mass spectrometer and reported as $\delta^{13}C$ (*9*) with an analytical precision of approximately ± 0.2 per mil).

The $\delta^{13}C$ values of eight subsamples taken from the hydrothermal vent mussel tissues range from −32.7 to −33.6 per mil, with an average value of −33.2 per mil (Fig. 1). These values are strikingly lower than a compiled range of $\delta^{13}C$ from a wide variety of littoral, pelagic, and bathypelagic organisms from temperate oceans and also lower than values reported for major reservoirs of organic carbon in the marine environment including dissolved, particulate, and sedimentary organic matter (Fig. 1). The $\delta^{13}C$ values of the two *Mytilus* species from different littoral environments are within the range for marine organisms utilizing photosynthetically derived organic carbon (Fig. 1) and give no evidence that mytilid mussels fractionate carbon isotopes to an unusual extent. The lower relative abundance of ^{13}C in the tissues of the hydrothermal vent mussel by comparison with other marine organic carbon is thus evidence that the primary food source of this organism is not photosynthetically derived. This result supports suggestions by other investigators (*1*) that chemoautotrophic bacteria may be an important food source for the hydrothermal vent communities of the Galápagos Rift zone.

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References and Notes

1. R. D. Ballard, *Oceanus* **20**, 35 (1977); J. B. Corliss and R. D. Ballard, *Natl. Geogr.* **152**, 441 (1977).
2. P. Lonsdale, *Deep-Sea Res.* **24**, 857 (1977).
3. E. T. Degens, in *Organic Geochemistry*, G. Eglinton and M. T. J. Murphy, Eds. (Springer-Verlag, New York, 1969), pp. 304–329.
4. P. M. Williams and L. I. Gordon, *Deep-Sea Res.* **17**, 19 (1970).
5. E. T. Degens, M. Behrendt, B. Gotthardt, E. Reppmann, *ibid.* **15**, 11 (1968).
6. M. J. DeNiro and S. Epstein, *Geochim. Cosmochim. Acta* **42**, 495 (1978); B. Fry, A. Joern, P. L. Parker, *Ecology* **59**, 498 (1978); E. B. Haines and C. L. Montague, *ibid.*, in press.
7. D. L. Buchanan and B. J. Corcoran, *Anal. Chem.* **31**, 1635 (1959).
8. G. Rau, *Science* **201**, 901 (1978).
9. $\delta^{13}C = \{[(^{13}C/^{12}C)_{sample}/(^{13}C/^{12}C)_{PDB}] - 1\} \times 10^3$ per mil, where PDB is the Peedee belemnite standard.
10. We thank Drs. J. Corliss and J. Baross (Oregon State University) for providing the hydrothermal vent mussel and R. C. Clark for furnishing samples of *Mytilus* from the coast of France and the State of Washington. We also thank Dr. M. Stuiver for allowing us to use his isotope laboratory facilities at the University of Washington. Address correspondence to J.I.H.

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Seasonal Cycling of Cesium-137 in a Reservoir

Abstract. *Studies of a reservoir in the southeastern United States show that cesium-137, introduced into the system from a leak in a nuclear fuel element, cycles between the water and sediment on a seasonal basis. The cycling, which coincides with the annual periods of thermal stratification in this monomictic lake, has been occurring for over 10 years.*

Considerable research effort over the past several years has been devoted to determining the environmental chemistry of ^{137}Cs. Studies have shown that ^{137}Cs introduced into a watershed is attached to soil particles, which are removed by erosion and runoff (*1*). Some of the eroded soil particles comprise the sediments of the catchment basins in the watersheds and act as "sinks" for ^{137}Cs (*2*). Because of the high distribution coefficients of ^{137}Cs even in areas of differing soil-sediment type (*3*) and the almost irreversible fixation of this element in clay interlattice sites in freshwater environments, it is unlikely that this nuclide will be removed from these sediments under normal environmental conditions except by exposure to solutions of high ionic strength such as estuarine environments (*4*).

Thus far, all the studies of ^{137}Cs interaction with solids have been conducted either with tracer or fallout ^{137}Cs except for that of Simpson *et al.* (*5*) who dealt with low-level reactor releases in an estuarine system. The results of these studies have been extrapolated to predict the environmental fate of ^{137}Cs derived from the operation of nuclear reactors in freshwater systems. However, the release of low concentrations of ^{137}Cs from an experimental reactor fuel element at the Savannah River Plant, South Carolina, in 1957 has made it possible for us to study the environmental chemistry of this material during the intervening 12-year period and to compare it to that reported for ^{137}Cs from other sources.

Lower Three Runs Creek, on the site of the Savannah River Plant (Barnwell County) was dammed in 1958 to provide cooling water to two production reactors (P and R). The impoundment was completed in 1960 to form Par Pond (Fig. 1), an 1100-ha reservoir with a maximum depth of 16 m and a capacity of 6.8 × 10^{10} liters. Chemical and physical measurements of the system have been compiled regularly since 1965 (*6*). The pond is thermally stratified from April through October and well mixed from November through March. During thermal stratification, the hypolimnion becomes intensely anoxic with ferrous iron being dissolved from the sediment and in evidence throughout the hypolimnion. Concentrations of dissolved iron range from 0.02 part per million (ppm) in the epilimnion in the winter to more than 10 ppm in the hypolimnion during the height of stratification. The flux of iron from the sediments is so great during this stratification period that the concentration of dissolved iron in the surface waters increases about fourfold (*7*).

The R reactor area discharged small quantities of ^{137}Cs into the Par Pond system from 1954 until 1964 (*8*), due principally to the rupture of an experimental fuel element during testing in 1957. Initially the releases were to Lower Three Runs Creek until Par Pond was impounded in that portion of the watershed in 1958.

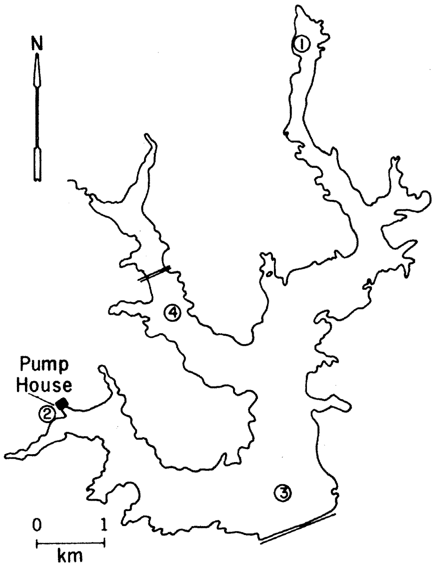
Studies of the ^{137}Cs concentration in Par Pond water have been carried on by three groups of researchers since 1960. The first studies were initiated by the Health Physics Environmental Monitoring Group at the Savannah River Plant. Cooling water pumped from Par Pond to the reactors has been monitored for ^{137}Cs. The pumping station's intake is located at a depth of 6 m, which is in the epilimnion of the pond. Water taken from the influent stream has been composited for a week to give a 1-liter total water sample which was analyzed for ^{137}Cs (*9*). The intake water has been monitored in that manner since 1960. In the second set of tests, from 1965 to 1968, J. S. Marshall sampled a station in the deepest area of the pond (Cold Dam station) on a monthly basis at depths of 1, 5, 10, and 15 m. These samples were analyzed for ^{137}Cs in cooperation with the Health Physics Group (*10*). Finally, during the period from March 1976 to February 1977 water samples were taken seasonally and with depth at the station occupied by Marshall in 1965 through 1968 and from two depths at a station in the bay of Par Pond (Hot Arm station) which currently receives thermal effluent (*11*).

The results of the monthly sampling from July 1965 through July 1968 demonstrate that ^{137}Cs concentrations in Par Pond water followed a seasonal cycle (Fig. 2). Elevated ^{137}Cs concentrations were observed at all depths from early summer until autumn. The concentrations drop to their lowest values during winter. This cycle mimics closely the cycle of thermal stratification observed in Par Pond (*6, 7*). In addition, the deep water samples have higher ^{137}Cs concentrations than the surface waters during the periods of anaerobiosis in the hypolimnion, whereas the surface and deep waters show only minor concentration differences during the periods of isothermal mixing in the pond. These concentration gradients suggest that ^{137}Cs is released to the water column from the sediments coincident with the dissolution of ferric oxides and then removed to the sediments when mixing introduces oxygen into the water column, hence causing reprecipitation of hydrous ferric oxides.

The ^{137}Cs concentrations in water samples taken between March 1976 and February 1977 (Fig. 3) confirm the observations that ^{137}Cs is cycling in the water column and that significant concentration gradients occur during the period of stratification. The similarity between the concentration profiles at the two stations indicates that the process is not an isolated phenomenon at the deep station but occurs over the entire pond. The difference in concentration between the two stations during August 1976 may be indicative of dilution of the surface waters in the Hot Arm by water from reactor operations and concentration of surface waters by evaporation during surface flow toward the Cold Dam.

In addition to the fact that ^{137}Cs continues to show a seasonal cycle in Par Pond over 10 years after having been introduced into the system, the overall concentration of ^{137}Cs in the pond water is lower during 1976 through 1977 than 1965 through 1968, however, apparently only by a factor of about 2 for surface waters. Examination of the yearly ^{137}Cs concentrations in surface water (Fig. 4), as determined by averaging the weekly collections at the Pump House station, shows the trend of surface water concentrations for the pond's history. The high concentrations for the years 1963 and 1964 (Fig. 4) agree with the releases of ^{137}Cs (83 and 47 Ci, respectively) for those years (*8*). In addition, the data support the surface water concentrations reported for 1965 through 1968 and 1976 through 1977 with their twofold concentration difference (*12*). Finally, the data show that most of the ^{137}Cs introduced into Par Pond rapidly left the water column (*13*). Within 3 years, the annual average surface water concentrations of ^{137}Cs returned to the values observed prior to the large inputs of 1963 through 1964 and have been maintained at those concentrations until the present. The slight decline in surface water concentrations beginning in 1971 through 1972 appears to be real. However, the cause is not understood at this time.

The seasonal cycling of ^{137}Cs observed in Par Pond apparently has been continuing over the past 12 years. However, the mechanism of the cycling remains unknown. The fact that the ^{137}Cs cycle closely follows the annual cycle of thermal stratification in the pond indicates that intense anoxic conditions may be an



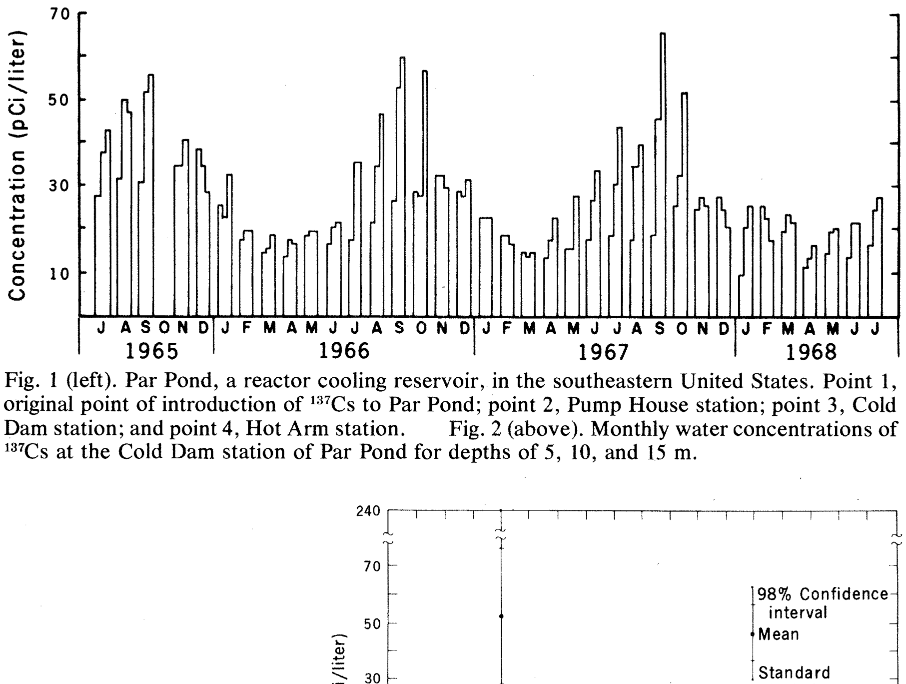


Fig. 1 (left). Par Pond, a reactor cooling reservoir, in the southeastern United States. Point 1, original point of introduction of ^{137}Cs to Par Pond; point 2, Pump House station; point 3, Cold Dam station; and point 4, Hot Arm station. Fig. 2 (above). Monthly water concentrations of ^{137}Cs at the Cold Dam station of Par Pond for depths of 5, 10, and 15 m.

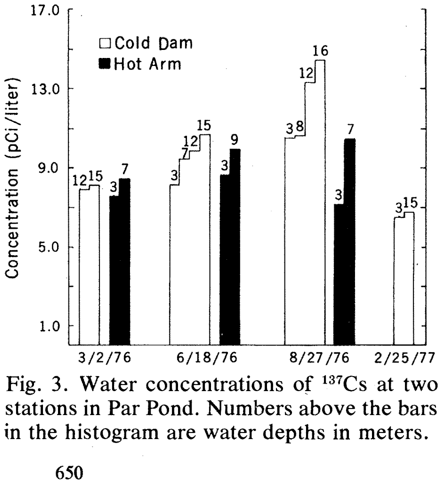
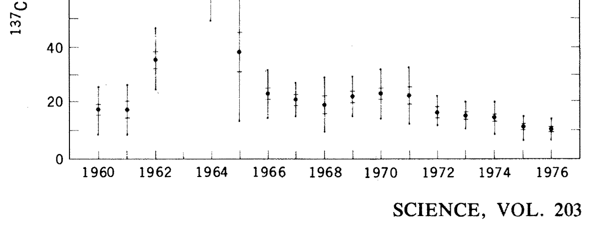


Fig. 3. Water concentrations of ^{137}Cs at two stations in Par Pond. Numbers above the bars in the histogram are water depths in meters.

Fig. 4. Annual average concentrations of ^{137}Cs in epilimnion water from weekly collections at Pump House station, Par Pond. Values were obtained by averaging the weekly ^{137}Cs concentrations over the calendar year.



important factor in its cycling. However, neither stable cesium in a montane lake which undergoes reduced oxygen concentrations (*14*) nor ^{137}Cs and stable cesium in a hypereutrophic lake which undergoes intensely anoxic, hypolimnetic conditions (*15*) show indications of a seasonal cycle. Cycling of ^{137}Cs coincident with thermal stratification has been reported in Lake Michigan (*16*). However, this cycling is apparently controlled by particulate transport and resolution in the water column well above the sediments. Hypolimnetic water in Lake Michigan does not become anaerobic over the vast bulk of the lake area, and ^{137}Cs concentrations are constant throughout the year in the deeper waters. This cycling observed in Lake Michigan and the mechanism controlling it are quite different from the case of intense anaerobiosis encountered during thermal stratification in Par Pond. Moreover, the cesium source terms in all the examples except Par Pond were either tracer material or fallout and weathering products rather than a nuclear fuel element. Because of the apparent anomalous behavior of ^{137}Cs in Par Pond and its relevance to the nuclear fuel cycle, further studies are required to define the cause of the ^{137}Cs cycling and the availability of this material to the biota of the system.

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References and Notes

1. A. S. Rogowski and T. Tamura, *Radiat. Bot.* **10**, 35 (1970).
2. D. N. Edgington *et al.*, *Transuranium Nuclides in the Environment* (International Atomic Energy Agency, Vienna, 1976), p. 493; L. D. Eyman and N. R. Kevern, *Health Phys.* **28**, 549 (1975); J. C. Ritchie and J. R. McHenry, *ibid.* **32**, 101 (1977).
3. Approximately 10^5 for midwestern and southeastern environments. J. J. Alberts and R. N. Muller, *J. Environ. Qual.*, in press; K. Garder and O. Skulberg, *Int. J. Air Water Pollut.* **8**, 229 (1964); E. A. Jenne and J. S. Wahlberg, *U.S. Geol. Surv. Prof. Pap. 443-F* (1968), p. F1.
4. J. J. Alberts, M. A. Wahlgren, K. A. Orlandini, C. A. Durbahn, in preparation.
5. H. J. Simpson, C. R. Olsen, R. M. Trier, S. C. Williams, *Science* **194**, 179 (1976).
6. L. J. Tilly, in *Mineral Cycling in Southeastern Ecosystems*, F. G. Howell, J. B. Gentry, M. H. Smith, Eds. (Publ. UC-11, National Technical Information Service, Springfield, Va., 1975), p. 394.
7. J. S. Marshall and J. H. LeRoy, in *Radionuclides in Ecosystems*, D. J. Nelson, Ed. (Publ. CONF-710501, National Technical Information Service, Springfield, Va., 1971), p. 465.
8. Approximately 222 Ci of ^{137}Cs were released from R reactor to the Lower Three Runs–Par Pond area between 1954 and 1964 [C. Ashley and C. C. Zeigler, "Releases of radioactivity at the Savannah River Plant, 1954 through 1976" (ERDA Report DPSPU 75-25-1, E. I. du Pont de Nemours and Company, Savannah River Plant, Aiken, S.C., 1977), p. 143].
9. The total water samples were fumed with HNO_3, and the ^{137}Cs was precipitated with KF. The ^{137}Cs concentration was then determined on a gas-flow proportional counter.
10. Water samples (25 to 50 liters each) were collected by means of a 10-liter polyvinyl chloride Van Dorn sampler bottle fastened to the hydrographic sampling wire horizontally so that the sample would represent a narrow lens of water at the sampling depth. The water samples were centrifuged through an eight tube, KSB continuous-flow centrifuge (Sorvall, SS-3) at 200 ml/min and 27,000*g*. Known volumes of the centrifugate were passed through ion-exchange columns. To determine ^{137}Cs, the radioactivity of the resins was counted with a NaI well crystal (22.9 by 22.9 cm).
11. The 50-liter samples were collected by means of the specially adapted Van Dorn sampler, filtered through 0.45-μm membrane filters (Millipore), and spiked with HCl (final $p\text{H} \sim 2$) and 5 mg of stable cesium to retard adsorption losses on container walls. The sample was evaporated in glass to a final volume of 500 ml; the ^{137}Cs concentration was determined by gamma-counting of the entire sample on a low-background NaI detector (7.6 by 7.6 cm).
12. Par Pond is a surface discharge reservoir with an average thermocline depth of ~ 6 m. The minimum replacement time of the reservoir calculated from rainfall discharge data and Savannah River makeup water volume is 8 to 10 months. Since this time period is long compared to the time of stratification, it would be expected that most of the ^{137}Cs in the reservoir would remain in the system rather than be flushed over the dam.
13. Further evidence of the retention of ^{137}Cs in the sediments may be found in the specific activities of the sediments. Although a ^{137}Cs sediment burden cannot be determined from existing data, the mean ^{137}Cs activity of the upper 2 inches of 45 cores taken in 1962 through 1963 was 181 pCi per gram of dry weight (range, 1 to 3559 pCi/g). The mean ^{137}Cs activity for three cores taken in 1976 was 224 pCi/g (range, 150 to 278 pCi/g). These data indicate that the ^{137}Cs activities of the sediments have not changed significantly during the intervening years.
14. T. E. Hakonson and F. W. Whicker, *Health Phys.* **28**, 699 (1975).
15. L. D. Eyman and N. R. Kevern, *ibid.*, p. 549.
16. M. A. Wahlgren and J. S. Marshall, *Impacts of Nuclear Releases into the Aquatic Environment* (International Atomic Energy Agency, Vienna, 1975), p. 227; M. A. Wahlgren, D. M. Nelson, E. T. Kucera, "1977 annual report" (Argonne National Laboratory Radiological and Environmental Research Division, Argonne, Ill., 1978).
17. We thank C. Ashley of the Savannah River Plant, Health Physics Group, for allowing us to use specific data, and K. A. Orlandini and D. Warner, Ecological Sciences Section, Argonne National Laboratory, for analytical assistance. Joint contribution from the Savannah River Ecology Laboratory (Institute of Ecology, University of Georgia) and the Savannah River Laboratory (Savannah River Plant, E. I. du Pont de Nemours and Company). This research was supported in part by contracts EY-76-C-09-0819 and AT(07-2)-1 of the U.S. Department of Energy with the University of Georgia and E. I. du Pont de Nemours and Company, respectively.

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Biological Control of Dissolved Aluminum in Seawater: Experimental Evidence

Abstract. *Experimental evidence supports the hypothesis that the concentration and distribution of dissolved aluminum in ocean water are controlled by biological activity in the surface waters. The growth of the diatom* Skeletonema costatum *in artificial seawater media spiked with aluminum reduced the aluminum concentration to that actually found in surface open ocean waters (about 0.5 micrograms per liter). Furthermore, aluminum had a catalytic and limiting effect on the growth of the diatoms.*

The concentration of dissolved Al in surface open ocean waters is less than 1 μg/liter (*1*). The mechanisms that maintain this low concentration are poorly known. In the past it has been assumed that the seawater Al concentration was the result of thermodynamic equilibria between the water and various aluminosilicate minerals (*2*). Several investigators attempted to reproduce these observed seawater Al concentrations either by the dissolution or precipitation of aluminosilicate minerals in aqueous solutions or by the adsorption of dissolved Al onto various types of suspended particles (*3*). However, in these experiments equilibrium concentrations of dissolved Al below 1 μg/liter were not obtained.

Recently, it was suggested that the dissolved Al concentration in the oceans is controlled by biological activity. Mackenzie *et al.* (*4*) observed a depletion of dissolved Al in the photic zone of the Mediterranean Sea parallel to an observed depletion in dissolved silica. This covariance was thought to be due to biological uptake of Al and Si by siliceous organisms. They concluded that "the silicon and aluminum cycles in the oceans are linked through the activity of diatoms" (*4*, p. 680). Van Bennekom and van der Gaast (*5*) identified authigenic smectites regularly distributed in the frustules of diatoms collected at sea. Lewin (*6*) suggested that Al and Fe are necessary to the formation of diatom frustules because the adsorption of these cations onto the silica frustules renders them resistant to dissolution.

To verify the hypothesis that dissolved Al concentrations in surface open ocean waters can be related to the activity of diatoms, I carried out culture experiments using the diatom *Skeletonema costatum*, a common neritic species, grown in an artificial seawater culture medium spiked with dissolved Al. The Al concentration in the medium was monitored during the development of the culture; even in a medium to which 25 μg/liter had been added, the dissolved Al concentration was reduced to less than 1 μg/liter during the growth of the diatoms.

The basic culture medium used in the