CONF-720708--6

RECEIVED BY TIC JUL 26 1972

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OCEANIC DISTRIBUTIONS AND RELATIONSHIPS OF ^7Be
AND FISSION PRODUCTS

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ABSTRACT

The vertical and horizontal distribution of ^7Be in the world's oceans provides a potential basis for estimation of the distributions of other conservative fallout radionuclides as a function of time and space. Such information is essential to any study of biogeochemical processes occurring within the marine biosphere. Surface air concentrations of ^7Be , a cosmogenic radionuclide, vary seasonally with the northern hemisphere maximum occurring during June and the minimum in December, however the concentrations are uniform from year to year. If the transfer of ^7Be across the air-sea interface is proportional to its air concentration, then ^7Be in surface water should parallel the seasonal atmospheric fluctuation, and the water column would reflect the transfer during the preceding months. The ratio of the surface concentration of ^7Be to its water column inventory, for twenty-one northern hemispheric profiles, did indeed exhibit a strong seasonal change which was independent of latitude, longitude, and year of sampling. Highest ratios were observed during July and August and the lowest during the early spring months, in accordance with the pattern that would be predicted from the variation in air concentration. It is estimated that the ^7Be water column inventory can be determined within $\pm 10\%$ at the 95% confidence limits from a single measurement of the surface water concentration, and the annual deposition of ^7Be be approximated within $\pm 20\%$.

Oceanic deposition of a given fission product radionuclide should be directly related to the proportionality between its measured air concentration and that of ^7Be . Thus, the cumulative input of fission products with half lives comparable to ^7Be could be determined by applying the time integrated ratios of fission product to ^7Be air concentration to the measured concentration of ^7Be in surface sea-water; this would require appropriate corrections for relative decay rates and vertical mixing within the water column. A more complicated system occurs in the case of longer lived fission products because advective water mass transport becomes an important factor in the oceanic distributions.

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Eventually, air concentration ratios of fission products and ^7Be , together with surface water concentrations of ^7Be would be employed in a dynamic model of ocean mixing and circulation to predict the temporal and spatial distributions of long-lived radionuclides in the north Pacific Ocean. Once completed and verified, this model should provide a useful tool for predicting future oceanic distributions of radioactivity from nuclear weapons testing.

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INTRODUCTION

Over the past several decades, ^7Be and fission products from nuclear weapons testing has provided a useful and diverse group of radioactive tracers that have found application in describing elemental transfer through the marine biosphere, geochemical processes and oceanic circulation and mixing rates. The wide variation of physical half-lives of this group of tracers, <100 days for ^7Be and $^{95}\text{ZrNb}$, ~1 year for ^{106}Ru and ^{144}Ce , and 30 years for ^{90}Sr and ^{137}Cs , permit their collective application toward description and analyses of processes occurring over widely different time scales. Evaluation of the interaction of radionuclides with marine biota requires intimate knowledge of the concentration of radionuclide in the water to which the organism was exposed. This knowledge is quite readily obtained in those studies of contained systems, such as the Humboldt Bay work presented by Heft and his co-workers.^[1] In open ocean studies, evaluation of the radionuclide concentrations and hence exposure to organisms becomes much more complicated. For example, due to the migratory nature of salmon, which range from feeding grounds in the Gulf of Alaska to their spawning areas, they are exposed to waters containing changing concentrations of radionuclides. Another example of varied exposure is represented by certain species of zooplankton which migrate vertically within the water column. In both these instances it would be virtually impossible to obtain a sample of water that would contain the radionuclide spectra representative of the integrated exposure of the organism. We are continually gaining better definition of the migratory patterns of the various organisms and their duration of existence within a given water mass; thus, if a means were available to estimate the radionuclide concentrations within these water masses, more meaningful interpretation of patterns and rates of radionuclide transfer through the marine food web and geochemical processes would be possible.

SAMPLING AND ANALYTICAL METHODS

All samples for this study were processed through Battelle large volume water samples, [2] units which first pass the water through parallel filters and then through a bed of aluminum oxide. The glass filters employed as the filtering medium have been shown to collect particulate radionuclides with essentially the same efficiency as a 0.3 μ pore size membrane filter. At the standard sampling flow rate of 35 l/min, the 6.4 mm thick alumina adsorption bed removes 70%, 50%, 18% and 50% of the nonfilterable ^7Be , $^{95}\text{ZrNb}$, ^{106}Ru and ^{144}Ce , respectively, as determined by laboratory tests verified by in-situ separations at sea. The sampling system employed a bundle of five one-inch diameter polyethylene hoses lowered from the hose reel, each terminating at a discreet depth, the maximum being 100 meters. Water was drawn simultaneously from each line with deck mounted centrifugal pumps, passed through a sampling unit and finally discharged through a water meter to ascertain the total volume processed. After passage of 2000-4000 liters of water, the units were disassembled, and the filters and alumina reserved for nondestructive radiochemical analysis by counting on large crystal [NaI(Tl)] anticoincidence shielded multidimensional gamma-ray spectrometers. [3] These instruments characteristically have a very low background, reduced Compton interference, good counting efficiency, and by coincident gamma-ray analysis provide high sensitivity for the direct measurement of many minor radioactive constituents. The disintegration rate of each radionuclide was calculated from their counting rate by computer solution of a series of simultaneous equations. These equations employ Compton correction factors and absolute counting efficiencies predetermined from a standardized source counted in identical geometry; in addition, corrections for sample volume, collection efficiency and decay from time of collection to date of analysis are made.

Radionuclide concentrations measured in samples taken in three vertical profiles at 36°N, 68°W are given in Table I. The recorded plus or minus values represent the 1 σ statistical variation due only to the counting measurements; however, the total measurement uncertainty is $\pm 7\%$ at the 1 σ level, as determined from analyses of a series of ocean samples which were processed and analyzed concurrently. [2]

VERTICAL RADIONUCLIDE TRANSPORT

The vertical distribution of individual radionuclides is typified in Figure 1, where the concentrations of ^7Be from the three profiles are plotted logarithmically as a function of depth. Mixing is essentially complete within the isothermal surface layer, is impeded by the barrier presented at the top of the thermocline, and the concentration then decreases exponentially with depth within the thermocline. From this depletion gradient, estimates of the vertical eddy diffusion coefficient can be made. Assuming boundary conditions of constant input, conservative behavior of the radionuclide, and the absence of horizontal advective transport, the radionuclide concentration as a function of depth is described as:

$$C/C_0 = e^{-\left(\frac{\lambda}{K_z}\right)^{1/2} \cdot Z},$$

where: C and C_0 are the concentrations at two depths
 Z is the depth interval
 λ is the radioactive decay constant, and
 K_z is the vertical eddy diffusion coefficient.

Values for K_z computed from the concentrations of ^7Be and $^{95}\text{ZrNb}$ in the depth interval between 40 and 100 meters, were 1.3 and 1.2 cm^2/sec , respectively. These values are in comparable agreement with Broeckers[4] north Atlantic values, but higher than the diffusion coefficients reported by Young and Silker[5] in water east of Barbados. Vertical eddy diffusion coefficients based on the distributions of the longer-lived ^{106}Ru and ^{144}Ce in the same depth interval were 0.3 and 0.6 cm^2/sec . The discrepancy between the K_z values for long and short half-life radionuclides is strong indication that one or more of the boundary conditions were not met. Biological or particulate settling is not considered a significant factor. Less than 5-10% of the measured radioactivity was found in the insoluble fraction and the ratio of soluble to insoluble was the same throughout the water column. It has been shown in other studies[6] that this ratio is maintained to the extinction depth for ^7Be and fission products; thus, in these instances the particulates are apparently transported vertically at a rate comparable to the soluble species. The differences in K_z must, therefore, require either a higher surface concentration or a lowered apparent depth concentration of the long-lived fallout isotopes, both of which are reasonable assumptions. First, the absolute input of atmospheric radionuclides is cyclic, with maximum transport across the air-sea interface occurring during June, as will be shown later. From air concentration data of Thomas, et al,[7] it appears entirely possible that sufficiently high surface concentrations of ^{106}Ru and ^{144}Ce could exist to account for the low calculated diffusion coefficients. If this is in fact the case, then the diffusion coefficients determined from the ^7Be and $^{95}\text{ZrNb}$ distributions should also be lower, but the magnitude of error should be quite small. On the other hand,

water at 100 meter depths could have received its initial radionuclide burden in an area far removed from, and transported by ocean currents to the point of measurement. Assuming a current of 0.1 kt, and a mixing velocity of 100 m/year, the material at depth could have been deposited nearly 1500 km from the point of measurement. Back trajectory to a lower latitude could be assumed, placing the point of initial atmospheric exposure of the deeper water in a region of reduced fallout. It is thus appropriate to consider both of these parameters in the interpretation of vertical radionuclide distributions, and recognize that the input function may be critical in the case of intermediate-lived radionuclides, while advective transport will be the major means of influencing the vertical distribution of long-lived species within any oceanic water column.

OCEANIC RADIONUCLIDE DISTRIBUTIONS

If it is assumed that transfer of a radionuclide across the air-sea interface is proportional to its atmospheric concentration, then its concentration in surface water should closely parallel the seasonal atmospheric fluctuations, and the water column inventory of a short-lived nuclide would be a function of the cumulative transfer during recent prior periods. The ground level air concentrations of ^7Be and ^{137}Cs at Richland, Washington are shown in Figure 2. The cyclic variation of each radionuclide is obvious with concentration maxima occurring during June, reflecting rapid transfer of stratospheric material through the tropopause discontinuity. The concentration of ^{137}Cs decreased annually with the stratospheric half-life of about 11 months from 1963, the first year of the nuclear testing moratorium, until 1967-8 when the French and Chinese started testing high yield devices. Annual beryllium-7 concentration variations on the other hand are essentially repeated from year to year, with a peak to valley ratio of 2.5-3. Thus, if transfer across the air-sea interface is proportional to the air concentration, both the surface concentration and, to a lesser degree, total quantity of ^7Be contained by a given water column should also show a similar repetitive annual variation from year to year.

Data from twenty-two depth profiles sampled from 1968-1971 between 8° and 45°N latitude in both the Atlantic and Pacific oceans are presented in Figure 3 as the variation with time of year of the ratio of the surface concentration of ^7Be to its water column inventory. A strong seasonal change of this ratio is apparent. Highest values occur in July and August following the period of highest air concentration, and hence the highest input, when the water column inventory is at the maximum. Low ratios during early spring reflect the combination of both minimum airborne radionuclide concentration and the larger ^7Be ocean inventory accumulated during higher input periods. The average deviation of individual data points from the curve is $\pm 10\%$ at the 95% confidence limits; thus, the water column inventory or integrated input of ^7Be at a given location can be predicted within these limits by measurement of the surface water concentration at that

location. For example, the ratio of surface concentration to inventory in August is about 0.019. Applying this value to the surface concentration given in Table 1 predicts respective inventories of about 47,000, 43,000 and 38,000 dpm/m², while values of 48,000, 41,000 and 46,000 dpm/m² were obtained by integration of the depth distribution curves. In the case of ⁷Be, with its relatively short half-life, steady state is approached so that its water column inventory is equal to the transfer rate of ⁷Be across the air-sea interface. It is therefore possible, with knowledge of the seasonal change of the ratio of surface concentration to inventory, to predict the rate of ⁷Be addition for other times of the year and also to predict the rate of addition of other radionuclides which will be proportional to their atmospheric concentrations relative to ⁷Be.

Variability of oceanic deposition of atmospheric radionuclides is dramatically illustrated by the surface water concentrations of ⁷Be in three north-south transects of the Pacific Ocean shown in Figure 4. The distribution of ⁷Be in samples collected along track B in midocean is in line with those reported for ¹³⁷Cs and ⁹⁰Sr both in Pacific Ocean surface water and on land surfaces which increase to midlatitude maxima.[8,9,10] Samples collected along the eastern boundary three years apart, but at the same time of year, show remarkable consistency in their latitudinal ⁷Be distribution, lending strong additional support to the hypothesis that air-sea transfer of this radionuclide should be the same from one year to the next. Of particular interest are the very high concentrations found at about 10°N latitude along these two lines, and the minor maximum at the same latitude in the mid-ocean track. These maxima unobserved in the case of the long-lived fission products, whose distribution reflects both the integrated long-term deposition and the subsequent transport, describe the recent transfer of atmospheric aerosols to the sea surface, probably by wet deposition. Total annual rainfall, both along the 160th meridian[11] and at Pacific coast stations,[12] exhibits a pronounced maximum between 5-10°N, decreasing to minima at both the equator and midlatitudes. Weather satellite data[13,14] reveal persistent and highly developed cloud cells over the eastern Pacific about 8°N at the intertropical convergence zone and decreasing cloudiness both north and south. The atmospheric aerosols are quite probably removed more completely by the increased precipitation in this region. Other possibilities are that the top of the clouds penetrate the tropopause and scavenge material from the stratospheric reservoir of high radionuclide concentration or as suggested by Dmitrieva et al,[15] parcels of more radioactive air from more northern latitudes intrude into the tropical zones.

Regardless of the mechanism involved, the important fact to be considered is that certain areas of the tropical ocean receive much higher quantities of fallout, at least at certain times of the year, than had previously been recognized. This phenomenon quite likely is isolated to rather limited geographical areas.

Previous data have shown that at higher latitudes in the open ocean uniform concentrations of ^7Be occur within narrow latitude belts across a rather wide range of longitude. [16,17] Latitudinal uniformity of seawater concentrations would be predicted if the atmospheric concentrations of ^7Be were independent of longitude. This, in fact, should be expected because of two reasons: first, the stratospheric ^7Be is injected primarily through the tropopause discontinuity, which circumscribes the earth, and then diffuses both north and south; and second, the tropospheric ^7Be production rate is latitudinally constant. [18] Thus, based solely on source function, the average air concentrations of ^7Be should be the same at all points along a given latitude. Similarly, air concentrations of the fission products of stratospheric origin should also exhibit latitudinal uniformity; although unlike ^7Be , their absolute concentration is governed by the changing stratospheric reservoir. The ratios of ground level air concentrations of ^7Be and fission products should thus also be uniform at a given latitude, and the quantities transferred across the air-sea interface should be in direct proportion to this ratio. Therefore, by measurement of the ^7Be in surface water, and knowledge of ratios of ^7Be and fission products in the overlying air, the deposition of a fission product on the sea surface can be estimated and historical data will provide the source function for the transport model so that hopefully the oceanic radionuclide distributions as a function of time since their first injection will be available.

TRANSPORT MODEL

The general equation describing the distribution of a property in the sea [19] is:

$$\frac{\delta s}{\delta t} = \frac{\delta}{\delta x} K_x \frac{\delta s}{\delta x} + \frac{\delta}{\delta y} K_y \frac{\delta s}{\delta y} + \frac{\delta}{\delta z} K_z \frac{\delta s}{\delta z} - \left(V_x \frac{\delta s}{\delta x} + V_y \frac{\delta s}{\delta y} + V_z \frac{\delta s}{\delta z} \right) - R \quad (\text{Equation 1})$$

where s is a scalar quantity--e.g., temperature, pressure or concentration; V_x , etc, and K_x , etc, are the spatial components of velocity and the diffusion coefficient respectively; and R is the sink distribution (sources are thereby implied). In words Equation 1 states that the local time change of s equals the effects of diffusion minus advection minus the local sinks. In principle a unique solution for Equation 1 exists if (1) the initial scalar field of s in a bounded region is specified; (2) the temporal variation of the scalar fields-- V_x , V_y , V_z , K_x , K_y , K_z , and R --in the region is known; and (3) the boundary conditions and their time dependence are given. The solution results in a description of s within this region during the same time period stated in conditions (2) and (3).

Previous workers [20,21,22] have used simplified forms of Equation 1 to fit observed oceanic profiles of oxygen, phosphorus, salinity, alkalinity and the carbon isotopes. By looking for stationary solutions in one dimension, i.e., setting $\frac{\delta s}{\delta t} = 0$ and

neglecting two components of the vectors for velocity and the diffusion coefficients and estimating a suitable source function, these investigators could analytically solve the resulting equation. These solutions were then fitted to observed data in the abyssal ocean to obtain values for the ratio of the diffusion coefficient to velocity in one dimension. The values obtained from different tracers agreed well when one considers that a different sink function for each tracer had to be estimated.

Our proposed circulation model is based on Equation 1 and is an attempt not to deduce the velocity and diffusion coefficients, but using the best known values for these variables to compute the oceanic distribution of long-lived radioactive tracers. Not only would we like a description of the three-dimensional distribution but also to know how this distribution changes with time. Such a comprehensive model requires a numerical solution of Equation 1 over a grid of points in the oceanic domain. We have chosen a finite difference form, Equation 2, in which spatial partial derivatives are represented by space-centered differences and the temporal partial derivative by a time-forward difference.

$$\begin{aligned} \frac{\Delta_t s}{\Delta t} = & \frac{\Delta}{\Delta x} \left(K_x \frac{\Delta s}{\Delta x} \right) + \frac{\Delta}{\Delta y} \left(K_y \frac{\Delta s}{\Delta y} \right) + \frac{\Delta}{\Delta z} \left(K_z \frac{\Delta s}{\Delta z} \right) \\ & - \left(V_x \frac{\Delta s}{\Delta x} + V_y \frac{\Delta s}{\Delta y} + V_z \frac{\Delta s}{\Delta z} \right) - R \end{aligned} \quad (\text{Equation 2})$$

where the time difference operator Δ_t is defined as $\Delta_t s = s(x, y, z, t+1) - s(x, y, z, t)$ and the space difference operators as $\Delta s = \frac{1}{2} [s(x+1, y, z, t) -$

$s(x-1, y, z, t)]$ and $\Delta^2 s = s(x+1, y, z, t) - 2s(x, y, z, t) + s(x-1, y, z, t)$ and similarly for the y and z directions. This equation is computationally stable if $\frac{K_m \Delta t}{\Delta m} \leq \frac{1}{2}$ [23] (where $m=x, y$ or z). Hence, one

must scale the grid system in space (Δm) and time (Δt) so that the inequality is satisfied.

Lack of sufficient data for the velocity and eddy diffusion coefficient fields and limitations of computer memory dictate the size of the ocean domain we can expect to investigate. Thus, we are beginning with the surface waters of the North Pacific. The horizontal grid consists of points separated by 5° intervals from the equator northward but excluding the western and northern marginal seas. The vertical grid consists of points spaced every 100 meters.

To fulfill the three necessary conditions we shall estimate the introduction of the tracer in the manner described in the preceding sections. Average surface velocity data are available for each 5° square for four months-February, May, August, and November[24]-and we hope to estimate the velocity field down to approximately 600 meters from published velocity profiles.[25] The horizontal components of the eddy diffusion coefficient can probably be neglected in the surface waters. Published profiles of the vertical component[26] and additional data from ⁷Be profiles will be our guide to estimate the field of vertical eddy diffusion coefficients.

Preliminary boundary conditions will be defined as follows: at the air-sea interface, the flux will be proportional to the air concentration; there shall be no flux from the continent margins; and at the boundary of the sediment interface, the flux shall also be zero. Of course, one can easily alter these conditions to reflect the probable latitudinal and temporal variation of air-sea transfer or possible fluxes from the marginal seas or infinite sinks at ocean depths, etc.

Using the scalar fields of the velocity-diffusion data, the described boundary conditions, and an appropriate sink function, we hope to compute a realistic model for surface circulation of the North Pacific Ocean at time intervals of three months. Such a model would be invaluable in predicting both the past and future horizontal and vertical transport of numerous tracers dissolved in seawater.

CONCLUSION

The radioactive half-life of ⁷Be is too short ($T_{1/2} = 53d$) for this isotope to provide information on long range advective water transport, but it is useful in tracing diffusion at the top of the thermocline which permits determination of one parameter of the transport equation. Beryllium-7 can, however, serve as an index of the oceanic input of longer-lived radionuclides which, themselves, are most useful tracers of advective transport. This can be accomplished by the following simplified arguments. The oceanic deposition of ⁷Be, and, by analogy, that of other constituents associated with atmospheric aerosols, is proportional to their concentrations in the overlying air mass. The annual cyclic variation of the ground level air concentration of ⁷Be results in a similar cycle in its surface concentrations and cumulative deposition in the oceans. This allows the determination of the annual ⁷Be deposition at any given point based on its surface water concentrations. Thus, by applying the direct proportion of the time integrated ratio of the air concentration of ⁷Be and ¹³⁷Cs at a given latitude and the surface water ⁷Be concentration, the rate at which ¹³⁷Cs is deposited on the sea surface can be estimated. From historical ¹³⁷Cs air concentrations it should be possible to determine what global air-sea transfer rates of ¹³⁷Cs have been as a function of time and space since its first injection, providing the differential source function for a dynamic model of ocean circulation.

TABLE I. RADIONUCLIDE CONCENTRATIONS IN
WATER COLUMNS AT 36°N, 68°W

Date	Depth (M)	^7Be	$^{95}\text{ZrNb}$	^{106}Ru	^{144}Ce
8-27-70	7 $\pm$ 2	892 $\pm$ 15	736 $\pm$ 2	445 $\pm$ 10	1010 $\pm$ 100
	20 $\pm$ 3	898 $\pm$ 14	793 $\pm$ 2	425 $\pm$ 10	990 $\pm$ 80
	39 $\pm$ 5	572 $\pm$ 12	544 $\pm$ 2	311 $\pm$ 7	700 $\pm$ 60
	65 $\pm$ 5	209 $\pm$ 11	234 $\pm$ 1	80 $\pm$ 5	360 $\pm$ 90
	88 $\pm$ 6	81 $\pm$ 29	133 $\pm$ 3	64 $\pm$ 10	260 $\pm$ 90
8-28-70	0	823 $\pm$ 21	722 $\pm$ 4	414 $\pm$ 11	860 $\pm$ 90
	11 $\pm$ 1	852 $\pm$ 14	702 $\pm$ 2	380 $\pm$ 9	840 $\pm$ 70
	26 $\pm$ 1	821 $\pm$ 12	680 $\pm$ 2	342 $\pm$ 7	1050 $\pm$ 70
	46 $\pm$ 2	249 $\pm$ 16	322 $\pm$ 2	125 $\pm$ 8	440 $\pm$ 60
	72 $\pm$ 1	98 $\pm$ 6	108 $\pm$ 1	39 $\pm$ 4	240 $\pm$ 30
	97 $\pm$ 1	54 $\pm$ 7	85 $\pm$ 1	40 $\pm$ 4	210 $\pm$ 40
8-29-70	0	729 $\pm$ 16	709 $\pm$ 2	417 $\pm$ 9	900 $\pm$ 60
	10 $\pm$ 1	727 $\pm$ 16	712 $\pm$ 2	409 $\pm$ 9	960 $\pm$ 90
	25 $\pm$ 1	724 $\pm$ 16	742 $\pm$ 2	406 $\pm$ 10	810 $\pm$ 70
	43 $\pm$ 1	302 $\pm$ 15	475 $\pm$ 3	269 $\pm$ 11	680 $\pm$ 90
	69 $\pm$ 1	127 $\pm$ 15	160 $\pm$ 2	60 $\pm$ 5	300 $\pm$ 50
	92 $\pm$ 1	88 $\pm$ 23	112 $\pm$ 2	75 $\pm$ 9	240 $\pm$ 80

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VERTICAL ^7Be CONCENTRATION GRADIENT IN PRE GEOSECS WATER COLUMN

FIGURE ^7Be AND ^{137}Cs IN GROUND LEVEL AIR AT RICHLAND, WASHINGTON.

SEASONAL CHANGE OF THE RATIO OF ${}^7\text{Be}$ WATER COLUMN INVENTORY TO ITS CONCENTRATION IN SURFACE WATER

⁷Be CONCENTRATIONS IN PACIFIC OCEAN SURFACE WATER