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Marine Plankton as an Indicator of Low-Level Radionuclide Contamination in the Southern Ocean

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ABSTRACT

We have initiated an investigation of the utility of marine plankton as bioconcentrating samplers of low-level marine radioactivity in the southern hemisphere. A literature review shows that both freshwater and marine plankton have trace element and radionuclide concentration factors (relative to water) of up to 10^4. In the years 1956-1958, considerable work was done on the accumulation and distribution of a variety of fission and activation products produced by the nuclear tests in the Marshall Islands. Since then, studies have largely been confined to a few selected radionuclides, and by far most of this work has been done in the northern hemisphere. We participated in Operation Deepfreeze 1981, collecting 32 plankton samples from the U.S. Coast Guard Cutter Glacier on its Antarctic cruise, while Battelle Pacific Northwest Laboratories concurrently sampled air, water, rain and fallout. We were able to measure concentrations of the naturally occurring radionuclides ^{7}Be, ^{40}K and the U and Th series, and we believe that we have detected low levels of ^{144}Ce and ^{95}Nb in seven samples ranging as far south as 68°. There is a definite association between the radionuclide content of plankton and air filters, suggesting that aerosol resuspension of marine radioactivity may be occurring. Biological identification of the plankton suggests a possible correlation between radionuclide concentration and foraminifera content of the samples.

INTRODUCTION

Atmospheric nuclear tests, reactor operations and waste disposal programs have injected significant quantities of radionuclides into the marine environment of the northern hemisphere. These releases have in general been documented, and considerable research has been done to characterize the fate of this radioactivity. The southern hemisphere, by contrast, has been generally free of radionuclide input with the exception of the nuclear tests in the Tuamotus and some limited discharges of reactor cooling water. Relatively little sampling of the marine environment has been done, especially in the open ocean, and only a few studies of atmospheric radionuclide concentrations, principally at Antarctica, have been carried out. We initiated this study to determine the levels of natural and anthropogenic radionuclides in the marine environment of the southern hemisphere, to identify and characterize their sources, and to develop more effective sampling techniques.

Since the southern hemisphere is nearly all ocean, any radionuclides that are released will likely find their way into the marine environment where rapid dispersion and dilution will make them difficult to detect. While the fate of large releases either to the atmosphere or directly to the ocean could possibly be predicted by computer models of atmospheric and oceanic transport in time for concentrated samples to be collected, chronic low-level sources cannot be so characterized and their contribution to the marine environment is not predictable. Fortunately, marine plankton, especially the phytoplankton, have large concentration factors for many trace elements and their radioactive isotopes. Furthermore, since they have relatively short lifetimes and are in constant equilibrium with the water in which they drift, they are excellent tracers of a particular water mass. If the origin of the water mass and influence of currents is known, conclusions can be drawn regarding the probable origin of the radioactivity. The marine plankton are particularly sensitive monitors of most anthropogenic radionuclides, having concentration factors of several hundred to several thousand for many elements.

The characteristics of the radionuclides detected can be used to infer their source (for example, neutron-induced activities, fission products or waste processing), while isotope ratios can give information regarding the age and origin of the debris. Truly quantitative measurements are difficult because concentration and separation factors are not well known and are probably quite variable.

The influence of plankton in trace metal recycling and the resultant effects on the composition of the underlying water column and sediments is well documented. They are the first step in the concentration of trace and radioactive elements in the marine and ultimately human food chain, while their decomposition products are major contributors to the sea surface microlayer and to the marine aerosol. Radionuclides in plankton can be used as tracers to give more information concerning some of these processes.

REVIEW OF THE LITERATURE

The principal conclusion of our computer-based literature search was quickly reached. While there has been considerable research on plankton trace element and radionuclide accumulation in the northern hemisphere, there has been virtually none in the southern hemisphere. What follows is a discussion of books and articles that we feel have particular bearing on the problems of low-level radioactivity monitoring in the Southern Ocean.

No literature review, regardless of scope, would be adequate without mention of the 1964 book by Polikarpov[1] that has become the classic in the field. Since then, several reports[2,3] have appeared that actually propose the use of various biological organisms to monitor contamination by radionuclides, trace elements and even hydrocarbons. Ref. 2 is especially complete, containing a good discussion of reasons for using bioaccumulators rather than other techniques such as ion exchange resins. The difficulties inherent in using bioaccumulators, requirements for good bioaccumulators, and studies being undertaken at the time are also discussed.

By far most of the experience and data with a variety of radionuclides in marine plankton were acquired during the nuclear atmospheric test series at the Pacific Proving Grounds, Enewetak and Bikini in the Marshall Islands, from 1954 to 1958. All of these data are contained in a series of reports[4-7] and summarized in a very interesting and readable book.[8] While there are far too many data from the Pacific programs to review, mention of a few significant experiments and results will give an indication of the potential utility of plankton as radionuclide monitors.

On May 16, 1958, the Wahoo event was detonated underwater two miles southwest of Enewetak. Plankton sampling was begun as soon as possible, and at H + 6 hours the major part of the total radioactivity was found in the top 25 m and about one-eighth at the thermocline, 110 m. By H + 28 hours the activity was distributed through the upper half of the mixed layer to about 50 m, but by H + 48 hours it was concentrated at 100 m, the upper edge of the thermocline. At no time was the activity uniformly mixed; it was always stratified. Gamma-emitting short-lived fission products were dominant immediately following the explosion, and were (from May 16 to 20) ^{99}Mo and ^{132}Te - ^{132}I. Large plankton contained ^{140}Ba, while the smaller plankton had higher percentages of ^{99}Mo-^{99}Tc. Measurements of the relative total radioactivity in filtered water and plankton showed the water inventory to be 3000 - 11,000 times higher, indicating that plankton played a small part in removing the radioactivity. This was because the total amount of plankton was very small compared with the water. Naturally occurring trace elements in seawater such as Zn, Co, Ru, Cs, Sr and I are usually present in solution, and their radioactive isotopes were found only in trace quantities in plankton. Those with the least solubility, such as ^{95}Zr and ^{144}Ce were found in the greatest amounts (Ref. 8, pp. 278-285).

While the experiments following the Wahoo shot were confined to a small area and involved high levels of many fission products, other surveys were undertaken

to look for low levels of radionclides throughout the North Pacific ocean. A local survey[4] of the Pacific Proving Grounds during the test period in June 1956 consisted of a grid of stations 45 miles apart covering an area of 78,000 sq. mi. Radioactive materials were found in plankton samples from every station with the average concentration 7100 times the average surface water concentration. A much more extensive survey involving lower levels of radioactivity had been carried out the year before between March 13, 1955, and May 3, 1955, one year after the tests in the spring of 1954; this was Operation Troll.[5] The Coast Guard Cutter Roger B. Taney left Kwajalein on March 13, 1955, sampled the area of Enewetak-Bikini and arrived in Guam on March 22, taking samples along the way. The ship then proceeded northwest to about 22°N 139°E, then southwest to the Philippines. From there the ship sailed to Okinawa, on to Tokyo, and departed on April 21 from Yokosuka for San Francisco, arriving on May 3. Samples were collected all the way. The overall average concentration factor for plankton relative to water was 300 (range 100-1000) and 80-90% of the activity was due to ^{144}Ce. This nuclide and ^{95}Nb constitute 26% and 14% respectively of the total radioactivity of one-year-old fission products and essentially all of the gamma radioactivity.[9] The lowest concentrations were found near the Pacific Proving Grounds and the highest in the western Pacific around the Philippines and Okinawa. The microplankton, less than 0.1 mm, generally had the highest specific activity. Some pertinent conclusions of Operation Troll were: "The high specific activity of plankton compared with seawater makes it a simple measure of the distribution of activity" (p. 11) ... "Measurements of plankton activity offer a sensitive indication of activity in the ocean ..." and ..."Similar operations would be valuable in assessing the activity from future tests and in gathering valuable data for oceanographic studies" (p. 13).

Another survey[6] conducted from Sept. 1-20, 1956, six weeks after the last test in the 1956 series, reported results similar to those cited. They also found a reduction in the concentration factor with time from 7000 to 300 and attributed it to a wider spectrum of radionuclides available at early times. Two other important conclusions were that plankton appears to be the most sensitive indicator of radioactivity in the sea, and microplankton is probably the principal source of radioactive particulate matter. Nuclides detected were $^{89,90}Sr$, $^{137,140}Ba$, ^{144}Ce, ^{103}Ru, ^{106}Ru-^{106}Rh, ^{95}Zr, $^{57,58,60}Co$, ^{65}Zn, ^{59}Fe and trivalent rare earths. The last report in the series[7] gives the results of some radiochemical separations made on plankton samples collected in and west of the Proving Grounds in September, 1956. "Fission products, mainly ^{95}Zr-^{95}Nb and ^{144}Ce-^{144}Pr, contributed an average of 29% of the total radioactivity. The remaining 71% of the activity was contributed by the nonfission radioisotopes ^{65}Zn, $^{57,58,60}Co$, ^{55}Fe and ^{54}Mn [in quantities of 24, 26, 21 percent and trace respectively]. Variations in ratio of occurrence for the different nonfission products with change in geographical location were observed." This last observation was no doubt due to differences in source terms for activation products from different locations.

The last atmospheric test series, Operation Dominic, was conducted in 1962 at Christmas Island, and the biological investigations are summarized in Ref. 10. Even though no tests were conducted in which the fireball touched the surface, so that little fallout was produced, plankton sampling showed elevated levels of a few fission products. The utility and sensitivity of plankton sampling were emphasized in the report by the facts that not only were ^{95}Zr-^{95}Nb, $^{141,144}Ce$ and other radionuclides detected, but the levels increased as the test program continued. Analysis of seawater samples revealed no increase in either the number of radionuclides or in the levels of radioactivity during the period of the study. In fact, two-thirds of the water samples showed no detectable levels of radioactivity above background, and most of those that did had counting errors greater than 50%.

Since the cessation of nuclear testing in the atmosphere, reports have been concerned with inputs from world-wide fallout, nuclear fuel reprocessing wastes, releases from nuclear power plants, specific isotope studies and some related data from stable trace element studies. Ref. 10 includes studies of Mn, Co, Ag and Sb isotopes with a discussion of their correlation in various marine organisms. Ref. 11 reports on two cruises taken to monitor leakage from the Nuclear Energy Agency (NEA), part of the Organization for Economic Cooperation and Development, dumping sites off the west coast of Spain. An interesting conclusion from this study is that the rate of biological vertical transport downwards is of the order of magnitude of some years.

Several papers[12-14] have been concerned with ^{239}Pu and other transuranic and alpha-emitting radionuclides. The paper by Pillai et al.[12] reports concentration factors for ^{239}Pu from "red tide" organisms (660), green alga (1570), giant and palm kelp (770 and 1080) and mixed coastal zooplankton (2590). The paper by Gromov[13] reports the results of experiments in the partitioning of the radionuclides ^{239}Pu, ^{106}Ru, and ^{99}Tc by both natural and single species phytoplankton populations and ^{137}Cs by krill. The mechanism of accumulation of ^{239}Pu and ^{210}Po by the brown algae Ascophyllum nodosum and marine plankton was studied by Zlobin and Mokanu.[14] They demonstrated that the accumulation of ^{239}Pu by marine algae is an active process involving energy expenditure by the cell. Fowler and Aston[15] used ^{235}Np tracer to investigate the potential uptake of ^{237}Np by marine zooplankton. They showed that Np exists almost entirely as a soluble species in seawater and is less available for uptake than either plutonium or americium. The uptake is probably related to surface adsorption processes, and when Np is ingested with food it is not effectively assimilated by a euphausid crustacean. A survey of the total alpha-activity of marine plankton by Shannon[16] showed that most of the activity was due to the presence of unsupported ^{210}Po, 5-6 pCi/g

dry. Thorium isotopes were variable, but the uranium series elements other than ^{210}Po were about 0.6 pCi/g dry zooplankton and 1.8 pCi/g dry phytoplankton. The wet-to-dry ratio averaged 16. Another survey of the relative amounts of natural and man-made alpha emitters in plankton was made by Paschoa et al.[17]

A study by Sick and Baptist[18] addressed particularly the concentration of Cd in the marine food chain and showed that the marine copepod Pseudodiaptomus coronatus incorporated ^{109}Cd at higher rates from ambient water than from phytoplankton food. A very complete study by Knauss and Ku[19] of the elemental composition of plankton and the content of natural radioactive decay chain nuclides reported not only concentration factors but cross-correlation coefficients calculated for all possible pairs of elements studied. They suggested that a biologic mechanism is likely as an explanation for the Ce depletion in seawater relative to the other rare earth elements, based on its high concentration factor in marine plankton. Not all trace elements are concentrated by the plankton as indicated by the Np work already cited. Two papers[20,21] report the non-accumulation of ^{95m}Tc in either the IV or VII oxidation states. Tc, like Np, is predominantly in solution as an ionic species in seawater.

When it comes to the southern hemisphere, very little work has been reported, especially regarding artificial radionuclide uptake. The work of Shannon[16] in the area of the Cape of Good Hope was concerned only with natural decay series isotopes. The paper by Kolesnikova et al.[22] reports finding $^{141,144}Ce$, ^{95}Zr-^{95}Nb and ^{103}Ru in zooplankton collected in the region of Mururoa Atoll, the French nuclear testing area. Jennings[23,24] reports ^{55}Fe in plankton increasing from below detectable levels at 20° N up to 130 nCi per gram of iron at 20° S and then decreasing along a track at 135° W longitude. The maximum is near Mururoa (22°S 139°W). He remarks that: "The paucity of data on radioactivity in marine organisms from the Southern Hemisphere is a serious deficiency because atmospheric nuclear tests are actively

being carried out there."[24] Ref. 23 is a discussion of the selective uptake by marine plankton of ^{55}Fe relative to other forms of stable Fe. It is interesting that our keyword search did not uncover a single reference similar to the University of Washington Fisheries Laboratory (UWFL) series on marine radioecology in connection with the Mururoa tests.

Freshwater organisms also have high concentration factors for many elements, and our literature search found several interesting references. The survey by Blaylock[25] summarizes the availability of data on concentration factors for use in dose assessment in the transfer of radionuclides from the aquatic environment to man. Factors influencing the variability of bioaccumulation concentration are identified, uncertainties associated with the validation of these factors are discussed, and some guidance is given for collecting data and making measurements. Aoyama et al.[26] followed the uptake and release of ^{137}Cs, ^{65}Zn, ^{60}Co and ^{45}Ca by a fresh-water phytoplankton. They found that the uptake is a biological process dependent on light and the growth phase. Marciulioniene et al.[27] also found that the accumulation of ^{90}Sr, ^{144}Ce, ^{137}Cs and ^{106}Ru in Charophyta depends on the species and that the accumulation of ^{90}Sr and ^{137}Cs depends on sampling time and vegetative period. Jenckes[28] investigated the feasibility of developing a quantitative method for measuring low levels of radionuclides in fresh-water systems through bioaccumulation, particularly by phytoplankton. An interesting report by Millard et al.[29] presented the results of an investigation of the radionuclide content and cycling in the leaching ponds adjacent to the Test Reactor Area located at the Idaho National Engineering Laboratory. These ponds were small enough and inputs were known well enough to do material balance studies among the water, surface sediments, seston, zooplankton, benthic periphyton, macrophytes, littoral vegetation, arthropods and small mammals as a function of seasonal variation. As an indication of the potential various organisms have for use as bioaccumulators,

the paper by Davis[30] discusses the accumulation of several radionuclides by aquatic insect larvae as well as plankton in the Columbia River. The concentration factors were dependent on the growth stages and water temperature but were often 1000 or more for the nuclides ^{32}P, ^{51}Cr, ^{64}Cu, ^{65}Zn and ^{76}As.

Our literature review to date has served to orient us with respect to the types of work being done, the methods used and results achieved. This helps us to plan our own research and gives some indication of how we might proceed and results we can expect.

ADVANTAGES AND LIMITATIONS OF BIOLOGICAL ACCUMULATORS

From both the literature review and our own experience in radiochemical analysis, we can identify areas of strength and weakness in the use of plankton as a monitor for low levels of radioactivity in the sea. On the positive side, the various reports from the UWFL work show that uptake is rapid relative to dispersal and dilution, and once it has occurred, recycling keeps the radionuclides in the biotic layers and retards losses. These processes also assure us that the plankton is a unique tracer for a particular water mass as evidenced by the great distances over which it was followed by the University of Washington groups. The concentration factors are high enough to give good sensitivity; our experience indicates plankton sampling provides a detection capability of the order of 0.1 pCi per liter of sea water. Furthermore, these high concentration factors exist for a wide range of nuclides, the principal exceptions being only those species in true solution and the major elements in sea water. From a more practical standpoint, we can collect plankton in the open ocean, without infringing on any political boundaries, and with a knowledge of ocean current patterns we can infer the origin of the water mass being sampled. Once a sea-going vessel is acquired, further equipment for plankton sampling is very simple, techniques are well worked out, and the final samples are easily transported, stored, and analyzed.

The principal disadvantage of plankton, or any bioaccumulator, is that calibration sufficiently accurate to produce quantitative data is very difficult. While some attempts have been made and procedures suggested[2,3,28] for calibration, the concentration factors may depend on uncontrollable or unknown circumstances, such as the species involved, growth stages and bioavailabilty. While ratios between isotopes of the same elements should be consistent, even here caution should be exercised as evidenced by the ^{55}Fe specific activity results of Jennings.[23] Isotope discrimination effects are probably reduced over time, and we would expect that as long as the radionuclides are all of approximately the same age, differrences should be small. Isotope ratios can be useful in estimating the age of the radioactivity and its origin, since the isotopic mixture in nuclear explosion debris is considerably different from that found in reactor or reprocessing effluent. In any case, with experience and some idea of the concentration factors involved, initial plankton analysis could be used to determine locations and procedures for sea water analysis by more quantitative techniques.

PROJECT DESCRIPTION

Our initial goals for the program were principally to obtain experience at plankton sampling, acquire the necessary equipment, establish contacts with the oceanographic community related to our work, and collect and analyze some samples from the southern hemisphere. We needed to investigate both scientific and logistical problems. Our analysis was to be principally by low-level γ-spectroscopy on entire samples, and we were concerned that natural levels of the U and Th series as well as ^{40}K might raise the background sufficiently to make detection of low levels of artificial radionuclides difficult or impossible.

We were funded concurrently with Battelle Pacific Northwest Laboratories (PNL) to make an investigation of the problems. PNL arranged for space on the U.S. Coast

Guard Cutter Glacier (Fig. 1) for its "Operation Deepfreeze 1981" and provided personnel for most of the sampling trip. The general route of the ship after departure from Long Beach, California, in November 1981, was to Hawaii, New Caledonia, the east coast of Australia, New Zealand, the Ross Sea, eastward along the ice to Palmer Peninsula and after several trips back and forth between Palmer and Punta Arenas, north along the west coast of South America and back to Long Beach in May of 1982 (Fig. 2). Battelle had its own personnel on board from Hawaii to New Zealand, and the remainder of the samples were taken by a Coast Guard marine technician trained by Battelle. In addition to our plankton samples, PNL also collected air filter samples, fallout, and large-volume water samples. The following year we participated in a similar project, "Operation Deepfreeze 1982," which will be the subject of Part 2 of this report.

Figure 1. U.S. Coast Guard Cutter Glacier.

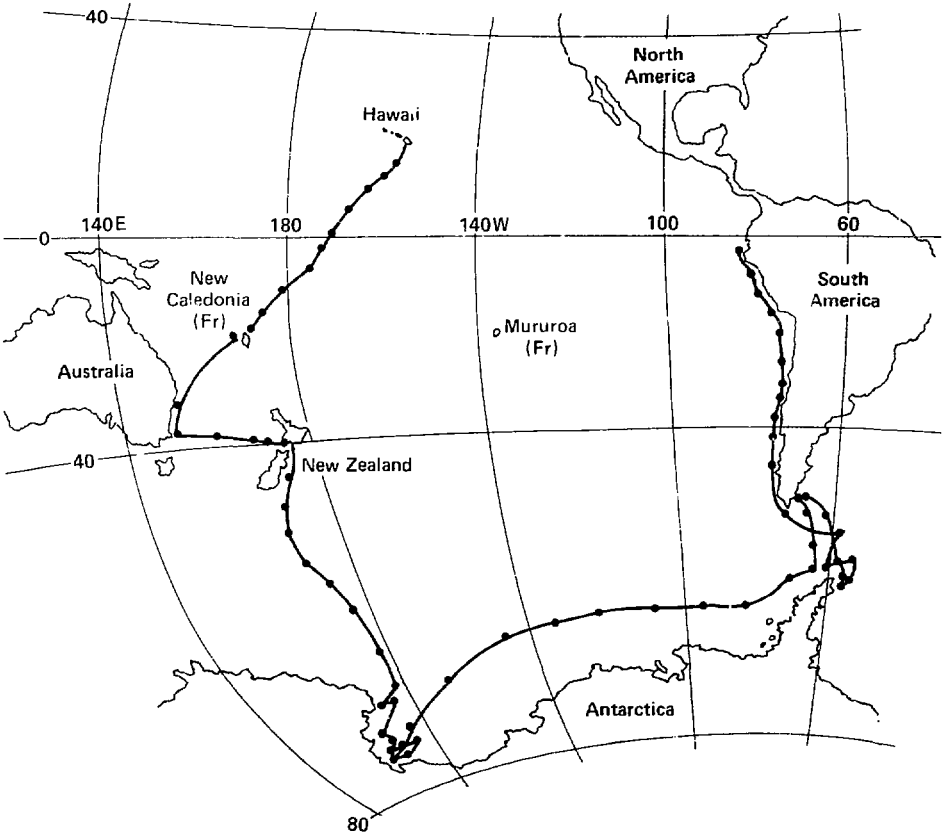

Figure 2. Cruise track for Operation Deepfreeze 1981. Dots indicate a sampling station of some type (air, fallout, rain, plankton, water).

METHODS

Funding for this initial project began as of October 1, 1981, which left us little time to acquire equipment for plankton collecting. We were able to assemble barely the minimum necessary and transport it to Honolulu in time to load aboard the Glacier by the first week in November.

All the plankton samples were collected by pumping water through the ship's fire mains and filtering it through a No. 10 plankton net suspended in a 55-gal drum chained outboard to the side of the ship. The net originally supplied was longer than the barrel was deep so the cod end was led back up and over the edge of the drum (Fig. 3). Later samples were collected in nets constructed to fit the drums that we were able to obtain and send to the ship in Wellington. The fire mains delivered about 250 gal/min directly into the net through a 3-in. canvas hose. The water flowed through the net into the drum, which when full acted as a cushion, and then simply overflowed into the sea. Pumping was generally carried out over a 24-h period, but various problems caused by other shipboard operations often resulted in the pumps cycling on and off or in reduced flow, requiring longer sampling times. Volumes were estimated by periodically reading the water pressure after rough calibration of the system by noting the time required to fill the drum at different pressures. Sample location and total volume were estimated by observing the time and position when sampling was started and concluded. While this arrangement was far from ideal, it produced fairly consistent samples after several days of practice. On two occasions the barrel and net were lost because of heavy seas.

After the samples were collected from the net, they were preserved in 5% buffered formalin in sea water for storage and transport. Volumes of sea water filtered varied from 250,000 gals to over one million gal in two cases with the average around 350,000 gal. Plankton wet weights were highly variable ranging from 25 g to 400 g with the average around 200 g. As the samples were collected, 25-ml subsamples were taken for species identification.

Figure 3. Plankton net setup for pumping. Note fire hose input at top left.

Dr. Wim Kimmerer of the University of Hawaii Institute of Marine Biology performed the species identification on the initial 11 samples. He first counted large organisms such as euphausiids in the entire sample, then took aliquots using a plankton splitter and counted everything in this aliquot. All counts were converted to number of individuals per original subsample. Professor John Wormuth

of Texas A&M University, Department of Oceanography, identified the species in the remainder of the samples. He first sorted them by size, using Nytex screens, and counted total organisms, or if there were too many to count in a single size fraction, took an aliquot with a plankton splitter.

The first 11 samples were returned to Livermore from New Zealand by air in mid-January 1982; the remainder returned with the Glacier in mid-May. The samples were drained on a Nytex screen and weighed wet without rinsing. The plankton was recombined with the drained liquid and the whole sample taken to dryness in an oven at 110°C. After drying, the samples were ground and homogenized with a mortar and pestle and packed into a suitable standard container for γ-spectroscopy. All samples were counted on our low-background Compton-suppressed γ-spectrometer[31] for at least 10^4 min. The resulting spectra were computer-analyzed by the GAMANAL[32] code and results expressed as pico Curies per gram wet weight. Radionuclides were all decay-corrected to the common time of December 1, 1981. This is a standard procedure in radioecological studies that facilitates comparison of radioactivity levels among samples collected over an extended time period and does not imply that December 1, 1981, was the date of production of any of the radionuclides.

We were able to calculate upper limits, or detection limits, for any radionuclide not positively identified and measured. Because it was not feasible to calculate limits for all possible radionuclides that could be expected, we settled on those that were positively identified in at least one sample (^{7}Be, ^{95}Nb, ^{144}Ce), and a few more that were known to have high concentration factors in plankton (^{54}Mn, ^{65}Zn, ^{103}Ru). ^{241}Am was included in the upper limit calculations because it is an indicator of ^{239}Pu, which is of considerable interest to marine radioecologists. We did not calculate upper limits for any of the naturally occurring radionuclides, because their detection is not indicative of anthropogenic contamination. A detailed description of the procedure used by GAMANAL

to calculate upper limits is given in Ref. 33, but a brief discussion will serve to show the type of upper limit used. If no peak is found in the γ-spectrum corresponding to the known energy of one emitted by the nuclide in question, the counts in the channels where the peak should be (due principally to the Compton continuum from higher energy peaks) are summed, the square root taken and multiplied by two. This quantity is then corrected by subsequent calculations for geometry, absorption, and decay and a concentration calculated just as if a peak had been found. The philosophy behind the method is that if, in the region of interest, counts exceeding twice the standard deviation of the background had been present, the peak-finding routine of GAMANAL would have identified a peak. For nuclides emitting more than one γ-ray, up to four such regions can be used, and the lowest value reported as the upper limit.

No significant oceanographic data were taken on this cruise as we were not equipped to make the measurements and only weather observations were taken by the Coast Guard. The Battelle air samplers were mounted as high as possible and run continuously with filter paper changed at irregular intervals. Battelle also collected dry fallout on a 6-m^2 corrugated aluminum collector by washing the surface with a spray of water and collecting it along with any rain water every 24 hours. Battelle also collected high-volume water samples at the rate of 30 l/min with a sampler of their own design. It will publish its own report on the results of its measurements.

RESULTS AND DISCUSSION

The locations of all the plankton samples and the results of the γ-spectrometry analyses are given in Table 1. The two most interesting radionuclides detected are ^{144}Ce and ^{95}Nb. In most cases, both these nuclides were measured at concentrations above upper limit values; in other cases they were near detection limits.

In the case of ^{144}Ce, only the gamma ray at 133.5 keV was detected. This gamma-ray has an overall abundance of 11% in the ^{144}Ce decay scheme; while there are others, their abundances are less by factors of 7 to 38. At the count rates we found for the 133.5 keV photon, we could not expect to find those with lower yields. We believe the ^{144}Ce to be real because there are no other reasonable candidates for the 133.5-keV photon, the ^{144}Ce is found by Battelle in its air filters, and it is known to have a high concentration factor[4-7] in plankton. The ^{144}Ce results for plankton along with the air filter data are shown in Fig. 4.

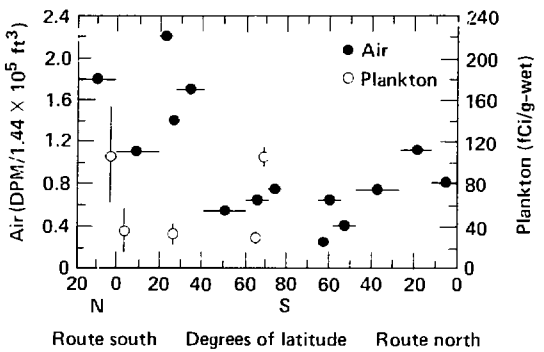

Figure 4. ^{144}Ce concentrations in latitude bands. Horizontal bars indicate distance covered during collection. Vertical error bars are 1 σ counting error (not available for air).

The air filter samples were collected continuously and sometimes over a considerable distance, while the plankton samples were generally collected over only a relatively short distance. Because the stations are plotted at the midpoint of the collection, they are not directly comparable on a station-by-station basis. In Fig. 4, the horizontal bars indicate the distances sailed during the collection. The abscissa

shows degrees of latitude, with the left side the route south, and the right side the return cruise north. There is a general relationship between the ^{144}Ce detected in the air and in the plankton. The higher levels in the air from about 20°N to 40°S on the western side of the basin (route south) are reflected in plankton from similar locations. Once the ^{144}Ce concentration falls below about 1.1 dpm/sample (normalized) in the air, however, it is not observed in the plankton (presumably below detection limits) except for the region between 60° and 70°S. ^{144}Ce in plankton in this band may represent higher oceanic content independent of local atmospheric input, as ^{144}Ce and other radionuclides were detected by Battelle in water but not in air samples from this area. These other radionuclides were ^{110m}Ag (2-5 dpm/m^3), ^{155}Eu(3 dpm/m^3), and ^{241}Am($3dpm/m^3$). The overlapping locations between 45° and 65°S on the east side of the basin (route north) represent samples collected during several transects of Drake Passage and indicate the range of concentration observed.

Figure 5 shows the equivalent data for ^{95}Nb. The arguments for and against the detection of ^{95}Nb are somewhat more complicated than those regarding ^{144}Ce. The only gamma ray detected from ^{95}Nb is the 765.8-keV photon, an energy similar to those of ^{238}U (actually its granddaughter ^{234}Pa) at 766.4, ^{102m}Rh at 767.0, and ^{226}Ra at 768.4. ^{226}Ra can be eliminated on several grounds: the energy is too different to actually constitute an interference given our resolution; the GAMANAL program would have found other Ra γ-rays that are much more abundant, and used them for appropriate corrections, and finally, Ra is not particularly concentrated by plankton. Several samples had detectable levels of ^{226}Ra with only average upper limits calculated for ^{95}Nb. The ^{102m}Rh can be eliminated on two grounds: it also has other γ-rays in much higher abundance that were not detected, and it is neither a fission product nor a common reactor product. This leaves the ^{238}U (^{234}Pa) as the principal candidate for interference. As can be seen in Table 1, all the positive results for ^{95}Nb, except Sample 39, are associated with the higher levels of

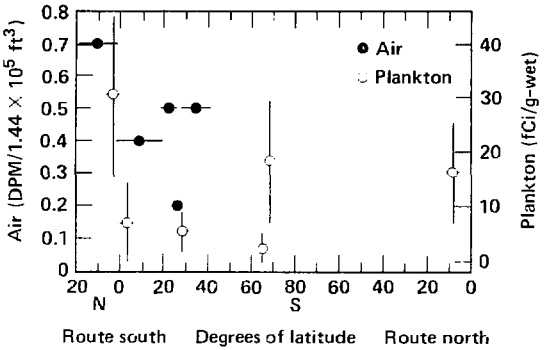

Figure 5. ^{95}Nb concentrations in latitude bands. Horizontal bars indicate distance covered during collection. Vertical error bars are 1σ counting error (not available for air).

^{238}U. However, our experience with the GAMANAL program over many years and hundreds of samples, such as Livermore Valley soil containing ^{238}U but no ^{95}Nb, has convinced us that the program accurately accounts for the 766.4-keV γ-ray of ^{238}U by means of the associative γ-rays at 63.3, 92.6, and 1001.1 keV when counting statistics are good. In the plankton samples the total counts in the 766 peak are very low, so the peak errors are large and the shapes not usually perfect. To be on the safe side, ^{95}Nb is only reported where the excess of the 766.4 peak over that contributed by ^{238}U is 50 percent or more. Another slightly disturbing finding is that we do not detect the gamma rays from ^{95}Zr, the parent of ^{95}Nb. There could be several reasons for this, any one of which would be sufficient: the γ-rays from ^{95}Zr are lower in abundance by about a factor of two, the equilibrium disintegration rate of ^{95}Zr is less than half that of the ^{95}Nb and there is some indication that more ^{95}Nb than ^{95}Zr is released to the marine environment. Because the ^{95}Nb is barely detectable, any factor of two less for the ^{95}Zr would make it undetectable. In fact, upper-limit values calculated for ^{95}Zr are consistent with the ^{95}Nb measured.

Table 1.
Radionuclides in plankton.[a,b]

Sample	Be-7	K-40	Mn-54	Zn-65	Nb-95	Ru-103	Ra-226	Ra-228	U-238	Am-241	Ce-144	Th-228	U-235	Location
half-life	53.6d	1.27 x 10^9 Y	312 d	245 d	35.1d (64 d)	40d	1622 Y	5.8Y	4.51 X10^9 Y	433 Y	285 d	1.40 X10^{10} Y	7.04 X10^8 Y	
1	<0.148	1.88 ±10	<0.008	<0.017	0.0306 ±26	<0.023	--	--	6.31 ±8	<0.062	0.107 ±42	--	--	3N177W
2	0.174 ±35	1.31 ±8	<0.004	<0.0010	0.00706 ±35	<0.014	0.0156 ±50	0.022 ±80	1.97 ±37	<0.031	0.0364 ±60	--	--	6S177E
3	<0.108	2.20 ±5	<0.002	<0.004	<0.008	<0.066	--	--	0.592 ±7	<0.014	<0.017	--	--	15S173E
4	<0.257	1.24 ±6	<0.004	<0.009	<0.018	<0.024	--	--	0.254 ±25	<0.020	<0.038	--	--	24S164E
5	<0.102	1.20 ±7	<0.002	<0.002	<0.006	<0.064	--	--	0.1754 ±16	<0.010	0.0324 ±28	--	--	26S158E
6	0.368 ±14	1.11 ±5	<0.002	<0.006	0.00534 ±68	<0.009	0.0091 ±80	0.017 ±32	1.98 ±17	<0.021	<0.026	--	--	28S154E
7	0.360 ±20	1.45 ±5	<0.001	<0.003	<0.004	<0.049	--	--	0.183 ±11	<0.009	<0.010	--	--	32S152E
8	0.275 ±14	1.01 ±4	<0.001	<0.003	<0.003	<0.006	0.0077 ±20	0.014 ±31	1.04 ±7	<0.010	<0.014	--	--	35S156E
9	0.149 ±14	1.12 ±7	<0.003	<0.008	<0.009	<0.016	--	--	1.30 ±7	<0.024	<0.034	--	--	36S159E
10	0.292 ±13	1.41 ±14	<0.0005	<0.001	<0.002	<0.018	--	--	0.187 ±18	<0.004	<0.006	--	--	37S161E
11	0.450 ±17	1.15 ±6	<0.003	<0.008	<0.002	<0.016	0.0142 ±33	--	2.42 ±7	<0.023	<0.034	--	--	39S168E
13	<0.148	1.92 ±3	<0.001	<0.003	<0.011	<0.008	0.284 ±7	--	0.0284 ±7	<0.006	<0.010	--	--	43S174E
14	<0.626	4.29 ±7	<0.005	<0.014	<0.040	<0.582	0.0422 ±25	--	0.635 ±14	<0.027	<0.042	--	--	46S172E
16	<0.185	1.22 ±5	<0.001	<0.003	<0.011	<0.168	0.00802 ±68	0.0154 ±41	0.0943 ±17	<0.005	<0.008	--	--	54S170E
17	<0.178	1.54 ±3	<0.001	<0.002	<0.011	<0.165	--	--	0.0430 ±26	<0.004	<0.008	--	--	58S173E
18[c]	<0.139	1.06	<0.0008	<0.002	<0.008	<0.162	0.0059 ±13	--	0.0600 ±18	<0.004	<0.006	--	--	175E 61.5S
18A	<0.475	1.04 ±3	<0.0007	<0.002	<0.021	<0.938	0.0051 ±40	--	0.0412 ±17	<0.0025	<0.006	--	0.0026 ±50	175E 61.5S

[a] The units are pCi/g of wet plankton; values preceded by "<" indicate the upper limit. Values are corrected for decay to 12/1/81.

[b] Errors are one standard deviation (in percent).

[c] Sample 18 was divided into two parts for counting purposes.

Table 1 (concluded).
Radionuclides in plankton.

Sample	Be-7	K-40	Mn-54	Zn-65	Nb-95	Ru-103	Ra-226	Ra-228	U-238	Am-241	Ce-144	Th-228	U-235	Location
		+3			+29				+6		+16	+24	+11	65S
19	<0.176	0.888	<0.0006	<0.0016	0.00233	<0.063	--	--	0.427	--	0.0286	0.00394	0.0164	178.5E
		+4			+18			+30	+6		+8	+14	+4	68S
20	<0.349	0.779	<0.0008	<0.002	0.0181	<0.416	--	0.0132	0.820	<0.004	0.1055	0.0179	0.0346	179.7E
		+4							+24					68.8S
22	<0.499	2.37	<0.0021	<0.0063	<0.0296	<0.749	--	--	0.114	<0.011	<0.018	--	--	138.7W
		+7							+15					56.2S
27	<0.194	0.452	<0.0006	<0.0018	<0.0101	<0.262	--	--	0.0588	<0.0026	<0.0051	--	--	63W
		+4												60.2S
28	<0.487	2.09	<0.0016	<0.007	<0.035	<0.780	--	--	<0.077	<0.009	<0.016	--	--	59.2W
		+5					+14		+10			+30	+38	59S
29	<0.202	0.624	<0.0008	<0.002	<0.011	<0.207	0.0143	--	0.197	<0.009	<0.024	0.0082	0.0076	59.2W
		+6						+23	+20					49.9S
30	<0.437	1.06	<0.001	<0.003	<0.020	<0.568	--	0.0145	0.0715	<0.006	<0.010	--	--	74.2W
		+4							+15					46.1S
31	<0.232	0.882	<0.0007	<0.002	<0.013	<0.336	--	--	0.0393	<0.002	<0.005	--	--	74.8W
		+3						+38	+11				+15	42.2S
32	<0.256	1.0	<0.0006	<0.002	<0.015	<0.474	--	0.0077	0.0553	<0.002	<0.005	--	0.00257	74.4W
		+2							+13				+12	37.6S
33	<0.289	1.84	<0.0006	<0.002	<0.012	<0.428	--	--	0.0575	<0.002	<0.005	--	0.00308	73.9W
		+2							+20					34.4S
34	<0.767	3.55	<0.0016	<0.005	<0.039	<1.31	--	--	0.0683	<0.006	<0.013	--	--	72.3W
		+2							+16				+20	31.2S
35	<1.34	5.26	<0.0024	<0.006	<0.065	<2.48	--	--	0.134	<0.008	<0.020	--	0.0058	72.1W
		+4							+18				+18	18.6S
37	<0.720	1.11	<0.001	<0.003	<0.036	<1.56	--	--	0.0839	<0.004	<0.010	--	0.00427	75.8W
		+2							+34					13S
38	<0.787	3.58	<0.0015	<0.004	<0.037	<1.37	--	--	0.0394	<0.006	<0.013	--	--	77.1W
39	<1.54	1.25	<0.0008	<0.003	0.0164	<1.22	0.0028	--	0.110	<0.002	<0.021	--	0.00448	81.0W
		+3							+8				+19	14S
40	<0.865	1.84	<0.001	<0.004	<0.050	<1.71	--	--	0.218	<0.005	<0.036	--	0.00784	84W

our conclusion is that because the ^{95}Nb is associated with the ^{144}Ce in our samples, is known to be highly concentrated by plankton, and is detected by Battelle in the air samples, it is real and its association with the ^{238}U is also real.

As with the ^{144}Ce, there is a relationship between the ^{95}Nb found in the air filters and in the plankton. We find positive indications of ^{95}Nb in both plankton and air samples from the same latitude bands of 20°N to 40°S on the southern leg of the cruise. Plankton, however, show two other positive results: in the same 60°-70°S latitude band as the ^{144}Ce and other radionuclides and at 8°S (Station 39, northwest of Lima), which could represent material in the Peruvian current. This last sample, while lacking confirmation in either the air or water samples, has the lowest ^{238}U concentration of the ^{95}Nb group and the identification is quite positive. Water samples from near this station were reported by Battelle to contain ^{110m}Ag (4.7 dpm/m^3) and ^{155}Eu (5.8 dpm/m^3).

The only published data we could find even remotely confirming our results are those of Kolesnikova et al.[22] from 1969 in the region of Mururoa Atoll. They reported finding ^{141}Ce in plankton at levels of 0.28-5.6 pCi/g-wet, ^{144}Ce at 0.16-4.0 pCi/g wet, ^{103}Ru at 0.2-4.0 pC/g-wet and ^{95}Zr at 0.24-9 pCi/g-wet. These concentrations are much higher than we found because of nuclear testing at the time, but indicate that Mururoa could be a source of radionuclides to the Southern Ocean. Fig. 6 shows all of our plankton stations; those where ^{95}Nb and/or ^{144}Ce were detected are circled.

Figure 7 shows the ^{7}Be concentration in both plankton and associated air filters on a station-by-station basis. ^{7}Be is produced by spallation reactions of cosmic rays on atmospheric constituents at high altitudes and, except for thermonuclear explosions, has no significant anthropogenic source. Because ^{7}Be has a half-life of only 53 days, we were able to detect it only in the samples returned from New Zealand. All the other samples returned with the ship either had very low ^{7}Be concentrations to begin with, or it had decayed below detection

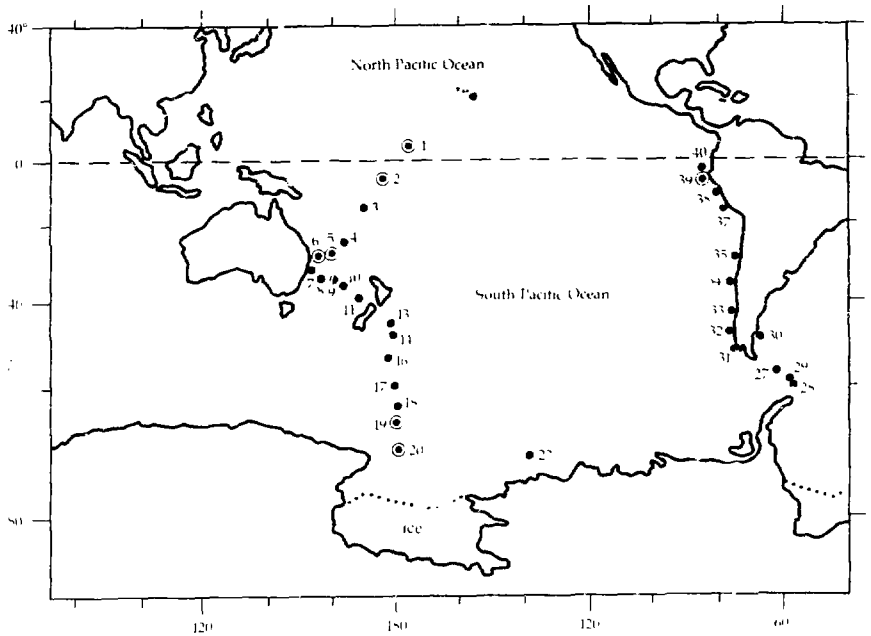

Figure 6. Plankton stations for Operation Deepfreeze 1981. ^{95}Nb and/or ^{144}Ce were detected at the stations circled.

levels before counting (this also could have been the case for ^{95}Nb). The plot does show the association between high concentrations in air and plankton for the trip south. ^{7}Be was not detected in plankton when the air concentration was below about 500 dpm/sample. The average ^{7}Be concentration in plankton between 28° and 39°S was 0.316 ± 0.10 pCi/g·wet, while the average concentration measured by Battelle in filtered sea water from these latitudes was $(114 \pm 34)10^{-6}$ pCi/g. This gives a concentration factor for ^{7}Be of 2770 ± 1200, which is in the range generally observed for these factors, but somewhat higher than the factor of 1000 reported by Lowman et al.[34]

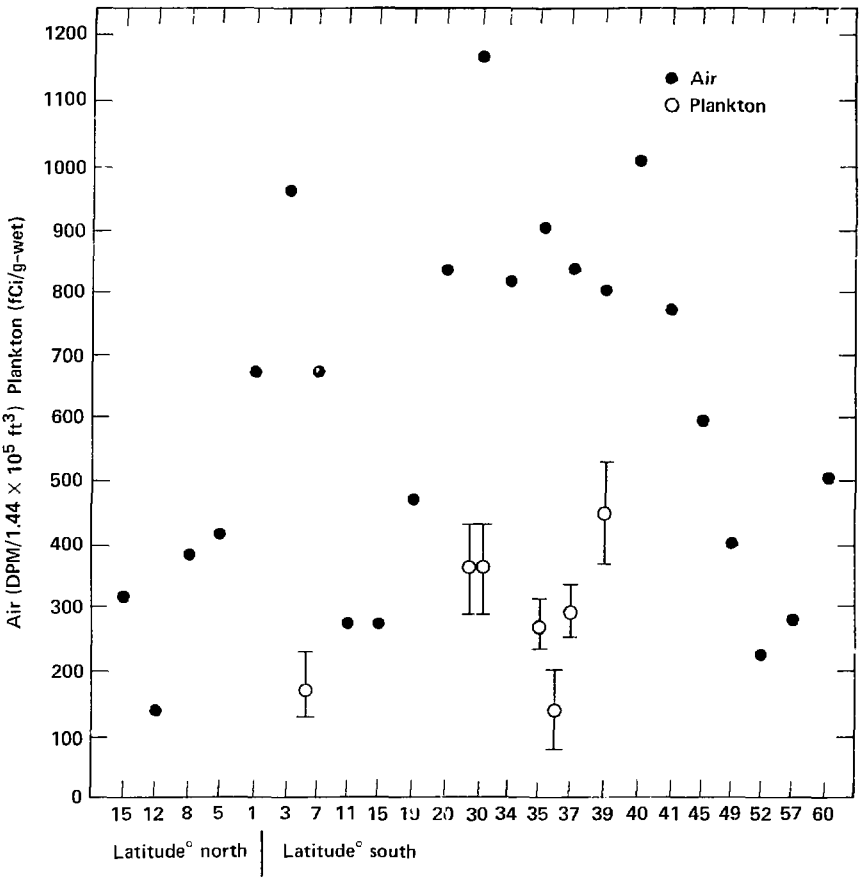

Figure 7. ^{7}Be concentrations at individual stations. Vertical error bars are 1σ counting error (not available for air).

Of the long-lived naturally occurring radionuclides, ^{40}K was detected in all 32 samples, ^{238}U in all but 1, ^{226}Ra in 11, and ^{228}Ra-^{228}Th in 9. The specific activity of ^{40}K is 0.84 pCi ^{40}K/mgK, which gives an average of about 1.4 mg K per gram of plankton vs 0.4 mg K/g of average sea water or a concentration factor of 3-4. Ten of the samples had enough U to enable measurement of the ^{235}U isotope as well as the ^{238}U. The average activity ratio of the ^{238}U to the ^{235}U in these samples was 22.3 in good agreement with that in sea water, 21.8. This implies that if the ratio were much different, for whatever reasons, it would be apparent in the plankton. The average U specific activity in plankton from Table 1 is 0.619 $\pm$ 1.2 pCi/g while sea water is known to be about 0.001 pCi/g. This gives an average concentration factor of about 600, ranging from less than 100 to over 1000. In any event, the concentration factor for normal U is very favorable and would be expected to apply to other isotopes of U from whatever source. We were pleased to see the Ra isotopes in low concentrations because their decay products, especially ^{214}Pb-^{214}Bi, have a large number of intense and energetic gamma rays that could seriously interfere with the detection of low levels of other radionuclides.

While the method used for calculating upper limits for undetected radionuclides has been described, a number of factors enter into the final result. It must be remembered that gamma spectrometry is simply the observation of the spontaneously emitted gamma rays from the sample. The only means of increasing sensitivity are to count large samples. to count the samples as close to the detector as possible (somewhat mutually exclusive), to count for as long as possible and to minimize the decay time between sample collection and analysis. The background in the energy region of interest is important and is composed mostly of Compton-scattered photons from higher energy gamma rays emitted by radionuclides in the sample. The Compton suppression circuitry in our counter reduces this background by a factor of 5 to 20, depending on energy. Other factors that tend to minimize the upper limit calculated for a particular nuclide are large sample weight, close-in geometry, short decay time, large branching intensity for the gamma ray sought,

and absence of other gamma rays at higher energy. It can be seen, then, that the upper limit for detection of a particular radionuclide varies from sample to sample and can be widely different from one radionuclide to the next. This explains, for example, why the upper limit for ^{7}Be is higher in the later samples. All numbers are decay-corrected to December 1, 1981, so the samples counted in February have smaller decay corrections than those analyzed in June.

It is interesting to note the low limits of detection for the nuclides ^{54}Mn and ^{65}Zn, and the implication these limits have on sensitivity. The detection limits are about 1 fCi/g, and we would like to have about ten times this for a good measurement, say, 10 fCi/g. Assuming a very conservative concentration factor of 10^3 for these two nuclides, we would be able to measure concentrations in sea water at least as low as 10 fCi/l (about 20 dpm/m^3), probably even lower.

Table 2 lists the results of our species identification work on the plankton. The method of pumping through the ship's fire mains and collecting for 24 h or more resulted in samples containing many broken pieces and in some cases only fragmented exoskeletons with no soft parts remaining, making identification difficult. At this stage of our experience, we are unable to say much about the influence of species composition on radionuclide content, and Table 2 is included principally to present the data. The samples generally consist mostly of copepods with varying amounts of foraminifera and lesser amounts of other organisms. It is interesting that only 13 samples contained 10 or more foraminifera, and 6 of the 7 samples containing either ^{95}Nb, ^{144}Ce, or both are included among these. While so few samples hardly represent a statistically significant number, possibly what we are seeing is the influence of the generally higher specific activity of smaller particles as reported by the UW groups. As data from future cruises accumulate, perhaps corrections will become more definite. A side benefit of the species analysis is that it alerts us to contamination by nonplanktonic material, such as sessile organisms or inorganic detritus. A summary of the seven samples containing ^{95}Nb and/or ^{144}Ce with a description of the principal biomass is given in Table 3.

Table 2.
Plankton species.[a]

Taxa	Sample No.	b 1	b 2	c 3	4	b,d 5	b,e 6	f 7	8	9	10	11	13	16	17	18
Crustacea																
Copepods																
Calanoids																
Calanus Sp.		341	4510	310	440	2780	3080	1030	400	82	248	276	36	799	713	128
C. Propinquus																
Candacia		60	60	30	--	290	30	30	--	--	--	4	10	1	--	--
Temora Sp.		--	60	--	--	4610	4760	5760	1860	--	--	--	76	--	--	--
Labidocera Sp.		--	--	80	--	190	--	30	--	--	--	--	--	--	--	--
Euchaeta Sp.		3	380	70	120	290	130	--	--	--	--	--	--	--	--	--
Metridia Sp.		--	--	--	--	--	30	--	--	--	16	--	72	1	--	--
Scolecithrix Sp.		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Pontella Sp.		--	--	--	--	--	--		--	--	--	--	--	--	--	--
Eucalanus		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Pleuromamma Sp.		1	120	60	400	540	--	--	1030	11	284	224	5	--	--	--
Centropages Sp.		60	60	40	40	470	90	120	--	--	--	--	--	--	--	--
Unident Large		--	60	170	--	420	--	700	--	--	--	--	37	--	4	--
Unident Small		1220	6300	--	40	--	290	--	--	--	--	--	--	1612	674	1496
Calanoides		--	--	--	--	--	--	--	--	--	--	--	2	--	--	--
Cyclopoids		3426	11850	1175o	190	2040	550	380	160	9	32	26	1	--	--	--
Copilia		--	--	--	--	--	--	30	--	--	--	--	--	--	--	--
Harpactocoids		540	1900	20	--	60	160	--	--	--	--	16	--	--	--	--
Gammerid Amphipod		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Amphipods		--	--	10	40	30	--	--	30	25	3	2	5	19	4	8
Caprellids		--	--	--	--	--	--	--	--	--	--	-	--	--	--	4
Euphausiid Pieces		--	--	--	--	15	--	1	10	12	3	18	--	--	--	--
Euphausiid Naupli		1	260	50	40	60	100	100	220	40	--	44	43	--	4	8
Euphausiid Calyptopis		--	--	--	--	--	--	--	--	--	--	--	11	--	6	--
Megalops		--	--	20	40	60	60	--	--	2	--	--	--	--	--	
Zoeas		--	--					--	--	--	--	--	--	--	--	--
Naupli		--	--	--	--	--	--	--	--	--	--	--	--	1	--	--
Ostracods		--	--	20	30	30	--	--	20	1	--	4	1	14	29	8
Barnacle naupli		60	--	--	30	30	--	--	--	--	--	--	--	--	--	--
Barnacle Cyprids		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Isopods		--	--	--	--	--	--	--	--	--	--	--	--	--	--	--

[a] Number of individuals per 25 ml.
[b] Artificial radionuclides detected in these samples.

Table 2 (continued).
Plankton species.

Taxa Sample No.	1	2	c 3	4	d 5	e 6	f 7	8	9	10	11	13	16	17	18
Radiolarians	--	1000	--	--	--	--	--	--	--	--	--	--	--	--	--
Foraminifera	1410	1900	--	20	190	160	160	--	1	--	10	--	--	19	376
Mollusca	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Gastropod Larvae	--	--	--	--	30	--	--	--	--	--	--	--	--	--	--
Pteropods	740	2400	40	90	770	60	60	60	1	12	20	--	--	--	--
Diacria Quadridentat	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Creseis Virgula	130	--	--	--	320	510	510	100	--	--	--	--	--	--	--
LINACINA INFLATA	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
L. Bulimoides	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
L. Helicina	--	--	--	--	--	--	--	--	--	--	--	--	48	59	416
Siphonophore	--	--	40	30	60	30	30	--	5	--	--	--	--	--	--
Chaetognaths	60	60	60	210	100	320	160	50	3	8	64	6	6	6	--
Salp	--	--	10	--	60	--	1340	20	--	--	--	1	--	--	--
Fish Eggs	350	450	180	880	19700	640	--	80	62	8	28	6	--	--	--
Various Larvae	690	2450	50	610	830	1372	1370	50	4	--	4	--	--	--	--
Centric Diatoms	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--

[c] Shrimp Lucifer Sp. - 10.

[d] Heteropods - 60; Cladocera - 220.

[e] Cladocera - 260.

[f] Cladocera - 260; Shrimp Lucifer Sp. - 30.

Table 2 (continued).
Plankton species.

Taxa Sample #	19 †	20 †	22	27	28	30	31	32	33	34	35	37	38	39 b
Crustacea														
Copepods														
Calanoids														
Calanus Sp.	12	--	--	1604	3	104	824	2888	928	380	108	4	2	--
C. Propinquus	--	--	--	--	1	--	--	--	--	--	--	600	--	--
Candacia	20	--	--	--	--	952	1464	144	80	44	500	--	--	3974
Temora Sp.	--	--	--	--	--	--	--	--	--	8	84	--	144	506
Labidocera Sp.	--	--	--	--	--	--	--	--	--	4	--	--	--	196
Euchaeta Sp.	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Metridia Sp.	--	4	--	--	4	256	--	1328	352	352	8	--	--	--
Scolecithrix Sp.	--	--	--	--	--	--	--	--	--	100	120	124	50	111
Pontella Sp.	--	--	--	--	--	--	--	--	--	--	--	132	--	2
Eucalanus	--	--	--	--	--	--	--	--	--	--	4	608	20	--
Pleuromamma Sp.	--	--	--	--	--	--	--	--	48	16	--	--	--	--
Centropages Sp.	--	--	--	--	--	--	--	1344	5152	220	8	448	2004	--
Unident Large	--	--	--	--	--	--	24	--	32	16	12	4	--	272
Unident Small	120	--	--	852	32	5488	368	15000	47800	12 6	2012	764	1140	5895
Calanoides	--	--	--	--	--	--	--	--	--	--	--	12	--	288
Cyclopoids	--	1	--	--	--	8	108	--	1112	152	56	584	34	3118
Copilia	--	--	--	--	--	--	--	--	--	4	4	4	--	1
Harpactocoids	--	--	--	--	--	--	--	--	16	4	--	--	2	44
Gammerid Amphipod	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Amphipods	--	--	4	52	5	8	20	232	--	4	4	16	--	184
Caprellids	20	3	--	--	--	8	8	--	--	--	--	--	--	--
Euphausiid Pieces	--	--	4	--	--	--	--	16	--	--	--	--	--	1
Euphausiid Nauplii	4	--	--	--	--	--	--	--	--	--	--	--	--	--
Euphausiid Calyptopis	--	--	--	--	--	--	,	8	24	152	--	24	--	--
Megalops	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Zoeas	--	--	--	--	--	--	--	16	16	148	12	--	4	17
Nauplii	--	--	--	--	--	16	--	32	--	8	36	12	2	--
Ostracods	--	--	--	16	--	24	12	32	--	12	4	--	--	33
Barnacle Nauplii	--	--	--	--	--	--	--	8	64	88	100	12	4	24
Barnacle Cyprids	--	--	--	--	--	--	--	24	24	4	4	4	2	--
Isopods	--	1	--	--	--	--	--	8	--	--	--	--	--	--

Table 2 (concluded).
Plankton species.

Taxa Sample #	19	20	22	27	28	30	31	32	33	34	35	37	38	39
Radiolarians	--	--	--	--	--	--	--	--	--	--	--	4	--	--
Foraminifera	16	--	--	--	--	--	--	8	112	24	4	--	--	539
Mollusca	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Pteropods	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Diacria Quadridentat	--	1	--	--	--	--	--	--	--	--	--	--	--	--
Creseis Virgula	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Linacina Inflata	--	1	2	--	--	--	--	24	16	--	--	--	--	296
L. Bulimoides	--	--	--	--	--	--	--	--	--	--	--	--	--	--
L. Helicina	4	5	--	--	--	--	--	24	--	--	--	--	--	--
Siphonophore	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Chaetognaths	--	--	--	4	--	--	56	8	32	100	64	52	4	257
Salp	--	--	2	--	--	--	--	--	--	--	--	--	--	--
Fish Eggs	--	--	--	--	--	--	--	968	312	408	76	204	84	75
Various Larvae	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Centric Diatoms	--	--	--	--	--	--	12	256	--	32	12	--	--	--

Table 3.
Summary of biomass associated with artificial radionuclides.

Sample	Radionuclides	Description of Principal Biomass[a]
1	^{95}Nb - ^{144}Ce	Cyclopoids, algae, shell fragments, rust
2	^{95}Nb - ^{144}Ce	Approximately equal numbers of calanoids and cyclopoids. Only sample with substantial number of radiolarians
5	^{144}Ce	Only sample with very large fraction of fish eggs
6	^{95}Nb	Other than copepods, has large contribution from larvaceans
19	^{95}Nb - ^{144}Ce	Mostly unidentified small calanoids, caprellids
20	^{95}Nb - ^{144}Ce	About equal fraction of calanoids, caprellids and pteropods
39	^{95}Nb	Substantial number of forams; rest mostly small calanoids and cyclopoids

[a] All samples contained forams and copepods except No. 20, which had no forams.

RESUSPENSION

The ocean is known to be a major source of atmospheric particulate matter. There is considerable evidence, however, that the chemical composition of the particles in the marine aerosol is often considerably different from that of seawater. Barker and Zeitlin[35] found enrichment factors for transition metals in the aerosol approaching and exceeding three and four orders of magnitude relative to sodium. Cattell and Scott[36] suggest that a biogenic agent may be responsible for the approximately 20,000-fold enrichment of copper during aerosol production in the ocean. The whole question of fractionation at the sea surface was the subject of a 1976 review article.[37] It seems possible, even likely, that the correlation we observe between radionuclides in plankton and in the air samples is due, at least in part, to resuspension. In general, we detect those, and only those, radionuclides in the air filters that are detected in the plankton at the same approximate location. Even the peculiarity of detecting only the ^{95}Nb but not its parent, ^{95}Zr, is reproduced in the air filter data. While the air filter inlet was mounted as high as practical on the ship, its elevation above water level was only about 80 feet. Cattell and Scott[36] present evidence that, at least for copper, the influence of the marine aerosol extends upward to 2000 m. If this is true, and given the sea surface conditions usually encountered in the Southern Ocean, it seems likely that most of the atmospheric particulate material we sampled was produced by resuspension. The air filters, however, often do show radioactivity not found in the plankton. This is probably because the air filter can collect the particles from a very large volume of air without reducing the gamma detection efficiency. If the smaller particles, 0.4 μm, or less are also collected, evidence suggests these may have very high enrichment factors. It seems likely that low-altitude air filter sampling could be a very sensitive technique for monitoring contamination in the sea as long as one realizes that it is principally the resuspended marine

aerosol that makes up the bulk of the sample, not primarily atmospheric constituents. The only published work we have found to date indicating concentration effects for radionuclides is that of Pattenden et al.[38] They present evidence that Pu isotopes, ^{241}Am and ^{137}Cs, are present in airborne material deposited in west Cumbria near Windscale and downwind from the Irish Sea. Most of the excess ^{137}Cs can be explained by contributions from Windscale discharges to air and material in sea water resuspended as spray. The excess actinides cannot be explained in terms of Windscale discharges to air and: "Some inconclusive evidence suggests that the excess actinides may come from a combination of (a) seaspray, raised by the wind from the sea surface which is (in some way) highly enriched with actinides (but not ^{137}Cs) compared to bulk seawater." Excesses of ^{106}Ru, ^{125}Sb, ^{134}Cs, and ^{144}Ce were also observed. It should be emphasized that these radionuclides are initially discharged directly into the the water.

SUMMARY AND CONCLUSIONS

We have begun an investigation into the potential utility of using marine plankton as indicators of low-level radioactive contamination in the Southern Ocean. A review of the literature indicates that marine and freshwater plankton, as well as other organisms, can have concentration factors relative to water for radionuclides and trace elements of up to 10^4. Most of the original work on the uptake and distribution of a variety of radionuclides in marine plankton was done by the University of Washington during the two atmospheric nuclear test programs in the Marshall Islands in 1956 and 1958. The work demonstrated that many radionuclides are rapidly assimilated by plankton with large concentration factors and retained within the biological strata over considerable time and distance. Other workers have confirmed these large concentration factors and measured additional ones for a variety of elements and organisms. Very little work has been done in the southern hemisphere, mainly a few studies in the region of Mururoa Atoll. In

addition to having high concentration factors, plankton is also easy to collect in international waters and simple to transport and analyze. Because of various uncontrollable factors, however, plankton is best used for detecting the presence of unusual radionuclides and measuring isotopic ratios.

Our cruise on the U.S.C.G.C. Glacier around the South Pacific Basin in 1981-82 resulted in the collection of 32 plankton samples along with numerous air filter, sea water, fallout and rain samples collected by Battelle Pacific Northwest Laboratory personnel. We were able to measure concentrations of ^{7}Be, ^{40}K and U and Th series nuclides in some or all samples. Concentration factors were within the ranges expected, and we found that ^{226}Ra and its decay products did not affect our ability to detect low levels of other radionuclides. The activity ratio of ^{238}U to ^{235}U in plankton was the same as in sea water within experimental error. We believe we detected low levels of ^{95}Nb and ^{144}Ce in seven samples from locations as far south as 68°. While there is a potential for misidentification (especially in the case of ^{95}Nb) because of interference from other radionuclides, the facts that these same nuclides were detected independently by Battelle in the air filters and sea water, and that they are known from previous work to have high concentration factors and to be associated in plankton, lead us to believe that they are real. We do not know the origin of these nuclides but note that they are high-yield fission products with good characteristics for high sensitivity of detection, and the levels we measured were very low. We calculated upper limit values for several radionuclides not detected, and these limits of a few fCi/g indicate that we could measure a concentration of the order of 10 fCi/l in sea water.

The close association of radionuclide content and concentration between the air filters and the plankton leads us to suspect that resuspension from the sea surface may strongly influence the aerosol composition. This effect has been reported in the literature for stable elements and suspected for radionuclides. Low-altitude air sampling could prove to be a very sensitive means of monitoring low levels of radionuclides in the sea.

Biological identification of the plankton indicates that there may be an association between the artificial radionuclides and the foraminifea content of the samples. With only seven samples containing artificial radionuclides, this is a tentative conclusion, but it could be the result of the generally high specific activity of the smaller plankton as reported by others.

FURTHER STUDIES

We have already participated in Operation Deepfreeze 1982, a cruise similar to that in 1981, and the results will be the subject of Part 2 of this report. We will continue to collect plankton, water, and air samples in the southern hemisphere, utilizing ships of opportunity. We intend to investigate the extent to which Mururoa is a source of radionuclides to the Southern Ocean. We also intend to pursue the idea that sampling the marine aerosol might be a very sensitive method for monitoring contamination in the sea.

We have been exploring the possible utility of other marine organisms with the Commonwealth Scientific Industrial Research Organization (CSIRO) Marine Laboratory in Perth, Australia, and have analyzed several samples of Ecklonia radiata, a subtidal kelp, for possible use as a bioconcentrator.

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REFERENCES

1. G. G. Polikarpov, Radioeklogiya Morskikh Organizmov (The Radioecology of Marine Organisms (Atomizdat, Moscow, 1964).

2. J. E. Portmann, Ed., Manual of Methods in Aquatic Environment Research, Part 2 - Guidelines for the Use of Biological Accumulators in Marine Pollution Monitoring, Food and Agricultural Organization of the United Nations, United Nations Environment Programme, FAO Fisheries Technical Paper No. 150 (1976).

3. J. J. Koranda and W. L. Robinson, "Accumulation of Radionuclides by Plants as a Monitor System," Environmental Health Perspectives 27, 165 (1978).

4. L. R. Donaldson, A. H. Seymour, E. E. Held, N. O. Hines, F. G. Lowman, P. R. Olson, A. D. Welander, Survey of Radioactivity in the Sea Near Bikini and Eniwetok Atolls, June 11-21, 1956, Applied Fisheries Laboratory, University of Washington, Seattle, Washington, UWFL-46 (1956).

5. J. H. Harley, Ed., Operation Troll, Health and Safety Laboratory, U. S. Atomic Energy Commission, New York Operations Office, NYO-4656 (1956.)

6. A. H. Seymour, E. E. Held, F. G. Lowman, J. R. Donaldson, D. J. South, Survey of Radioactivity in the Sea and in Pelagic Marine Life West of the Marshall Islands, September 1-20, 1956, Applied Fisheries Laboratory, University of Washington, Seattle, Washington, UWFL-47 (1957).

7. F. G. Lowman, Radionuclides in Plankton Near the Marshall Islands, 1956, Laboratory of Radiation Biology, University of Washington, Seattle, Washington, UWFL-54 (1958).

8. N. O. Hines, Proving Ground: An Account of the Radiobiological Studies in the Pacific, 1946-1961 (University of Washington Press, Seattle, 1962).

9. R. C. Bolles and N. E. Ballou, "Calculated Activities and Abundances of U^{235} Fission Products," Nuclear Science and Engineering 5 (3), 156 (1959).

10. A. D. Welander and R. F. Palumbo, Radionuclide Content of Oceanic Fish, Plankton, and Water in the Vicinity of Christmas Island, March through August, 1962, Laboratory of Radiation Biology, University of Washington, Seattle, Washington, UWFL-88 (1963).

11. W. Feldt, G. Kanish, R. Laver, "Radioactive Contamination of the NEA Dumping Sites," Proc. Symp. Impacts of Radionuclide Releases into the Marine Environment, Vienna, 1980 (IAEA-SM-248/111), pp. 465-480.

12. K. C. Pillai, R. C. Smith, T. R. Folsom, "Plutonium in the Marine Environment," Nature 203 (4945), 568 (1964).

13. V. V. Gromov, "Uptake of Plutonium and Other Nuclear Wastes by Plankton," Marine Science Communications 2 (3&4), 227 (1976).

14. V. S. Zlobin and O. V. Mokanu, "Mechanisms of the Accumulation of Plutonium-239 and Polonium-210 by the Brown Alga Ascophyllum Nodosum and Marine Phytoplankton," Voprosy Morsk, Biol. (Problems of Marine Biology) 12, 160 (1969).

15. S. W. Fowler and S. R. Aston, "Application of ^{235}Np in Experimental Aquatic Radioecology: Preliminary Observations on Neptunium Behaviour in Sea Water, Sediments and Zooplankton," Health Physics 42 (4), 515 (1982).

16. L. V. Shannon, The Alpha-Activity of Marine Plankton, Republic of South Africa Department of Industries, Division of Sea Fisheries Investigational Report No. 68 (1969).

17. A. S. Paschoa, G. B. Baptista, L. E. Urenn, and M. Eisenbud, "Dosimetry of Natural and Man-Made Alpha Emitters in Plankton," Proc. Symp. Impacts of Radionuclide Releases into the Marine Environment, loc. cit., pp. 695-716.

18. L. V. Sick and G. J. Baptist, "Cadmium Incorporation by the Marine Copepod Pseudodiaptomus coronatus," Limnol. Oceanogr. 24 (3), 453-462 (1979).

19. K. Knauss, T. Ku, "The Elemental Composition and Decay-Series Radionuclide Content of Plankton From the East Pacific," Chemical Geology 39, 125-145 (1983).

20. N. Fisher, "Bioaccumulation of Technetium by Marine Phytoplankton," Envirn. Sci. Technol. 16, 579-581 (1982).

21. T. M. Beasley, Biogeochemical Studies of Technetium in Marine and Estuarine Ecosystems, School of Oceanography, Oregon State University, Marine Science Center, Newport, Oregon. DOE/EV/10251-3 (1981).

22. A. M. Kolesnikova, G. V. Barinov, A. Ya. Zensenko, "Radioecological Investigation in the Region of Mururoa Atoll (Tuamotu Island), "Radiobiologiya 9 (1), 139 (1969).

23. C. D. Jennings, "Selective Uptake of ^{55}Fe from Seawater by Zooplankton," Mar. Sci. Comm. 4 (1), 49-58 (1978).

24. C. D. Jennings, "^{55}Fe in Pacific Ocean Plankton," Mar. Biol. 44, 223-226 (1977).

25. B. G. Blaylock, "Radionuclide Data Bases Available for Bioaccumulation Factors for Freshwater Biota," Nuclear Safety 23 (4), 427 (1982).

26. I. Aoyama, K. Inomo, Y. Inoue, "Uptake and Release of Some Radionuclides by Fresh Water Phytoplankton in Batch Culture," J. Radiat. Res. 17, 69-81 (1976).

27. D. P. Marciulioniene, R. F. Dusauskiene-Duz, and G. A. Cibiraite, "Radioecological Study of the Charophytes," Hydrobiological Journal 12 (1), 6-16 (1976).

28. T. A. Jenckes, The Feasibility of Using Phytoplankton in a Quantitative Method of Detecting Low Levels of Radionuclides, Ph.D. Thesis, Temple University, Philadelphia, PA (1975).

29. J. B. Millard, F. W. Whicker, O. D. Markham, "Gamma Emitting Radionuclides of the Test Reactor Area Leaching Ponds," Ecological Studies on the Idaho National Engineering Laboratory Site 1978 Progress Report, O. D. Markham, Editor, U. S. Department of Energy Idaho Operations Office Radiological and Environmental Sciences Laboratory, IDO-12087 (1978).

30. J. J. Davis, Accumulation of Radionuclides by Aquatic Insects, Biology Laboratory, Hanford Laboratories, General Electric Co., Richland, WA, HW-SA-2848 (1962).

31. D. C. Camp, C. Gatrousis, L. A. Maynard, "Low Background Ge(Li) Detector Systems for Radioenvironmental Studies," Nucl. Instrum. Methods 117, 189 (1974).

32. R. Gunnink and J. B. Niday, Computerized Quantitative Analysis by Gamma-Ray Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-51061, Vols. I-IV (1972).

33. R. W. Hoff, J. W. Meadows, H. D. Wilson, A. L. Prindle, R. Gunnink, K. O. Hamby, "Analytical Program," Enewetak Radiological Survey, W. E. Nervik and R. Ray, Eds., USAEC Nevada Operations Office, Las Vegas, NV, NVO-140, Vol. I (1973), pp. 426-485.

34. F. G. Lowman, T. R. Rice, F. A. Richards, "Accumulation and Redistribution of Radionuclides by Marine Organisms," Radioactivity in the Marine Environment (National Academy of Sciences, Washington, D. C., 1971), p. 173.

35. D. R. Barker and H. Zeitlin, "Metal-Ion Concentrations in Sea-Surface Microlayer and Size-Separated Atmospheric Aerosol Samples in Hawaii," J. Geophys. Res. 77 (27), 5076 (1972).

36. F. C. R. Cattell and W. D. Scott, "Copper in Aerosol Particles Produced by the Ocean," Science 22, 429 (1978).

37. R. A. Duce and E. J. Hoffman, "Chemical Fractionation at the Air/Sea Interface," Ann. Rev. Earth and Planet Sci. 4, F. A. Donath, F. G. Stehli and G. W. Wetherill, Eds. (Annual Reviews, Inc., Palo Alto, CA, 1976) pp. 187-228.

38. N. J. Pattenden, R. S. Cambray, K. Playford, J. D. Eakins, E. M. R. Fisher, "Atmospheric Measurements on Radionuclides Previously Discharged to the Sea," Proc. Symp. Impacts of Radionuclide Releases into the Marine Environment, loc. cit., pp. 695-716.