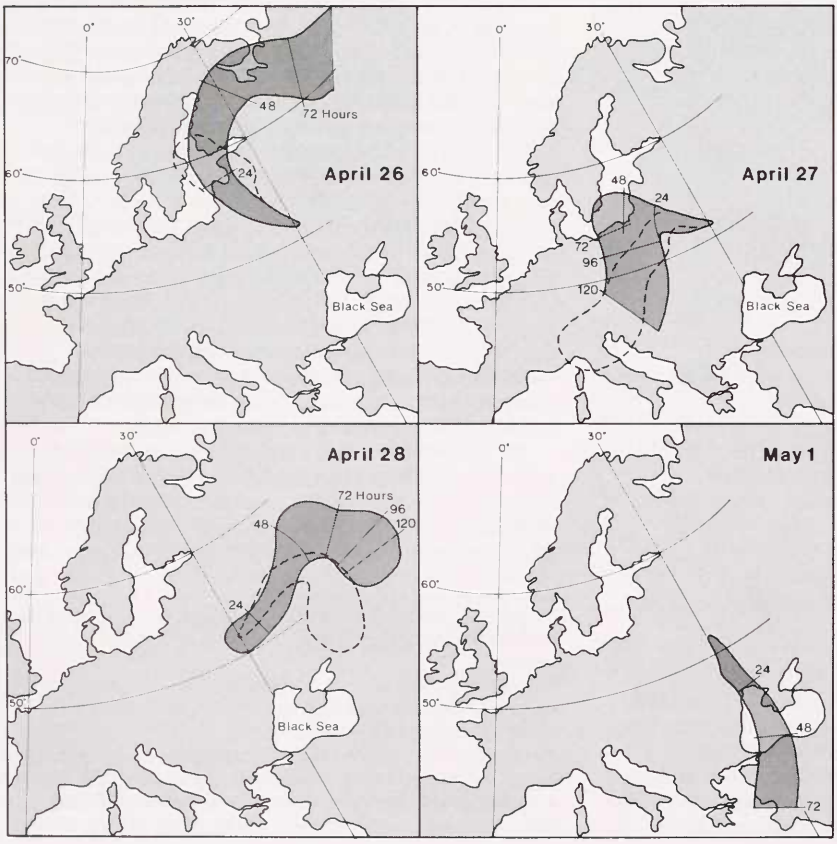


Figure 3. The atmospheric pathway of radioactive fallout from Chernobyl at 750 meters (dashed line) and 1,500 meters (shaded plume) originating from the accident site on different days.

boundary. Also, at the boundary between the oxygenated surface waters and deep anoxic region, a variety of chemical transformations are thought to occur.

Given the known Chernobyl fallout releases, it was important to sample these waters as soon as possible after the accident. Our plan was to determine the fallout levels and range of radionuclides present, and then to use these Chernobyl radionuclides as tracers of water or particle-related processes. In general, this work is similar in nature to studies that use fallout from the atmospheric nuclear weapons testing programs in the 1950s and 1960s to trace oceanic processes. Of course, many of the short-lived radionuclides from weapons testing fallout have decayed away, while the more recent Chernobyl fallout still included the shorter-lived radionuclides.

With rapid planning and cooperation from our European colleagues, we were able to participate in two cruises aboard the Turkish research vessel, *K. Piri Reis*, to the Black Sea in June and September of 1986. The cruises involved international participation, including German scientists from the University of Hamburg, Turkish scientists and crew from the University of Izmir, and WHOI chemists and geologists. Overcoming Turkish, German, and English language barriers, we were successful in obtaining the first set of samples from the Black Sea surface waters on June 17, just under 2 months after the Chernobyl releases.

Back at WHOI, it was only a matter of minutes before the samples revealed an indication of the levels of Chernobyl fallout. Using highly sensitive radiation detection techniques, cesium-134 and cesium-137 signals from Chernobyl were detected in the surface water samples. Within 24 hours, 8 additional Chernobyl radionuclides were identified:

K. Piri Reis, *the Turkish research vessel used for Black Sea studies.*

cerium-144, cerium-141, ruthenium-106, ruthenium-103, lanthanum-140, barium-140, niobium-95, and tellurium-129. With further radiochemical analyses, the isotopes of plutonium, americium, and curium were also detected.

While these radionuclide activities were not high relative to radiation exposure safety guidelines, they were certainly elevated compared to any preexisting activities. For example, the cesium-137 activity in the Black Sea surface waters was found to be elevated over its previous activity levels by a factor of 10 to 20. The Chernobyl signal appeared across the entire southern Black Sea (our sampling efforts were confined to Turkish territorial waters). Some of the patchiness seen in the fallout levels on land appeared in the surface water data as well.

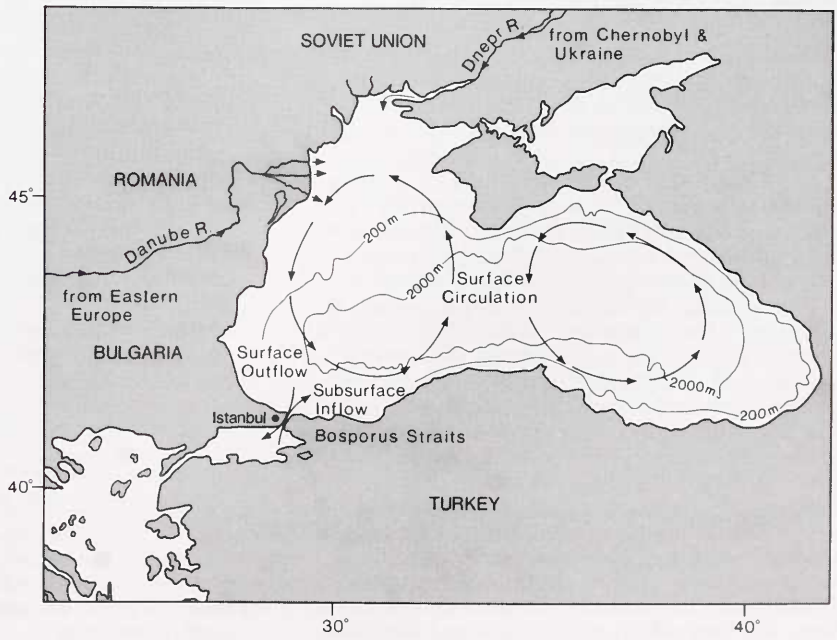


Figure 4. Circulation of the Black Sea, with river inputs.

Cleaning the streets of Kiev, the nearest large city to Chernobyl, on June 3, 1986, shortly after the accident. (Sygma Photo News)

Results to Date

Based on their chemistries, the Chernobyl fallout radionuclides can be broadly grouped into two categories. The highly soluble elements, such as the cesium isotopes, are used to trace water movement and mixing processes. The more particle-reactive elements, such as cerium, ruthenium, plutonium, americium, and curium, are used to trace the rapid scavenging and removal of an element from the surface waters—due to their strong affinity for particle surfaces, and their potential rapid transport to depth.

Cesium-137 is a good example of a highly soluble radionuclide whose marine chemistry has been well characterized—because of its relatively long half-life and previous studies on this major component of atmospheric nuclear weapons testing fallout. Because the Chernobyl fallout contains in abundance the shorter lived cesium-134 isotope in addition to the cesium-137 (about half as much cesium-134), it is possible to easily separate the pre- and post-Chernobyl cesium signals. We found in the Black Sea that surface water cesium-137 levels had jumped from 15 to 340 becquerels* per cubic meter in the surface waters near the mouth of the Bosporus. By September 1986, the Chernobyl cesium signal had penetrated down to the base of the summer thermocline,** and with future samplings, we should be able to see the progression of this signal deeper into the water column. With time, this Chernobyl cesium signal may penetrate into the deeper waters, if some limited mixing exists between the surface and deeper layers. It is precisely this progression of the cesium signal over time that will quantify the rate at which the surface waters are being mixed to depth.

For several other elements, movement through the water column may be somewhat more complex. While cesium-134 and cesium-137 primarily trace water movement, because of their chemistries, many of the Chernobyl radionuclides are considered highly particle reactive. Such particle-reactive tracers are rather "sticky," and their oceanic behavior is often determined by the particles to which they are attached. Therefore, the fate of these particle-reactive tracers is determined not only by the water mixing processes, but also by their partitioning onto the large and small particles in the water column. The removal of particle-reactive elements to the deep waters and underlying sediment is termed "scavenging." The rate of scavenging of particle-reactive elements is a topic of major concern in oceanographic studies, because the balance between the supply of an element to the ocean, and its ultimate removal from the ocean is largely determined by this scavenging removal rate. The scavenging rates of Chernobyl radionuclides also will be of value to local authorities interested in how long the radioactivity will remain in the Black Sea surface waters.

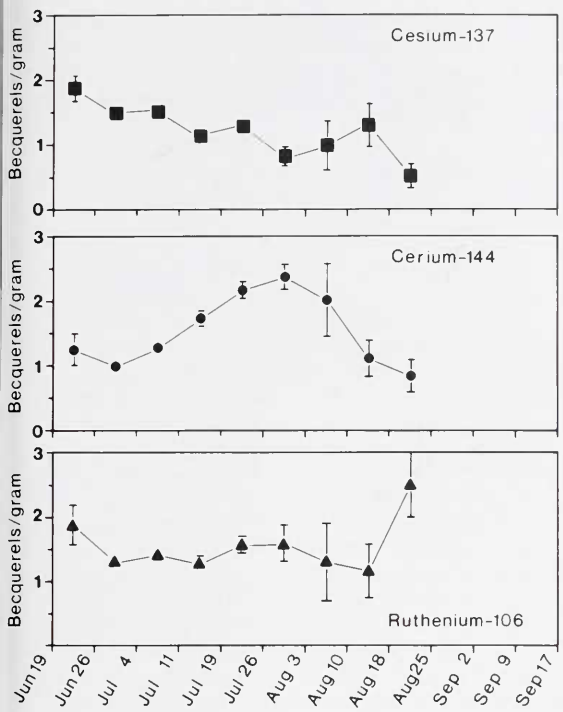
Recently, much attention has been focused on large biogenic (produced by living organisms) particle aggregates, which are formed in surface waters, and their role in transporting a variety of particle-reactive elements to depth. A variety of organisms, either through their feeding or defecation processes, act to aggregate smaller particles into larger ones. This increases the rate of particle settling, perhaps up to hundreds of meters per day. As such, these sinking particles can form the major removal pathway for a variety of elements from the surface oceans.

A great deal of data on particle activity in the oceans have been collected from sediment traps deployed by Susumu Honjo, Senior Scientist in WHOI's Geology and Geophysics Department. One sediment trap (Figure 5) was moored at 1,071 meters in the southwestern region of the Black Sea from June to September, 1986. This has provided a set of preliminary data on Chernobyl radionuclides.

Sediment traps are essentially large, cone-shaped funnels that collect settling particles as they fall through the water column. With an electronic switching device, it is possible to obtain a time series of sediment trap samples from a single trap deployment, by rotating new sample collection cups into position at the base of the funnel. In this case, a

* Becquerel—an international unit of radioactivity equal to 1 nuclear disintegration per second. A curie equals 37 billion becquerels.

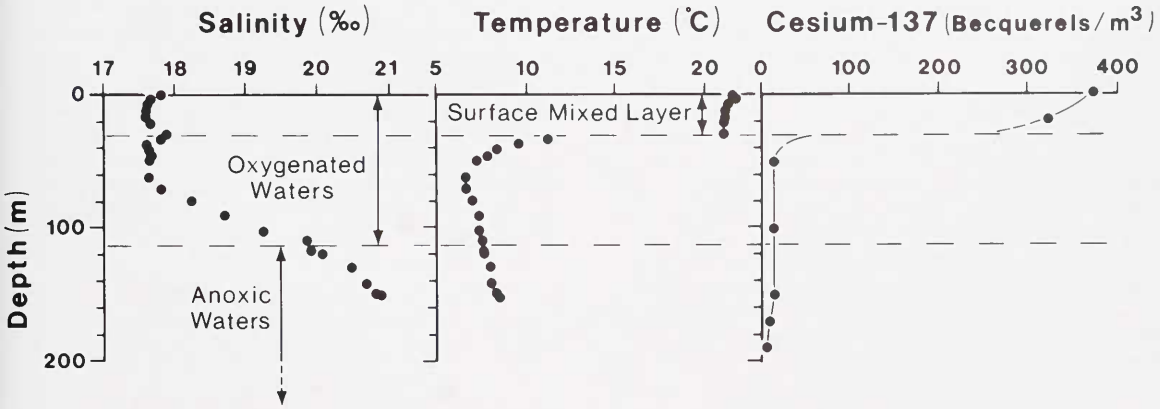
** Thermocline—A zone where the water temperature decreases more rapidly than the water above or below it. When the gradient is steep, mixing of the surface and deep waters is impeded.



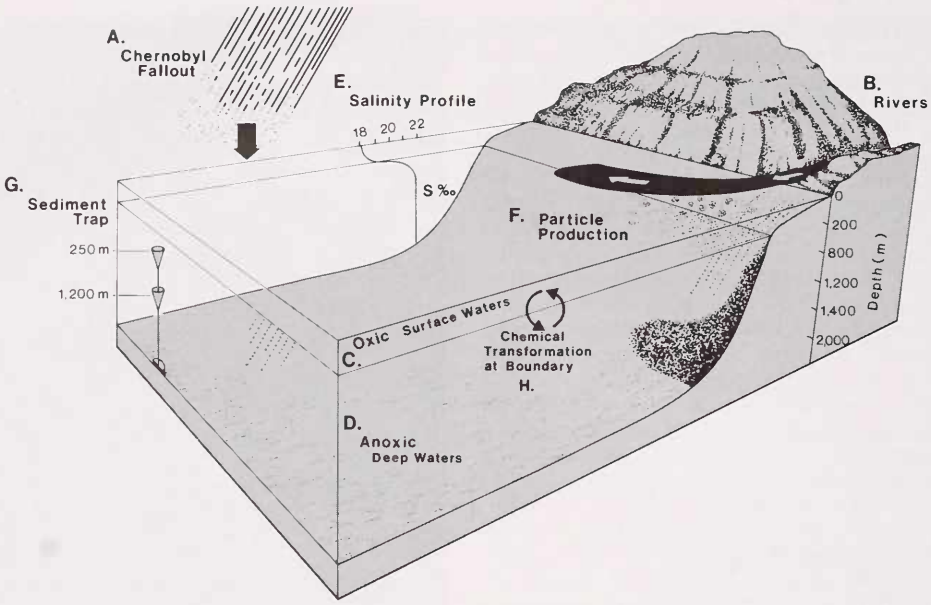
Collection of Chernobyl radionuclides by Black Sea sediment trap. The dates correspond to sediment trap sampling intervals. The error bars on several data points are statistical uncertainties relating to radioactive counting techniques. Note that the cesium-137 activity is decreasing, ruthenium-106 is fairly constant, and the cerium-144 appears to peak around the end of July. The reason for the cerium-144 peak is presently unknown.

Figure 5. The Black Sea sediment trap on the stern of the research vessel.

weekly time interval was used. Researchers have shown that, on a seasonal basis, the large particle fluxes can vary quite substantially. This variability in particle flux is often due to local productivity levels in the overlying waters, but in the Black Sea the fluxes also vary due to physical processes that transport particles from the shelf regions into the interior of the Black Sea.



The September 1986 cesium-137 activity profile (right-hand graph) from a site north of the Bosporus Strait as compared to the depths of the surface mixed layer (middle graph) and the salinity gradient (left-hand graph). Note that the very high surface-water cesium-137 signal, which had been labelled by Chernobyl fallout, had only penetrated to about 35 meters, or the depth of the summer thermocline. Cesium-137 activities below this depth represent background cesium-137 remaining from nuclear weapons testing fallout. The progression of this surface water cesium-137 with time will be of major interest. (From Livingston and others, 1987)



Black Sea Processes

Key:

A. *Atmospheric deposition of Chernobyl fallout to the Black Sea was the major initial source of new radionuclides to this basin.*

B. *The Danube and Dnepr rivers will become important for the delivery of dissolved and particulate Chernobyl radionuclides from run-off in their watersheds. These rivers also help maintain the low salinity in the surface waters.*

C. *Surface waters. Because of density differences, they are almost completely sealed off from the deep waters. While the surface waters are highly productive, the deep waters (D) are devoid of most life.*

E. *Because of the strong salinity gradient, the surface oxygenated waters remain isolated from the anoxic deep waters.*

F. *Large particles produced in the surface waters can sink rapidly to depth at speeds of 50 to 100 meters per day. We sample this settling particulate matter with sediment traps (G), which are moored at fixed depths in the water column.*

H. *The boundary between the oxygenated surface waters and the anoxic deep waters is a site for chemical transformations of some elements. For instance, the geochemistry of many trace elements can be directly or indirectly tied to chemical transitions at this boundary, which can strongly affect the scavenging (attachment to sinking particles) rate of the element, and its transport across this boundary.*

Our data show that at least some of the same Chernobyl radionuclides we had detected in the surface waters had been very rapidly removed to the trap at 1,071 meters—within less than 2 months of the initial fallout deposition at the surface. As expected by their differing chemistries, the removal rates for a more soluble element like cesium are much slower than that of the more particle-reactive cerium-144 and ruthenium-106. If the fluxes we measured continue (we have in place a program to monitor these trap samples for the next two years), the Chernobyl cesium-137 signal would take 100 to 200 years to be scavenged from the surface waters, while the cerium-144 and ruthenium-106 signals will be removed in only a matter of years.

Long-term Studies

In the future, the Chernobyl signal will be used as a tool to study the geochemistry of the Black Sea. With the spring run-off in 1987, the input of Chernobyl radionuclides from the rivers should be seen. We hope to use this riverine signal to trace surface circulation rates as the river waters travel from the Northern Black Sea region to the south, and to some extent out of the Black Sea, and through the Bosporus Straits.

Studies of Chernobyl fallout at other oceanic sites are underway, and comparisons between the behavior of the Chernobyl radionuclides in different settings will be possible. Future Chernobyls must be avoided, and cannot be excused for any reason, but we must act responsibly to learn as much as we can from this tragic accident.

Ken O. Buesseler is a Visiting Investigator in the Chemistry Department, Woods Hole Oceanographic Institution.

Acknowledgment

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