Reports

Radioactivity of the Columbia River Effluent

Abstract. *Chromium-51 and zinc-65 were detected as far as 115 and 15 kilometers, respectively, from the mouth of the Columbia River; zinc-65 also was found at a few isolated stations. Zirconium-95 and niobium-95 from atmospheric fallout occurred in the river effluent but they were most abundant in surface waters further offshore. Distribution of the radionuclides is controlled largely by surface currents and by upwelling of sea water near the coast.*

Surface sea waters near the Washington-Oregon coast contain radionuclides derived from the reactors at Hanford, Washington (*1, 2*), making possible the study of the dispersal of Columbia River water at sea and the behavior of various radionuclides in the marine environment. Radionuclides from the Columbia River are distinctly different from those naturally present in sea water or in marine sediments (*3*) or from fission products derived from worldwide atmospheric fallout (*4*).

During *Brown Bear* cruise 331, 13–24 August 1963, radioactivity of surface sea water near the mouth of the Columbia River was measured *in situ* at stations where the salinity, temperature, and dissolved oxygen content were determined from samples of surface water. Gamma-ray spectra (Fig. 1) of the surface waters (approximately 3 m deep) were obtained with an underwater scintillation detector (*5*) consisting of a 12.5-cm by 15-cm crystal of NaI (Tl) optically coupled by a lucite light-pipe to a photomultiplier tube. The instrument, enclosed in a steel pressure housing, was lowered by an armored, single-conductor, coaxial cable which connected the detector system to the high-voltage supply, a multichannel spectrum analyzer, and other electronic equipment aboard ship. To prevent contamination by absorption of radioactive materials, the outer surface of the pressure housing was cleaned with solvent and coated with waterproof grease at each station immediately before the probe was lowered; the grease had no detectable gamma-ray activity.

Unfortunately, radiochemical analyses of sea-water samples taken during the cruise failed; thus identification of radionuclides in the waters is based on the photopeak energies in the spectra. Because the sources of the radioactivity are known to be the Columbia River, worldwide fallout, and naturally occurring radionuclides, our identification of the radionuclides was greatly simplified (*6*).

The Columbia River discharge forms a plume of surface water with salinity intermediate between the negligible salt concentration of the river water and that of the ambient sea surface, which is variable but generally slightly greater than 32.5 per mil. The salinity of the surface waters increases with distance from the river mouth along the axis of the plume and laterally away from the axis. Most of the low-salinity water from the river remains within 40 m of the surface. The effluent, with salinity less than 32.5 per mil, moves generally southwestward during the summer months and may extend south of 40°N during September and October, the period of its maximum detectable spread (*7*).

The core of the plume during August 1963 is outlined by the 31-per-mil isohaline (Fig. 2). Surface waters of variable salinity, generally less than 30 per mil, occurred within approximately 60 km of the river mouth where the water from the river and sea initially mix (*7*). Upwelled water, recognized by high salinities and low concentrations of dissolved oxygen, was present at or near the surface just south of the river mouth and in the inshore area extending southward from Tillamook Head.

Chromium-51 (half-life, 28 days) from the Columbia River was detected in surface sea water over a large area (Fig. 2), with the highest activities in the river estuary. Within 15 to 40 km of the river mouth the activity of Cr^{51} exceeded that of the naturally occurring potassium-40 in sea water. Chromium-51 was detectable in a band, generally parallel with the coast, up to 115 km south of the river mouth but only 25 km north and west of the mouth.

The concentration of Cr^{51} in the surface sea water decreases rapidly with distance from the mouth of the river mouth because of the mixing of river water with uncontaminated sea water, by radioactive decay of Cr^{51}, and by any other process(es) that remove Cr^{51} from the surface layers, such as sedi-

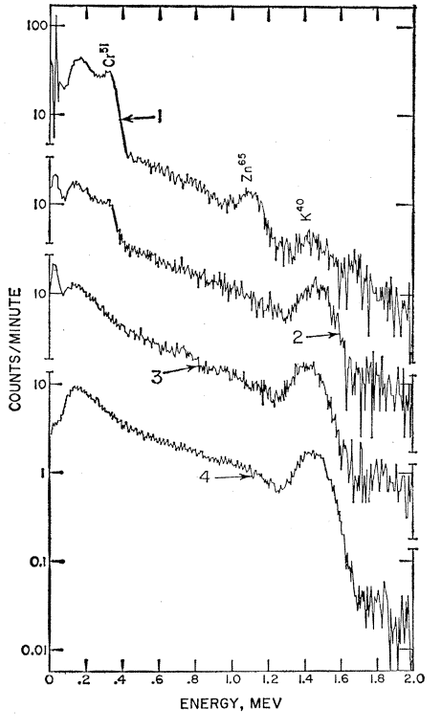

Fig. 1. Typical gamma-ray spectra obtained by the detector system. *1*, Columbia River water, Astoria, Oregon, 14 August 1963. *2*, Sea water; depth 3 m; salinity, 22.35 per mil; 24 August 1963. *3*, Sea water; depth, 3 m; salinity, 30.56 per mil; 20 August 1963. *4*, Sea water; depth, 190 m; salinity, 33.95 per mil; 13 August 1963.

mentation of any particulate matter. South of Tillamook Head, the absence of Cr^{51} is due largely to upwelling, which keeps Columbia River water away from the coast.

Concentrations of zinc-65 (half-life, 245 days) were highest in the Columbia River estuary near Astoria, Oregon, and in surface sea water within approximately 15 km of the river mouth. It appears that Zn^{65} is rapidly removed from surface sea waters or diluted to such low concentrations that it cannot be used to trace the movement of river effluent. Zinc-65 is detectable in planktonic marine organisms near the Columbia River (*2*). Three isolated occurrences of Zn^{65} detected during this survey may be due in part to marine organisms. Two of the three stations were occupied in the predawn hours when planktonic organisms are commonly near the surface.

Zirconium-95 and niobium-95 (half-life, 65 days) were ubiquitous in offshore surface waters but greatly reduced in concentration in surface waters within 30 to 60 km of the coast, except near the mouth of the Columbia River (Fig. 3); no other fission products were detected.

The distribution of these radionuclides is partly controlled by combined effects of the ocean currents near the coast and of the winds which cause the surface waters to move generally southwestward in late summer (*7*). The low-salinity, less-dense water formed by the effluent of the Columbia River tends to spread over the ambient waters in the coastal and offshore regions. Fronts characterized by surface confluence and sinking are particularly well defined at the boundary between the plume of river water and the upwelled waters near the Oregon coast where the upwelling is more or less continuous.

The upwelled water brought to the surface was previously offshore at depths of 100 to 200 m and shielded from recent atmospheric fallout. Consequently, upwelled coastal waters can be expected to have low concentrations of fallout nuclides with relatively short half-lives, such as Zr^{95} and Nb^{95}. In contrast, surface waters seaward of the plume remain in the surface, wind-mixed, layer during the warming part of the annual cycle, retaining the fallout nuclides above the seasonal pycnocline. Fallout radionuclides brought into the ocean with Columbia River water probably remain in the less-dense, less-saline waters of the plume and also contribute to the general level of Zr^{95} and Nb^{95} activity in the surface waters.

These studies confirm the surface current patterns determined by standard oceanographic techniques (*7*) and demonstrate that where suitable radio-

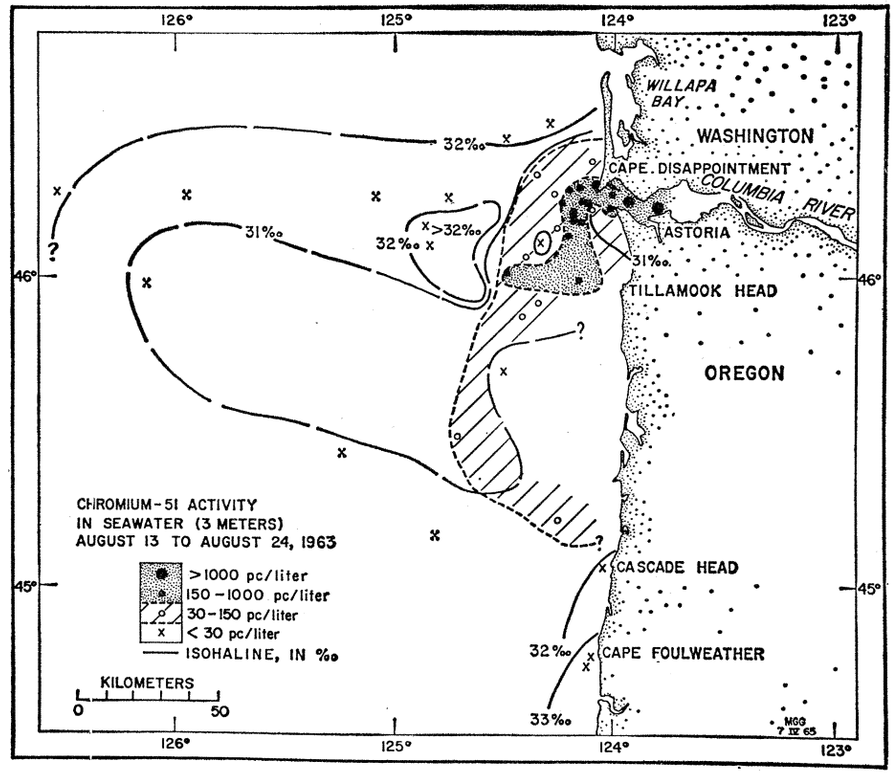

Fig. 2. Distribution of chromium-51 in sea water at a depth of 3 m.

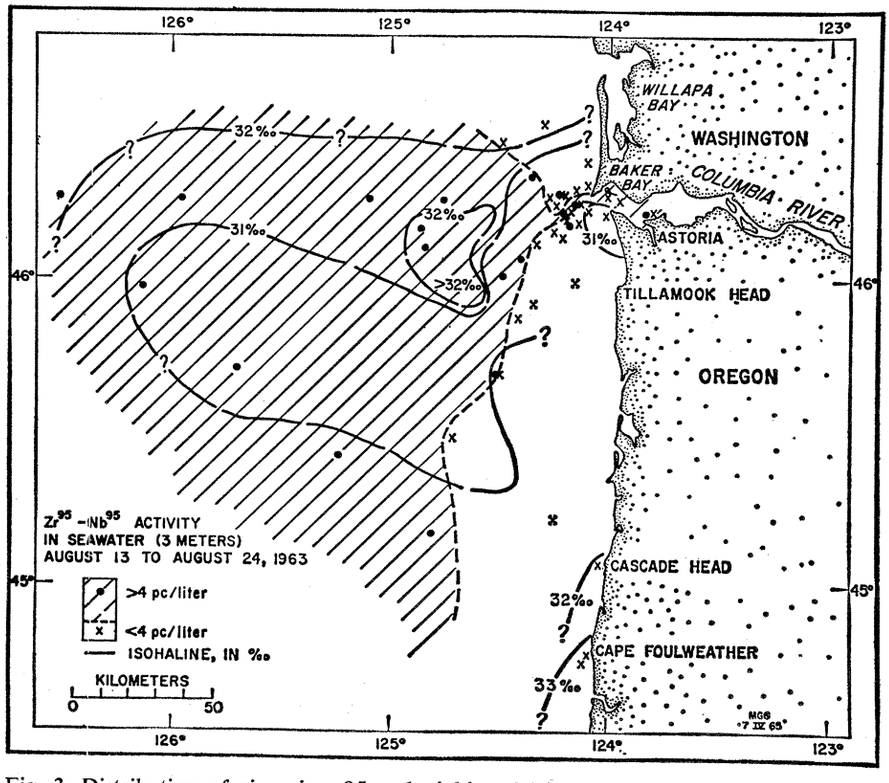

Fig. 3. Distribution of zirconium-95 and niobium-95 in sea water at a depth of 3 m.

nuclides are introduced into the ocean in large quantities they may be used to study local water circulation. Use of an *in situ* detector greatly simplifies the procedure and many more analyses can be made than would be possible if extensive radiochemical separations were required.

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References and Notes

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6. The data were analyzed by a spectrum-stripping technique in which the total number of counts in the photopeak of each radionuclide was corrected by subtracting the counts due to Compton scattering associated with photopeaks of higher energy. The concentration of each radionuclide was calculated after determining the efficiency of the detector by measuring its response in 10^5 liters of various solutions containing known concentrations of Cr^{51}, Cs^{137}, La^{140}, and K^{40}. The efficiency for Zr^{95}-Nb^{95} and Zn^{65} was estimated by interpolation on a plot of detector efficiency as a function of the photofraction of the crystal, divided by the square of the mean free path in water of the gamma photon. The data, computed as picocuries per liter for each radionuclide, are considered significant if the activity was greater than two standard deviations estimated from the counting statistics. Zirconium-95 and Nb^{95} were ubiquitous in the surface waters, so spectra obtained in the upwelled coastal waters were arbitrarily taken as background. Subsequent analysis of the spectra used to obtain the correction for the Compton effect indicates that these waters contained Zr^{95}-Nb^{95} at 3 to 4 pc/liter.
7. T. F. Budinger, L. K. Coachman, C. A. Barnes, "Columbia River effluent in the northeast Pacific Ocean, 1961, 1962: Selected aspects of physical oceanography," Dept. of Oceanography, Univ. of Washington, Seattle, Tech. Rept. 99 (February 1964).
8. We thank A. H. Seymour and D. E. Engstom, Laboratory of Radiation Biology, and R. Pedrick, U.S. Naval Oceanographic Office, for assistance. Dick Duffy, Department of Nuclear Engineering, University of Maryland, assisted in the design and construction of the *in situ* gamma-ray detector. Studies conducted in a cooperative program with the Laboratory of Radiation Biology, University of Washington. Supported by AEC contract AT(45-1)-1725 and by ONR contracts Nonr-477(10) and 477(37), project NR 083 012. Contribution No. 344, Department of Oceanography, University of Washington.

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Immunoglobulin Structure: Partial Amino Acid Sequence of a Bence Jones Protein

Abstract. *Sequence analysis and ordering of the soluble tryptic peptides of one Bence Jones protein and comparison with partial sequence data for another have revealed many structural differences in the half of the molecule with the terminal amino group, but only one structural difference in the half of the molecule having the terminal carboxyl group. Somatic chromosomal rearrangements may effect such changes and account for variability in antibody structure.*

Because Bence Jones proteins are the L-chains of the myeloma globulin from the same patient and are related to normal L-chains (*1*), analysis of the amino acid sequence of Bence Jones proteins facilitates study of the structure of normal human immunoglobulins (*2, 3*). There are two wholly different types of L-chains that correspond to the two antigenic types of Bence Jones proteins (type I and type II); these differentiate each of the structure of normal human immunoglobulins (γG, γA, and γM) into the two corresponding antigenic types. The two types of L-chains differ in terminal amino groups, peptide maps, and composition of their tryptic peptides (*4*); hence, they differ greatly in primary structure. Furthermore, within each antigenic type the Bence Jones proteins of individual patients differ in primary structure in multiple positions rather than in just one, as in the abnormal hemoglobins.

From comparison of peptide maps and partial sequence studies (*3*), we have concluded that the NH_2-terminal portion of type I Bence Jones proteins is subject to variation whereas the COOH-terminal octapeptide is an invariant part of the structure of type I L-chains. We now report the amino acid sequence (*5*) of about three-fourths of an individual's type I Bence Jones protein (specimen Ag) including the consecutive sequence of 118 residues in the COOH-terminal half of the molecule. Figure 1 presents a comparison of our data with the partial amino acid sequence of another individual's type I Bence Jones protein (specimen Roy) and with the tryptic peptide composition of a third specimen (Cu), both of which have recently been reported (*6*). From these data we deduce that Bence Jones proteins—and presumably normal L-chains—differ individually in primary structure in a manner heretofore unknown for proteins with analogous function in the same species. With the exception of a minor homologous interchange, the COOH-terminal portion of the molecule is apparently identical in the two specimens (Ag and Roy) for which partial sequence is known and is probably the same for the third protein (Cu) for which only the order and composition of the tryptic peptides is known. On the other hand, the NH_2-terminal portion has many replacements involving the exchange of both homologous and nonhomologous amino acids. Although most of these interchanges appear randomly distributed, some are clustered in a tetrapeptide sequence (designated the "switch peptide") that is located just before the exact middle of the polypeptide chain where the sequence becomes almost invariant.

The procedures for the isolation of the tryptic peptides of the type I Bence Jones protein Ag and for their sequence determination have been described (*3*). The sequence of the larger tryptic peptides was established by comparison of the fragments resulting from peptic and chymotryptic digestion of these peptides. The ordering of the tryptic peptides is based on two kinds of evidence: (i) on the overlaps we have established independently from the amino acid composition of peptides obtained from a separate chymotryptic digest of the intact carboxymethylated protein, and (ii) on the deductions drawn by Hilschmann and Craig (*6*) who compared their results with our data (*2*). Furthermore, just as they have used identity or similarity in the amino acid composition to order our tryptic peptides relative to theirs, we have tentatively assigned amino acids whose position was not determined by one laboratory to the position determined by the other. If the sequence of a peptide is not known in any of the three proteins, the amino acids comprising it are written in the order of their elution from the column of the amino acid analyzer. For comparative purposes the polypeptide chain is assumed to contain 212 amino acids, and the numbering system proposed by Hilschmann and Craig is adopted. In this way we have constructed a chart comparing the tentative sequence of three Bence Jones proteins of type I.