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INVENTORIES OF SELECTED RADIONUCLIDES IN THE OCEANS

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FOREWORD

In March 1984 an ad hoc Review Committee composed of senior experts in the marine radioactivity field met in Monaco to review the on-going scientific activities of the International Laboratory of Marine Radioactivity, IAEA, and to give guidance on the future orientation of its programmes. The Committee made several recommendations, one of which stated that "the Laboratory should be engaged in compiling and evaluating the input of radionuclides into the marine environment". The Committee felt that this task was important, not only for the IAEA programmes of waste management and radiological safety, but also for the work performed by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The task was subsequently approved at a meeting of the IAEA's Scientific Advisory Committee in December 1984.

To initiate the Laboratory's new task on a clearly defined basis a group of consultants, which included the Secretariat of the UNSCEAR, met in July 1985 in Monaco. At this meeting the extent of the work to be covered, its time-table and guidelines for approaches to be followed were discussed. The Group recommended that work should commence on selected radionuclides, viz., ^{14}C, caesium isotopes, plutonium isotopes, ^{210}Po and ^{210}Pb followed by ^{226}Ra. Depending on the radionuclides involved the assistance of competent experts from outside as well as inside the IAEA was sought. In November 1986 the experts' work was reviewed by a group of consultants.

The present document is a product of the work carried out within the framework of the above-mentioned task and contains reports on ^{14}C, ^{90}Sr, ^{137}Cs, ^{238}Pu, $^{239+240}$Pu, ^{210}Pb, ^{210}Po and ^{226}Ra.

Although the eventual goal of the work, viz., the estimation of the inventory in the marine environment and related input and output fluxes, is the same for all radionuclides concerned, different approaches were followed by the experts to achieve this objective. These approaches depended on the geochemical characteristics of the radionuclides and the availability of data for different times and locations. For regions where data were lacking, extrapolation on the basis of specific assumptions has often been necessary. As the work was initiated during the pre-Chernobyl period, the radionuclides derived from the Chernobyl incident were not, in general, considered.

Since the work for preparing the forthcoming report of the UNSCEAR is scheduled to be completed by 1991, it is hoped that the information contained in this volume will be beneficial. At the same time, as the reports indicate where a lack of data and information exists, it is also hoped that the scientific community at large will be motivated to fill these gaps so as to for arrive at more precise inventories and fluxes.

EDITORIAL NOTE

CONTENTS

ASSESSMENT OF THE INVENTORY OF CARBON-14 IN THE OCEANS: AN OVERVIEW*

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Abstract

The report overviews an earlier treatise (INS-R--348, Nov 1986) on the oceanic distribution of ^{14}C of both cosmogenic and bomb origin. The total oceanic inventory of cosmogenic ^{14}C is assessed at 1.96×10^{30} atoms (7500 PBq). The bomb-carbon inventory as of 1972 is assessed at 303×10^{26} atoms (116 PBq), based on analysis by Broecker, Peng, Ostlund and Stuiver. The atmospheric source of ^{14}C and its flux between atmosphere and ocean is discussed and quantified, from which the post-1972 transfer of bomb-carbon to the oceans is estimated to have bolstered the inventory there to 435×10^{26} atoms (167 PBq) by 1985.

1. INTRODUCTION

1.1 Background

This report is an overview, prepared at the request of the IAEA, of an earlier report (1) covering the oceanic distribution of ^{14}C. Estimated inventories of both natural (cosmogenic) and man-made (mainly bomb-origin) ^{14}C are presented, along with estimated rates of influx and eflux. Such estimates are summaries of published data.

1.2 Scope of Report

The only three significant sources of ^{14}C into the environment at this time are (a) that produced naturally by cosmic rays in the earth's atmosphere; (b) that produced since 1945 by explosion of nuclear weapons in the atmosphere; and (c) more recently an increasing input into the environment from the nuclear fuel cycle, associated with fission reactors used to produce electricity.

The numerous available measurements of ^{14}C concentrations in the ocean are almost exclusively since environmental contamination by bomb ^{14}C, so that prenuclear levels can only be obtained by inference. Such inferences are further complicated by the dilution of ^{14}C in the carbon pool by fossil-fuel CO_2 over the industrial era.

This report summarises the apportionment of carbon in the major reservoirs of the carbon cycle (section 2), the sources of atmospheric ^{14}C (section 3) and in section 4 the various surveys of oceanic ^{14}C (notably the GEOSECS surveys of 1972-8). Section 5 discusses the

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* This work was supported by the International Atomic Energy Agency under Technical Contract 4400/TC.

dynamics of oceanic carbon and its exchange with the atmosphere, and updates a post-GEOSECS estimate of the oceanic inventory of ^{14}C.

1.3 Citing Radiocarbon Data

For convenience, 10^{26}atom ^{14}C is referred to as 1 RCU (radiocarbon unit). Then 1 RCU = 0.384 PBq = 10.4 KCi.

Data in this report cite the ^{14}C ratio relative to the 0.95 NBS oxalic acid standard for ^{14}C which is an estimate of pre-industrial atmospheric ^{14}C ratios. Explicitly

$$\delta^{14}C = (R/R_{std} - 1) \times 1000 \qquad (1.1)$$

where R is the measured $^{14}C/C$ ratio and R_{std} is that for the 0.95 NBS standard which is (2)

$$R_{std} = 1.176 \times 10^{-12}. \qquad (1.2)$$

The customary $\delta^{13}C$ and $\Delta^{14}C$ values are defined by

$$\delta^{13}C = (R_{13}/R_0 - 1) \times 1000 \qquad (1.3)$$

and by

$$(1 + 10^{-3}\Delta^{14}C) = (1 + 10^{-3}\delta^{14}C)\left(\frac{.975}{1 + 10^{-3}\delta^{13}C}\right)^2 \qquad (1.4a)$$

or, to first order,

$$\Delta^{14}C = \delta^{14}C - 2(\delta^{13}C + 25)(1 + 10^{-3}\delta^{14}C). \qquad (1.4b)$$

Here R_{13} and R_0 are the $^{13}C/^{12}C$ ratios in the sample and PDB limestone standard, respectively. $\Delta^{14}C$ cites a "fractionation-corrected" $^{14}C/C$ ratio, normalised to a $\delta^{13}C$ value of $-25°/_{oo}$.

2. THE CARBON CYCLE

The natural production rate of ^{14}C has not been precisely constant in the past, being effected by the earth's magnetic field and solar activity. Thus even the distribution of natural ^{14}C between and within the different carbon reservoirs on the earth is not in a steady state.

It is also likely that this carbon cycle is effected by major oceanic circulation patterns which have changed in time.

Many researchers have developed mathematical models to simulate ^{14}C and carbon dynamics in the oceans, atmosphere and (terrestrial) biosphere. Such models serve to interpret surveyed ^{14}C and carbon concentrations and to guide estimates of inventories.

The three main reservoirs of carbon are the ocean, atmosphere and biosphere with approximately 37000, 615, 800 Pg, respectively (pre-industrial values(4)). A further 1500 Pg in the terrestrial humus is usually assumed to be isolated from these reservoirs. Of the large oceanic inventory, about 1000 Pg is in the form of organic carbon and the rest as dissolved inorganic carbon (DIOC). [Note that 1 Pg = 10^{15}g].

3. THE SOURCES OF RADIOCARBON

3.1 Natural ^{14}C Production

The ^{14}C isotope is produced continuously in the atmosphere by neutrons originating from cosmic rays. Low energy thermal neutrons and higher energy neutrons at resonance energies interact with atmospheric nitrogen in the reaction

$$^{14}N + n \rightarrow {}^{14}C + p \tag{3.1}$$

to yield ^{14}C atoms which are quickly oxidized to carbon monoxide and then to carbon dioxide. The ^{14}C isotope is radioactive and decays according to

$$^{14}C \rightarrow {}^{14}N + \beta \tag{3.2}$$

with a half life of 5730 years (mean life 8267 years). As the lifetime of ^{14}C is long compared to the time scales of the physical and chemical processes that affect carbon, ^{14}C becomes widely distributed in the atmosphere, biosphere and oceans. However because of its low specific activity (about 0.2 Bq/gC) and low radiation dose per decay, naturally produced ^{14}C in environmental carbon does not constitute a significant health risk.

Approximately 65% of the cosmogenic production is in the stratosphere, with the remaining 35% being in the troposphere and predominantly the upper troposphere(5). Cosmic ray flux studies and specific activity measurements provide concordant estimates of production rates ranging from 2.8 RCU/yr to 3.5 RCU/yr. Generally the lower end of this range is favoured by carbon cycle modelling studies.

Cosmogenic production is modulated by several external factors - principally the cycle in the earth's magnetic field strength with a period of 9000 years, and the solar sunspot cycle with a period of 11 years. The former has periodicity of same order as the ^{14}C lifetime and so must be accounted for in estimating cosmogenic ^{14}C inventories based on production and decay rates. The geomagnetic field, which fluctuates by about 50% of its current value, affects the cosmic ray flux which is intercepted by the earth's atmosphere. Models of this geomagnetic field modulation suggest that the global ^{14}C inventory fluctuates by about 8% of its mean with periodicity equal to that of the field but with a phase lag with respect to the field which places the current estimated inventory of 22000 RCU close to its lowest value.

3.2 Transport of ^{14}C in the carbon cycle

Atmospheric CO_2 exchanges with dissolved carbon in the oceans and is incorporated into the biosphere by photosynthesis. The ocean contains the greatest store of potentially exchangeable carbon, but the bulk of this is in the deep ocean where carbon residence times are several hundred years. The well-mixed ocean surface exchanges carbon with the atmosphere with time scale of 7-10 yr.

Exchange with the biosphere is complex, involving a wide range of different mechanisms and time scales. Leaf litter, some root decay and agricultural cropping gives rise to release within a year or two of photosynthesis, while dying forest trees etc. release carbon several decades after photosynthesis.

Consider briefly carbon exchange between the pre-industrial atmosphere and ocean mixed layer in just sufficient detail to show how the cosmogenic atmospheric ^{14}C inventory is determined.

The rate of change of the ^{14}C inventory in the atmosphere is given by combining the production rate, the loss due to flux into the oceans, the gain due to the reverse flux, and radioactive decay. After allowing for mass balance of carbon (equal and opposite fluxes across the interface), the atmospheric ^{14}C inventory I(t) may be written

$$dI/dt = Q - [k_{am}f(1 - A_m/A_a) + \mu]\, I. \qquad (3.3)$$

In this equation Q is the production rate and μ the decay rate; A_m and A_a are the $^{14}C/C$ ratios in the mixed layer and atmosphere, and k_{am} the exchange rate coefficient for the carbon flux from atmosphere to ocean; $f = 1.052$ is the isotopic fractionation at the interface (6).

The equilibrium inventory can be related to production by setting $dI/dt = 0$. Surface ocean carbon has on average $A_m = 0.955A_a$ (based on an extrapolation to the pre-industrial era of $\Delta^{14}C = -45°/_{oo}$ with perhaps $10°/_{oo}$ uncertainty - see section 4.3.2). For $k_{am} = 1/7.2\ yr^{-1}$ (see section 5.3.1) the equilibrium inventory satisfies $I/Q = 149$ yr which, for $Q = 2.8$ RCU implies an atmospheric inventory of 420 RCU (cosmogenic).

The effect upon this estimate of including biospheric exchange is small since returning carbon fluxes are much "younger" than those evading the oceans.

More direct estimates of atmospheric inventory based on pre-nuclear ^{14}C measurements are complicated by the need to make proxy measurements in tree rings (e.g., ref (7)) and because those measurements pertain only to the surface atmosphere. For a well-mixed pre-industrial atmosphere for which $\Delta^{14}C = 0°/_{oo}$ (and therefore $^{14}C/C = 1.176 \times 10^{-12}$) the carbon inventory of 615 Pg(4) includes a ^{14}C inventory of 363 RCU. A similar calculation by Telegadas (8) deduces an inventory of 373 RCU.

These "direct" inventory estimates are about 12% lower than those based on (3.3). Some of this discrepancy might be due to ^{14}C ratios being higher in the upper troposphere and stratosphere than at the surface. Indeed Telegadas (8) has noted 2% variations between surface measurements and aircraft measurements in the lower troposphere. Also Rafter and O'Brien (9) note variations of 2% between Australian continental ^{14}C ratios and those at other South Pacific coastal sites.

3.3 Bomb ^{14}C

Testing nuclear weapons in the atmosphere since 1945 has produced additional ^{14}C in an amount slightly exceeding the natural cosmogenic atmospheric inventory.

The intense neutron fluxes produced by nuclear explosions give rise to ^{14}C via the same mechanism as cosmic ray neutrons. Neutron yields depend upon the nature of the weapon, particularly upon the fission/fusion mix (10). Nevertheless it is common to cite a production per Mt of yield, with surface bursts rated at half nominal yield due to absorption of about half the neutron flux by the ground. An often quoted figure due to Machta (11) is 2 RCU per Mt, a value based on estimated neutron production and the proportion of these which would react with atmospheric nitrogen atoms.

B.J. O'Brien in report by UNSCEAR(12) estimates bomb production of ^{14}C at 570 RCU, and Enting and Pearman (13) estimate it at 550 RCU. The former estimate exploits correlations between ^{14}C and ^{90}Sr measurements. Enting and Pearman used a one-dimensional carbon cycle model with the nominal Mtonnages as input and the ^{14}C production per Mt explicitly adjustable to provide the best fit to ^{14}C measurements; the best fit revised the Machta estimate to 1.2 RCU per Mt. Considering the different approaches used, these estimates are surpringly concordant. The apparently large Machta figure may be due to a built-in conservatism often employed when health effects are being estimated.

The bulk of the yield from atmospheric nuclear tests occurred in 1961 and 1962, these two years accounting for over 70% of the total. Prior to this there had been an increasing number of tests from 1951 through to 1958, followed by a brief moratorium. These features apart, there has been a much lower level of testing throughout the period from 1945 to 1976, with rare explosions since then.

Figure 3.1 shows smoothed stratospheric and tropospheric Δ^{14}C values inferred from figures given by Telegadas(8) and values from surface measurements by Levin et al.(14) and by Manning et al.(15). Table 3.1 gives annual average inventories inferred from the data shown. Note that the inventory estimates based on surface measurements are expected to be too low at least until 1975 due to higher Δ^{14}C values in the stratosphere than in the troposphere.

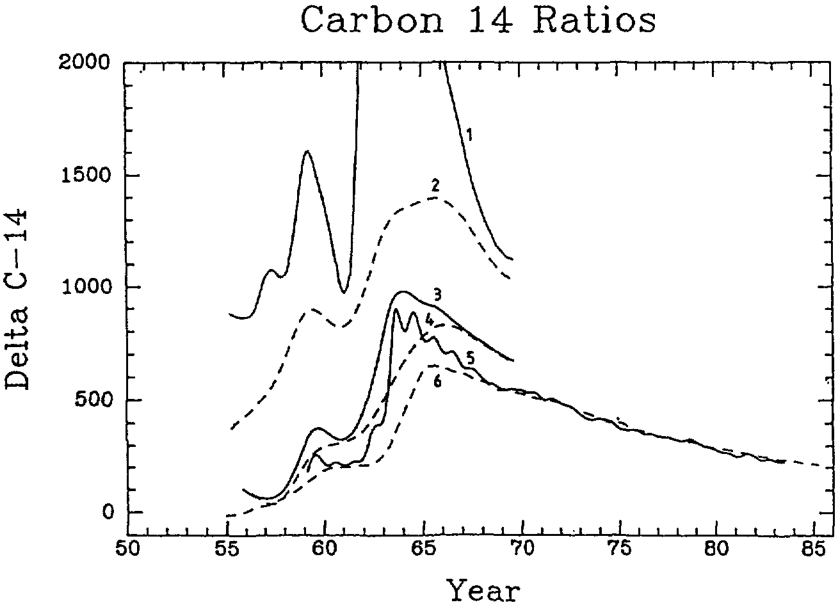

Fig. 3.1: Δ^{14}C excesses in the atmosphere as a result of nuclear bomb tests. Curves 1 and 2 are for the northern hemisphere (NH) and southern hemisphere (SH) stratospheres (respectively), after Telegades(8). Curves 3 and 4 are for aircraft measurements in the NH and SH tropospheres [Telegades (8)]. Curves 5 and 6 are based on surface air measurements for the NH [Levin et al. (14)]and SH [Manning et al. (15)].

The stratospheric data given by Telegadas(8) show some inconsistencies. For example the earliest values for atmospheric inventories would imply very high ^{14}C yield per Mt detonated prior to 1955. Similarly some of the early tropospheric values given show far more variation from one 3 month period to the next than appear reasonable from later studies. These probably reflect the difficulty of obtaining reliable inventory estimates from relatively sparse data.

The problem of sparse data coverage is particularly relevant for stratospheric bomb ^{14}C inventories. This is because the mixing times in the stratosphere are around 2 years, which is similar to the exchange time into the troposphere. As a result isolated measurements in the stratosphere soon after a nuclear explosion may miss the new input entirely, and measurements a short while later detect only part of it.

Table 3.1

INVENTORIES OF BOMB ^{14}C IN THE ATMOSPHERE
(in 10^{26} atoms of ^{14}C)

Year	Bomb yield (Mt)[1]	Stratosphere[2] NH	Stratosphere[2] SH	Troposphere[3] NH	Troposphere[3] SH	Total[4]	Surface NH[5]	Surface SH[6]
(Pre-1955	14.94)							
55	3.04	35	18	21			0	-1
56	14.94	35	18	10	13	76	0	5
57	16.46	45	21	11	5	82	0	9
58	48.72	45	30	21	18	114	0	19
59	0.00	64	40	62	47	213	45	30
60	0.05	51	31	47	45	174	41	38
61	111.01	48	31	57	50	186	42	40
62	220.50	189	50	88	65	392	69	43
63	0.00	267	53	146	86	552	137	65
64	0.01	157	52	144	111	464	161	103
65	0.02	99	57	140	123	419	145	124
66	0.90	80	55	130	126	391	133	120
67	3.08			120	119		120	115
68	9.30			113	111		108	107
69	3.10	45	41	105	103	294	105	103
70	5.40						102	99
71	1.67						96	96
72	0.11						90	91
73	2.50						81	84
74	0.50						75	78
75	0.00						71	74
76	4.10						67	69
77	0.02						64	65
78	0.04						63	61
79	0.00						57	58
80	0.60						51	55
81	0.00						49	51
82	0.00						46	48
83	0.00						43	45
84	0.00							42
85	0.00							41

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[1] Figures taken from Enting's and Pearman's summary of nuclear weapons testing.(13)

[2] Stratospheric measurements as reported by Telegadas(8)

[3] Tropospheric measurements from aircraft as reported by Telegadas(8)

[4] An estimate of total atmospheric inventory based on the preceeding 4 columns

[5] Estimates of Northern Hemisphere (troposphere + stratosphere) inventory based on surface measurements of Δ^{14}C at Vermunt, Austria by Levin et al(14)

[6] Estimates of Southern Hemisphere (troposphere + stratosphere) inventory based on surface measurements of Δ^{14}C at Wellington, New Zealand by Manning et al.(15)

Bomb ^{14}C production can be summarized as follows. Nuclear weapons testing in the atmosphere produced about 550 RCU predominantly in 1961 and 1962. For a brief period this nearly doubled the atmospheric inventory of ^{14}C. The bomb-produced excess of ^{14}C in the atmosphere has now reduced to about 90 RCU and is further depleting at about 6.1% per year.

3.4 The ^{14}C Production in Nuclear Reactors

Production of ^{14}C by nuclear power reactors currently in operation averages to about 2.0×10^{23} atoms per MW(e)-yr$^{(16)}$. With installed operating capacity of 240 MW(e) world-wide, production is about 0.5 RCU/yr, or about 17% of the cosmogenic production rate. Not all ^{14}C produced in nuclear reactors has yet been released into the environment, as some of it remains in unreprocessed fuel. However, up to 1985, the total emission of ^{14}C from nuclear reactors is only of the order of 1% of the total input from nuclear tests.

4. MEASUREMENTS OF RADIOCARBON IN THE OCEANS

In this chapter we examine the data base for radiocarbon in the world's oceans, and interpretations and analyses of that data. The oceans (a euphemism for all marine bodies) are subdivided geographically into three regions, termed the Atlantic, Pacific and Indian Oceans. These regions include those smaller marine bodies which are normally separately identified.

Hereafter the abbreviation DIOC denotes "dissolved inorganic carbon" (including the bicarbonate and carbonate ions) with DIO^{13}C and DIO^{14}C denoting the isotopically labelled species; square brackets enclosing a chemical entity denote its concentrations. Pre-industrial values of $\Delta^{14}C$ are denoted $\Delta^{14}C^{\circ}$ and $\Delta^{14}C^{*} = \Delta^{14}C - \Delta^{14}C^{\circ}$ denotes a Suess-effected bomb component, also called a $\Delta^{14}C$ excess.

4.1 Oceanic Surveys

Several systematic surveys of oceanic radiocarbon have taken place in the last 15 years. The most extensive is the Geochemical Ocean Section Survey (GEOSECS) which extended throughout the 1970's and covered the major marine bodies. A section of the North Pacific Ocean was surveyed in detail as part of the NORPAX program in 1979. The Transient Tracers in the Ocean (TTO) program surveyed portions of the Atlantic Ocean in 1981-3.

All of the above surveys are discussed in more detail below, as is a project entitled "Radiocarbon in the Sea" conducted by A. W. Fairhall and collaborators between 1968 and 1975.

4.1.1 The GEOSECS ocean surveys

The GEOSECS survey initiated in 1969 proceeded throughout the 70's to survey the chemical constituents of the world oceans. The objective was twofold: to provide a baseline study for future chemical changes; and to investigate large-scale oceanic transport and mixing processes. Measurements pertinent to radiocarbon determinations (including $\delta^{13}C$, $\delta^{14}C$ values and DIOC concentration) embrace only a few of the many geochemical species surveyed.

The major surveys covered the Atlantic(17) and Pacific(18) Oceans during 1972-3 and 1973-4 and the Indian Ocean(19) (with some Mediterranean and Red Sea stations) during 1977-8.

A determination of $DOI^{14}C$ distribution and an understanding of its sources, sinks and transport requires a good knowledge of physical oceanography. To the extent that radiocarbon measurements (especially the bomb component) are among the tracer information from which oceanographic data is obtained, the learning process is somewhat circular and iterative. GEOSECS radiocarbon data has helped deduce :

- upwelling rates and fluxes in the equatorial Atlantic necessary to account for depleted column inventories of $DIO^{14}C$ (20,21);
- mechanisms and magnitudes of vertical mixing in the Atlantic and Pacific thermoclines(22);
- Atlantic circulation patterns(6);
- replacement times of Pacific, Atlantic and Indian abyssal waters of about 510, 275, 250 years, respectively(23);
- gross features of oceanic circulation and interactions with atmospheric CO_2 (24).

A notable feature of all ^{14}C depth profiles(25) is the near-surface effects of "bomb carbon". Profiles of $\Delta^{14}C$ feature a pronounced peak at or near the surface of ~100°/₀₀ or larger (see fig 4.1) which is attributed to an invasion of bomb-produced $^{14}CO_2$.

4.1.2 The NORPAX shuttle experiment (Leg 3)

This experiment surveyed the upper 1000m of a N-S section of the central equatorial Pacific Ocean (between Hawaii and Tahiti) in April 1979. Quay et al.(26) report latitudinal variations in $\Delta^{14}C$ and in nutrient profiles from which are inferred equatorial upwelling rates and advective fluxes. The NORPAX results form a portion of the data interpreted by Broecker et al.(24)(section 4.3.5).

4.1.3 The TTO program

This program was designed to trace the movement of man-made geochemical tracers (principally tritium and radiocarbon from weapons tests) into the Atlantic interior as a follow-up to the GEOSECS survey. Surveys of the North Atlantic and tropical Atlantic regions were conducted in 1981 and 1983, respectively. The data, so far incompletely analysed, are the subject of a sequence of 12 papers in the Journal of Geophysical Research volume 90, number C4.

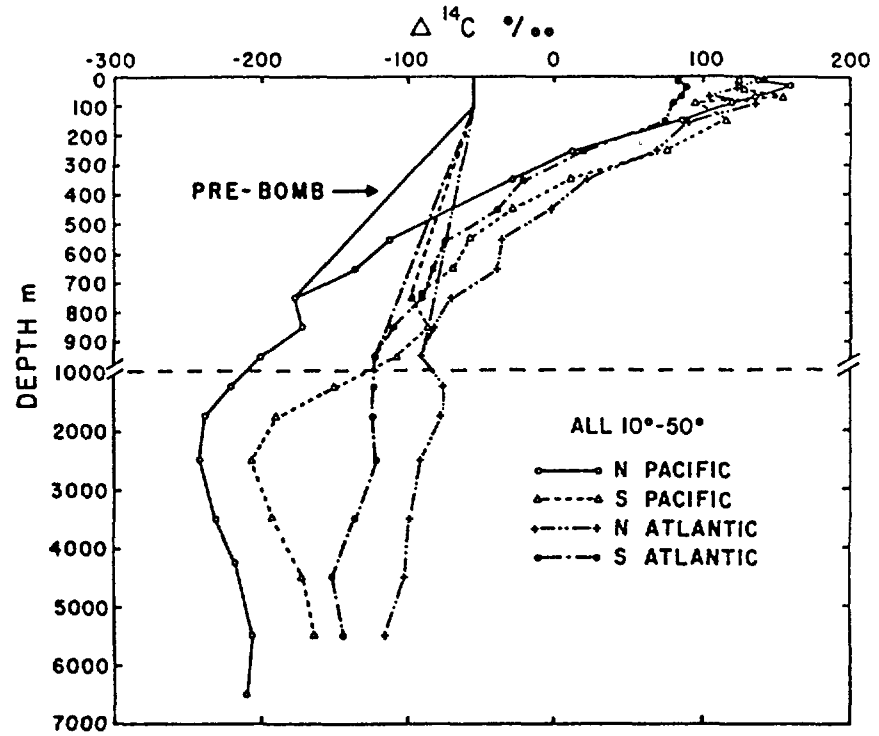

Fig. 4.1(a): $\Delta^{14}C$-depth profiles for the Atlantic and Pacific Ocean gyre reservoirs (10°-50° latitude). The assumed pre-bomb $\Delta^{14}C$-depth profiles all extend to $\Delta^{14}C = -55°/_{oo}$ at the surface. (Taken from Stuiver et al.$^{(25)}$).

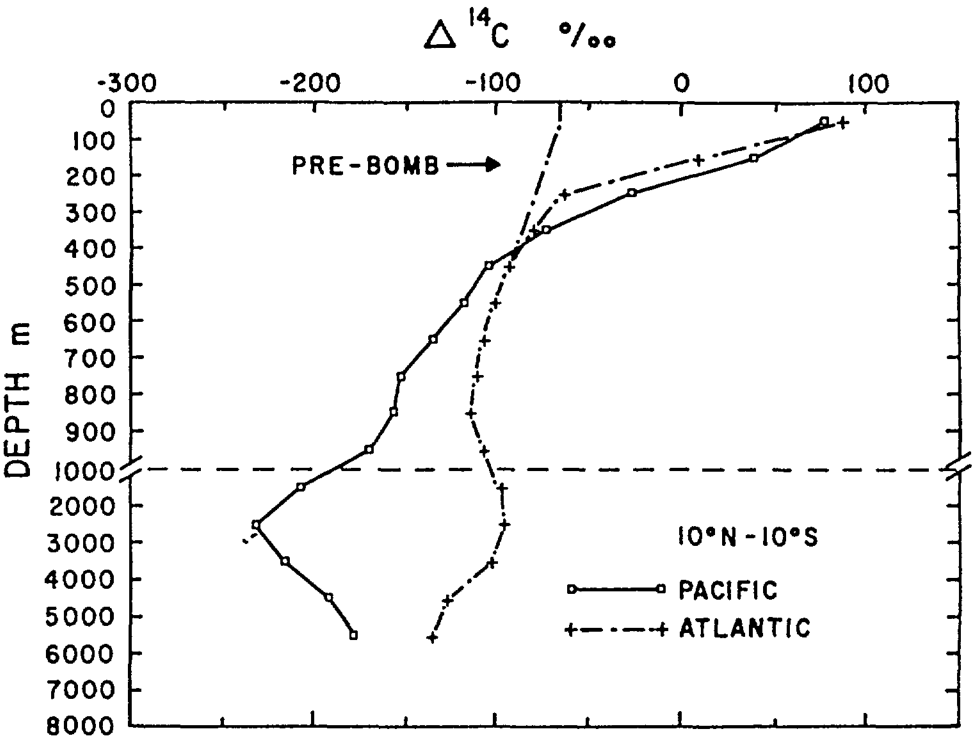

Fig. 4.1(b): $\Delta^{14}C$-depth profiles for the equatorial (10°N-10°S) regions of the Atlantic and Pacific Ocean. The assumed pre-bomb depth profile extends to a $\Delta^{14}C$ surface value of $-65°/_{oo}$. (Taken from Stuiver et al.$^{(25)}$).

4.1.4 "Radiocarbon in the sea"

A.W. Fairhall of the University of Washington and his collaborators surveyed marine radiocarbon(27-30); this followed a prior study of the fate of atmospheric bomb carbon which indicated a high rate of transfer to the oceans. The survey involved sampling marine waters (vertical profiles) at many locations from ships of opportunity between 1968 and 1975. A summary of the collected data is presented by Fairhall and Young(31), along with miscellaneous earlier data as early as 1958. Analyses of this data noted the near constancy of the cosmogenic [DOI^{14}C] throughout the oceans - see section 4.3.4.

4.2 A Summary of Physical Oceanography

This section summarises the main physical features of oceanic circulation, providing background and jargon for succeeding sections.

The oceans occupy a surface area of about 3.6 x 10^{14} m^2 (71% of the earth's surface) with volume 1.35 x 10^{18} m^3, and mean depth therefore 3.7 km. Despite being relatively thin, the oceans are highly stratified. The well-mixed surface layer, homogenised vertically by wind and thermal stresses, varies between about 20 m and 200 m in depth, averaging about 75 m. It is deepest in the temperate regions (roughly delineated by 15-50°N and S) and somewhat shallower in both tropical and polar regions - in the latter of which it is barely discernible all year round(4). The mixed layer is underlain by the main thermocline with lower boundary typically 1km below the surface.

The stratification follows isopycnal surfaces (constant density) which are deepest (i.e. the density gradients are lowest) in the temperate regions and noticeably shallower in the tropics (~15°S-15°N). The isopycnal surfaces approach the polar surfaces where they may actually "outcrop" (i.e. intersect the ocean surface). Fig 4.2 illustrates these isopycnals for the Atlantic Ocean.

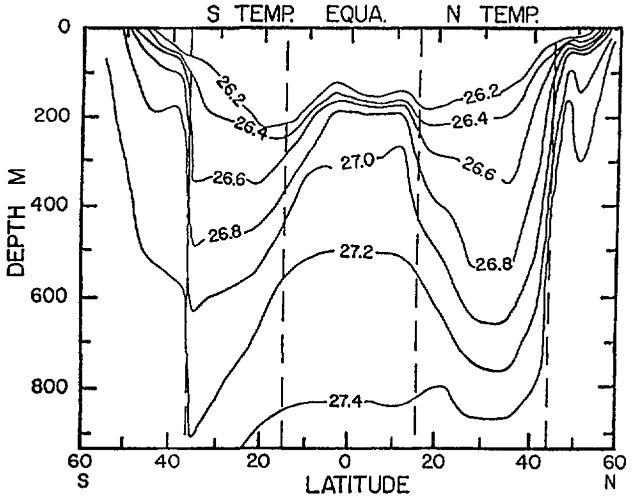

Fig. 4.2: Density versus depth along the western Atlantic GEOSECS track. The dashed lines separate the deep temperature thermocline from the shallow equatorial thermocline. The density units are per mil deviation from 1 kg/l. (Taken from Broecker and Peng,(32)).

The densest water is thus sourced near the poles from where it can flow equatorward and downward (along isopycnal surfaces) - particularly during winter when outcropping is more prominent. All the major oceans (Atlantic, Pacific, Indian) are thus fed with deep water from the easterly Antarctic circumpolar current (ACC), whereas only the North Atlantic has ready access to northern polar waters. Consequently, the Atlantic Ocean exports some deep water to the Indian and thence to the Pacific Oceans via the ACC, with compensatory counterflow of warmer thermocline water(33); see fig 4.3. That the deep Atlantic is a nett exporter of water to the ACC is supported by Δ^{14}C measurements(23).

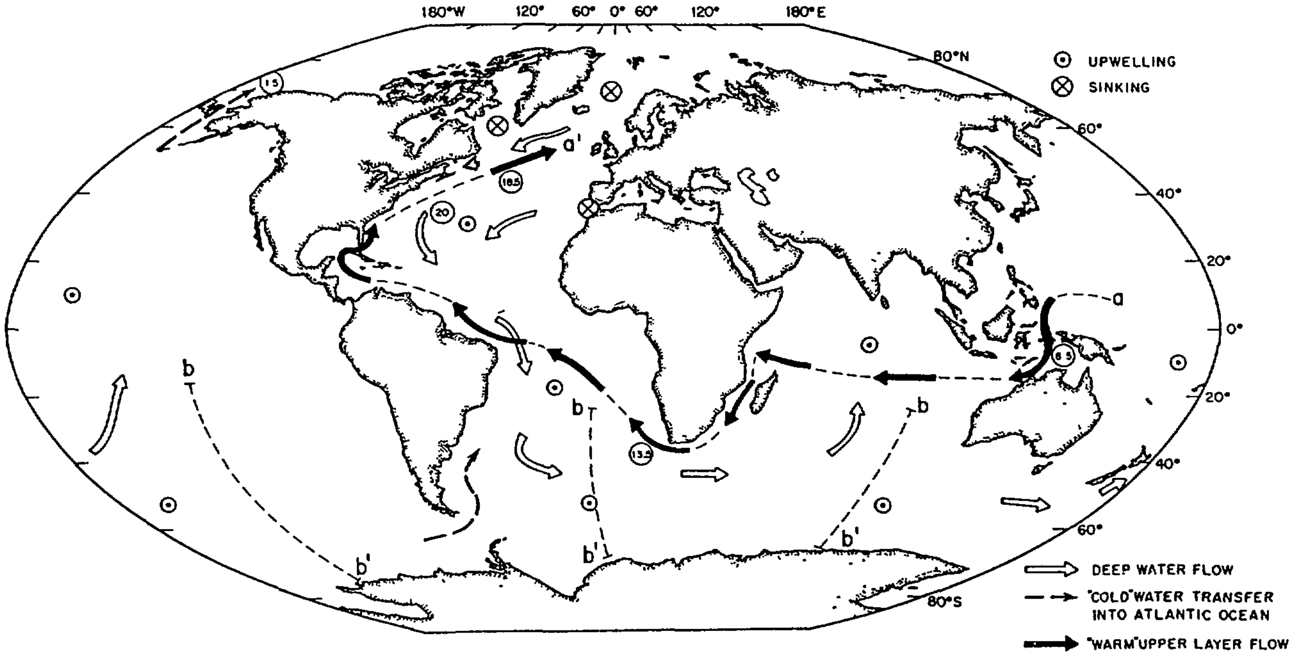

Fig. 4.3(a): Global structure of the thermohaline circulation cell associated with NADW production. The warm water route, shown by the solid arrows, marks the proposed path for return of upper layer water to the northern North Atlantic as is required to maintain continuity with the formation and export of NADW. The circled values are volume flux in 10^6 m^3/s which are expected for uniform upwelling of NADW with a production rate of 20×10^6 m^3/s. These values assume that the return within the cold water route, via the Drake Passage, is of minor significance. (NADW = North Atlantic Deep Water.) (Taken from Gordon(33)).

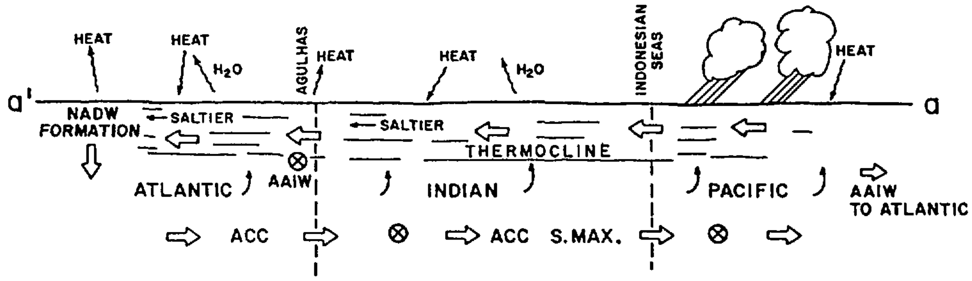

Fig. 4.3(b): Schematic representation of the thermohaline circulation cell associated with NADW production and the warm water route along line a-a' shown in Figure 4.3(a). The upper layer water within the main thermocline begins its passage to the North Atlantic in the Pacific as low-salinity water. It enters the Indian Ocean via the Indonesian seas, where its salinity and volume flux increase by excess evaporation and further upwelling of NADW, respectively. The thermocline water enters the Atlantic south of Africa and spreads to the northern Atlantic, continuing to increase in salinity and volume flux (from Gordon(33)).

The relative shallowness of tropical isopycnal surfaces indicates a nett upward advection (equatorial upwelling), with a downward advection (downwelling) in the neighbouring temperate regions(20,32). Such advective fluxes are confirmed by $\Delta^{14}C$ values, which signify relative depletion and enhancement in these respective regions (6,20,22,24). However, quantitative fluxes are not yet in concensus (21).

The temperate regions coincide with the gyre reservoirs, so named in recognition of the large-scale anticyclonic vortices which slowly stir the water in those regions.

4.3 Separation of Cosmogenic and Bomb Radiocarbon

The virtue of segregating the bomb contribution from the oceanic ^{14}C record is that the latter represents a useful tracer of oceanic DIOC. Furthermore, validated models of the global carbon cycle should be able to account for both the near-steady cosmogenic ^{14}C distribution and the environmental response to the bomb-carbon signal. The terms "pre-industrial" and "cosmogenic" $DIO^{14}C$ are used as if synonymous, even though the recent history of the latter may be Suess-effected; see section 4.4 for more detail.

The segregation of the bomb and cosmogenic $DIO^{14}C$ is complicated by the dearth of pre-nuclear $DIO^{14}C$ measurements. This dearth is partly offset by proxy measurements which can be made in certain banded species of coral.

4.3.1 The coral record

A convincing documentation of the Suess and nuclear effects in the surface ocean is recorded in banded corals, in much the same way as the atmospheric record is set in tree rings. These corals grow in many tropical and temperate areas and draw their carbonate skeleton from the DIOC of their host waters(34-39). That a reliable record of historic $\Delta^{14}C$ values for those host waters is thereby laid down has been confirmed by cross-checking with historic oceanic records.

The coral record is a valuable aid to assessing the bomb-carbon history in local surface waters, by extrapolating the pre-nuclear era. The Suess effect does, however, complicate such extrapolation. Of necessity this technique is available only for certain coastal tropical and temperate environs where suitable corals grow. A summary of the coral $\Delta^{14}C$ record is displayed in fig 4.4.

4.3.2 Pre-nuclear oceanic carbon-14

Few oceanic ^{14}C measurements were taken before the nuclear era. Broecker et al.(40) have summarised most of those available from the Atlantic Ocean and those early in the nuclear era (predating the significant bomb-carbon production of 1961-2). Data from the early 1960s in the Pacific and Indian Oceans are reported by Bien et al.(41). Other early Pacific data is reported in refs (31,42).

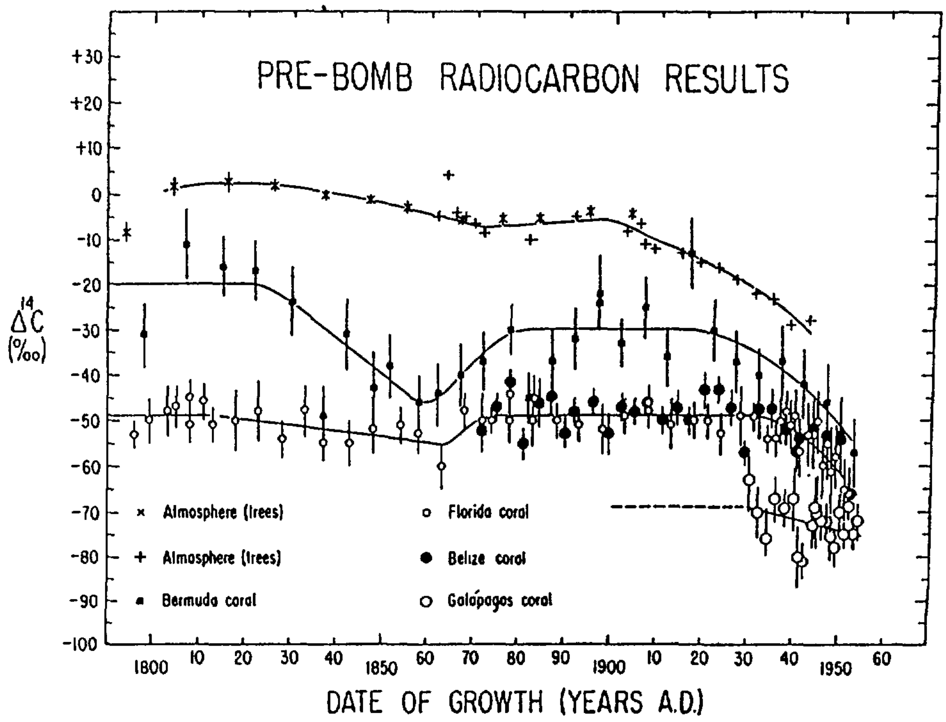

Fig. 4.4(a): Radiocarbon measurements for corals and tree rings that grew during the period A.D. 1800-1952. The trees were collected from the Pacific Northwest in the United States and from the Netherlands. The corals were collected from Florida and Belize(35, 36), from the Galapagos Is.(37), and from Bermuda(34). (Taken from Druffel and Suess(39).)

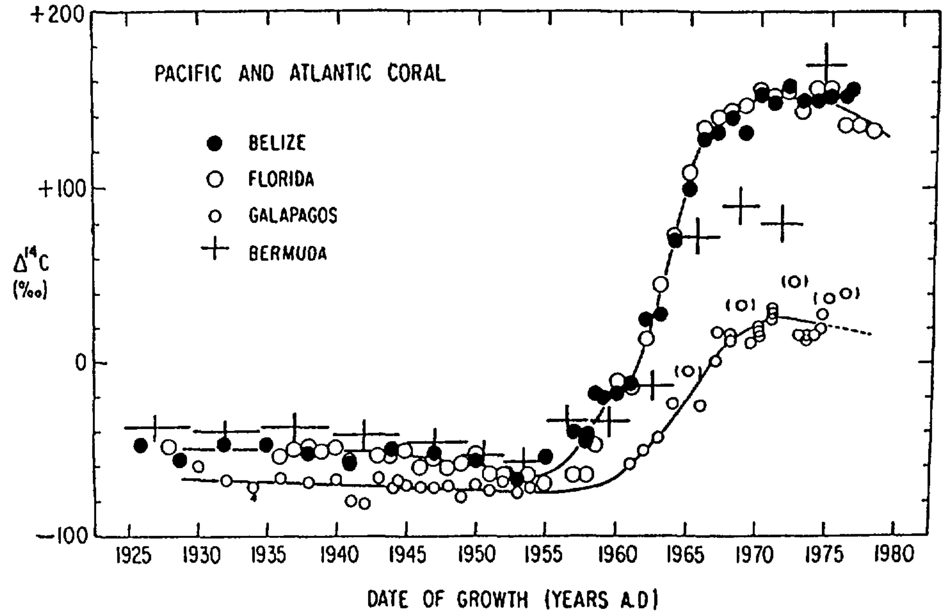

Fig. 4.4(b): Radiocarbon measurements for the corals of fig. 4.4(a) growing from 1925 to 1978. A considerable difference is apparent among the coral locations. This is attributed to the source of surface waters for each location. (Taken from Druffel and Suess(39).)

Killough and Emanuel(43) have assembled a pre-industrial record from sources cited above. A surface value of $\Delta^{14}C° = -40°/_{oo}$ is assumed in preference to measured values, which may be affected by bomb carbon. See section 4.3.6.

The $\Delta^{14}C°$ value $-40°/_{oo}$ has been widely accepted as a reasonable average for the pre-industrial ocean surface. Broecker and Li(44) proposed that value after surveying the pre-nuclear data; however, for reasons unstated, they excluded data taken poleward of 45° latitude. In an analysis of coral records, Druffel and Suess(39) proposed $\Delta^{14}C° = -40 \pm 2°/_{oo}$ for the mid-gyre region of the North Atlantic and North Pacific Oceans, and an equatorial value of $-69°/_{oo}$ (the latter based upon Galapagos Is. corals). This reflects an expected latitudinal variation in $\Delta^{14}C°$, with lower surface values in areas of upwelling - indeed values of less than $-100°/_{oo}$ for southern ocean surfaces have been reported in references cited above. Broecker <u>et al.</u>(45) have recently recommended $\Delta^{14}C° = -45°/_{oo}$ as an appropriate oceanic average.

In the pre-nuclear 1950s Suess-effected surface $\Delta^{14}C$ values in the range -50 to $-70°/_{oo}$ are often cited for low and mid latitudes.

4.3.3 The Stuiver et al. analysis

Stuiver et al.(25) summarised Atlantic and Pacific GEOSECS data into three latitude bands: equatorial (10°N-10°S), gyre (10°-50°N and S); the polar regions (latitude >50°) were not considered in detail. That summary is presented in fig 4.1.

The bomb carbon is segregated by assuming a pre-nuclear ocean surface with $\Delta^{14}C = -55°/_{oo}$ for the gyre reservoirs and $-65°/_{oo}$ for the equatorial zones, and by assuming that radiocarbon penetration parallels that of tritium. The reconstructed pre-nuclear profiles are included on fig 4.1.

The ^{14}C concentration averaged in each latitude band in each of the two oceans is computed from the formula (with modified notation)

$$[DIO^{14}C]_S' = 1.176 \times 10^{-12}(1 + 10^{-3}\Delta^{14}C)([DIOC]). \qquad (4.1)$$

Here, the molar concentrations $[DIO^{14}C]_S'$ and [DIOC] have the same units; the significance of the prime on the left-hand side is discussed in section 4.3.7.

The $DIO^{14}C$ concentrations are displayed in fig 4.5. Table 4.1 records the CO_2 invasion rates and column inventories of bomb carbon in each latitude band, as estimated by Stuiver et al. (the former from the atmospheric $\Delta^{14}C$ history in that latitude band). Also computed in table 4.1 are the bomb carbon inventories for the gyre and equatorial zones. The near equality of the average column inventories for the two oceans is noteworthy (though regions poleward of 50° are excluded). A rough estimate of the total oceanic inventory can be inferred by

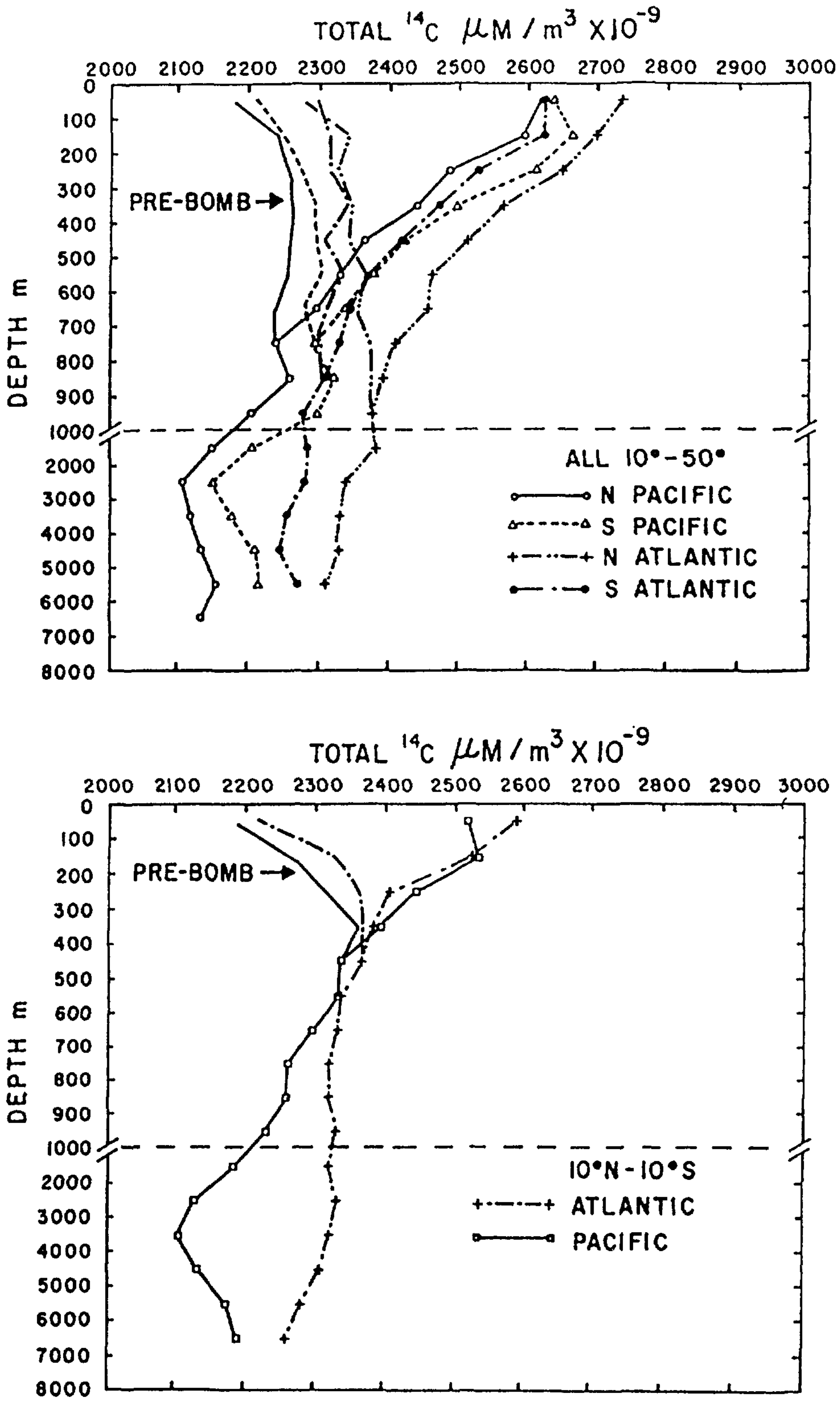

Fig. 4.5: Dissolved inorganic ^{14}C versus depth corresponding to the profiles of fig. 4.1. Note that 2000×10^{-9} µM/m^3 is equivalent to 1.2×10^9 atoms (^{14}C)/l. (Taken from Stuiver et al.(25).)

Table 4.1

Atlantic and Pacific GEOSECS bomb-carbon inventories and CO_2 invasion rates summarised over mid latitude bands*

Zone	CO_2 invasion rate ($mole/yr/m^2$)	Water area (10^{12} m^2)	Col. inventory (10^2 $atoms/m^2$)	Inventory (10^{26} atoms)
Atlantic Ocean				
50°N-10°N	27.2	29.8	110	32.7
10°N-10°S	11.1	12.7	42	5.4
10°S-50°S	20.2	27.9	72	20.2
50°N-50°S	21.4	70.4	83	58.2
Pacific Ocean				
50°N-10°N	21.0	53.4	89	47.3
10°N-10°S	12.1	40.2	48	19.4
10°S-50°S	26.5	57.9	100	57.9
50°N-50°S	20.8	151.5	82	124.5

* The invasion rates and column inventories are reported by Stuiver et al.(25), and the water areas by Broecker et al.(24); the inventories are computed in this work.

extrapolation of 82 x 10^{12} atoms(^{14}C)/m^2 to the world oceans, yielding 290 RCU.

4.3.4 The Fairhall analysis

Fairhall and Young(31) reported their results of oceanic DIOC surveys conducted in the early 1970s (see section 4.1.4). Those results included $\delta^{13}C$, $\delta^{14}C$, $\Delta^{14}C$ and the DIOC molarity as a depth profile at each of many oceanic stations. From each result was computed the "absolute ^{14}C concentration" (denoted herein as $[DIO^{14}C]_F'$) via the formula

$$[DIO^{14}C]_F' = 7.02 \times 10^8 \left(1 + \frac{\Delta^{14}C + 40}{1000}\right)([DIOC]) \text{ atoms } (^{14}C)/\ell, \quad (4.2)$$

where [DIOC] is expressed in mmole/litre. The supporting rationale for the sum ($\Delta^{14}C$ + 40) is that "a value -40°/₀₀ is assumed for pre-industrial surface ocean water, and the specific activity of surface ocean water is taken to be the same as modern wood, i.e. equivalent to 5.85 x 10^{10} ^{14}C atoms/g(C)" (28). The "Fairhall estimate" (4.2) exceeds by a factor 1.040 an estimate based on (4.1).

Fairhall et al.(27,31) noted that in deep oceans (e.g. below 1500m) where infiltration of bomb carbon should so far be minimal $[DIO^{14}C]_F'$ was remarkably constant, citing a value of (1.42 ± 0.02)x10^9 atoms per litre. Slightly lower values were noted for the North Pacific's older water mass (1.38×10^9) and larger values for the North Atlantic (1.47×10^9), which may be contaminated at depth by bomb carbon. Typical depth profiles are presented as fig 4.6. Since, with measured surface DIOC concentrations of about 2.0 mmole/litre, the pre-industrial ocean surface seems also compatible with 1.4×10^9 atoms(^{14}C)/litre, Fairhall et al. conjectured that such near constancy must have featured throughout the pre-industrial ocean.

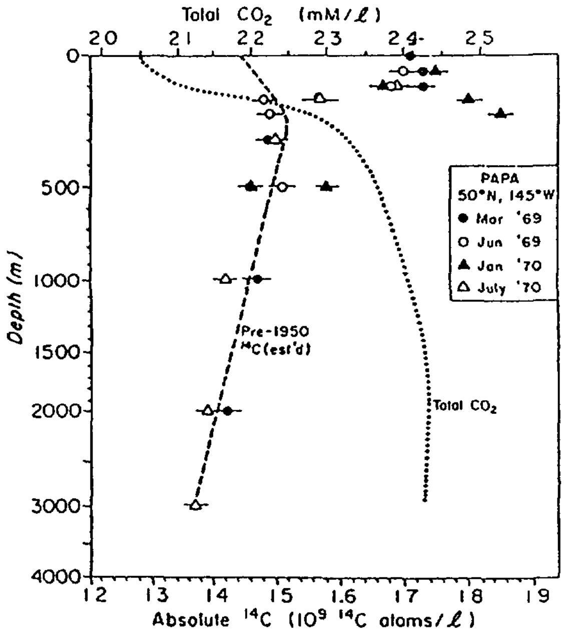

Fig. 4.6(a): ^{14}C concentrations at the North Pacific weather station PAPA. The dotted line gives the total DIOC profile. The dashed line is the estimated natural ^{14}C level. (Taken from Fairhall et al.[27].)

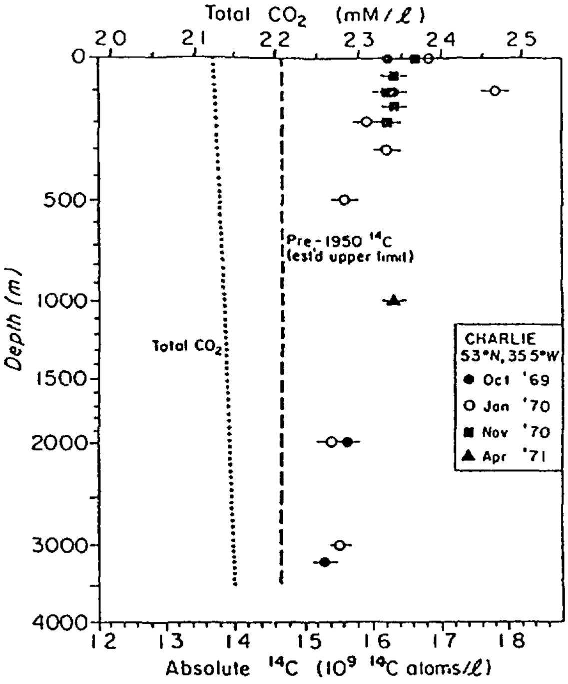

Fig. 4.6(b): As for (a), but for the North Atlantic weather station CHARLIE.

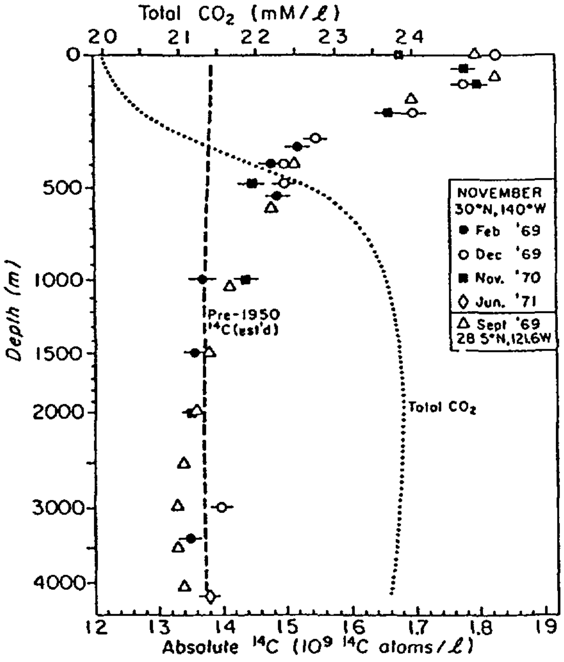

Fig. 4.6(c): As for (a), but for the mid-Pacific Ocean weather station NOVEMBER. Open triangles are data from the GEOSECS station.

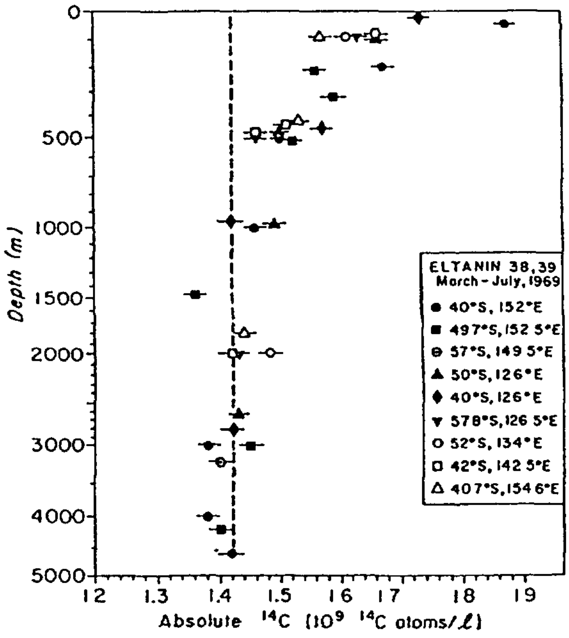

Fig. 4.6(d): As for (a), but for the ocean south of Australia in mid-1969.

A conceptual difficulty with a universal ^{14}C concentration is the absence of an obvious mechanism for maintaining this balance. Fairhall(29) explains this pre-industrial steady state as a balance between a downward rain of inorganic ($CaCO_3$) and organic detritus into the deep water where it dissolves, and an upward vertical diffusive flux driven by the DIOC concentration gradient. Quantitative estimates of these opposing fluxes add credibility to this accidental balance.

Craig(46) had earlier also noted a near constancy of ^{14}C concentrations in abyssal waters and speculated on a similar mechanism for its maintenance.

While Fairhall does not assess the oceanic cosmogenic inventory of $DIO^{14}C$, the straight-forward product of 1.42×10^9 atoms/litre and oceanic volume (1.35×10^{21} litre) suggests a cosmogenic inventory of 1.92×10^4 RCU. However there remains a question-mark over the interpretation of equn (4.1): see section 4.3.7.

4.3.5 The Broecker at el. analysis

Broecker *et al.*(24) analysed GEOSECS, NORPAX and TTO data, matching ocean-average bomb ^{14}C inventories to concentrations in overlying air and thereby deducing ocean-average invasions rates. Systematic discrepancies which emerge between station-by-station inventories and invaded input then point to possible departures from "ocean average" behaviour.

A bomb-carbon balance at the ocean-atmosphere interface requires a knowledge of the history of the $\Delta^{14}C$ excess (viz, $\Delta^{14}C^*$) in both the atmosphere and ocean surface. Broecker *et al.* assume that the required atmospheric history is represented by the many reported surveys, and that the coral record provides sufficient information to estimate the ocean-surface history of $\Delta^{14}C^*$. Each ocean-average invasion rate is deduced by equating time-integrated input to inventory; these rates (denoted I in the tables) are 22.3, 19.4 and 19.2 mole/m^2/yr for the Atlantic, Pacific and Indian Oceans, respectively. A map of $\Delta^{14}C^*$ segregated in this way is presented as fig 4.7.

Broecker *et al.* map the bomb-carbon column inventories (i.e. inventories of unit oceanic columns). They are deduced from segreated $\Delta^{14}C^*$ profiles at each station, integrating down to the penetration depth. The column inventories are integrated into 5° latitude bands and normalised against the atmospheric invasion assessed from the atmospheric $\Delta^{14}C$ record in that band. The resulting inventory/input quotients average to unity in each ocean and are mapped in Fig. 4.8. These quotients, which should be insensitive to the timing of the particular survey, evidently exceed unity only in the temperate zones; this is attributed to the convergence and downwelling of surface water in these zones in conjunction with latitudinally variable invasion rates.

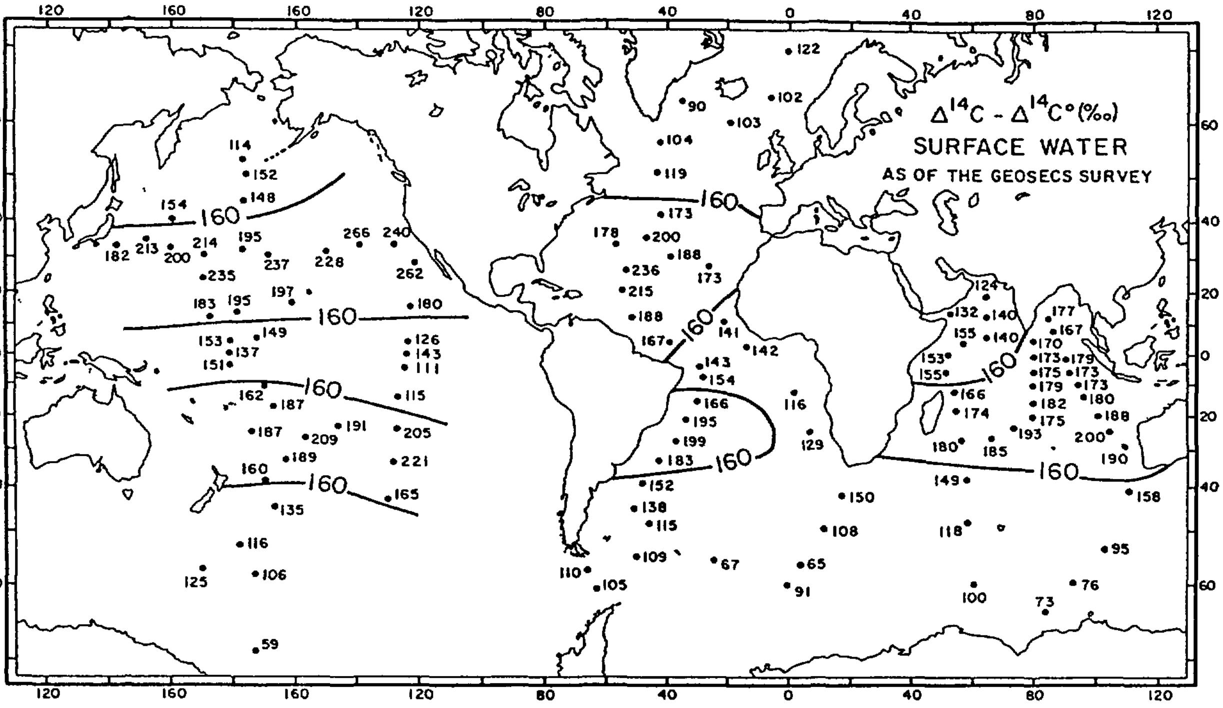

Fig. 4.7: Map showing the distribution of $\Delta^{14}C$ excesses (over the prenuclear values) for surface waters at the time of the GEOSECS surveys. (Taken from Broecker et al.[24].)

Bomb ^{14}C inventories are tabulated in two different ways. The first (table 4.2a) facilitates comparison with table 4.1; such a comparison awaits section 4.3.7. The second (table 4.2b) presents inventories and inputs in broad latitude bands which, according to fig 4.8, mainly separate regions of excess inventory (over input) from those of deficit. The total inventory of 289 RCU updates an earlier estimate[47] of (314±35) RCU based on a smaller data base.

Table 4.2a

Bomb-carbon inventories calculated by Broecker et al. for mid-latitude Atlantic and Pacific Oceans*

Latitude	Atlantic Ocean Col. inventory (10^{12} atoms/m^2)	Atlantic Ocean Inventory (10^{26} atoms)	Pacific Ocean Col. inventory (10^{12} atoms/m^2)	Pacific Ocean Inventory (10^{26} atoms)
50°N-10°N	122	36.5	93	49.4
10°N-10°S	30	3.8	50	20.1
10°S-50°S	86	24.0	94	54.7
50°N-50°S	91	64.3	82	124.2

* The latitude bands duplicate those of Table 4.1; the data is adapted from Table 6 of Broecker et al.[24]. All inventories estimated as of 1972.

Table 4.2b

Bomb-carbon Summary for Individual Ocean Regions Showing Either Excess or Deficient Inventories Relative to the Amount Expected if the CO_2 Invasion Rate Were Uniform Over the Ocean*

Latitude Band	Water Area (10^{12} m^2)	Inventory (10^{26} atoms)	Input (10^{26} atoms)	Inventory-Input# (10^{26} atoms)
		Atlantic Ocean (I = 22.3 mol/m^2/yr)		
80°N to 40°S	18.6	26.6	19.7	+ 6.9
40°N to 20°N	15.8	23.0	14.5	+ 8.5
20°N to 20°S	26.7	10.8	23.1	-12.3
20°S to 45°S	18.4	18.5	14.2	+ 4.3
45°S to 80°S	15.1	5.2	12.6	- 7.4
80°N to 80°S	94.6	84.1	84.1	0.0
		Indian Ocean (I = 19.4 mol/m^2/yr)		
25°N to 15°S	27.0	13.2	24.1	-10.9
15°S to 45°S	29.8	40.5	20.3	+20.2
45°S to 70°S	20.7	15.9	25.2	- 9.3
25°N to 70°S	77.5	69.6	69.6	0.0
		Pacific Ocean (I = 19.2 mol/m^2/yr)		
65°N to 40°N	15.1	7.3	13.4	- 6.1
40°N to 15°N	35.0	35.9	27.8	+ 8.1
15°N to 10°S	50.0	23.7	39.1	-15.4
10°S to 55°S	63.0	61.3	43.6	+17.7
55°S to 80°S	13.8	6.6	10.9	- 4.3
65°N to 80°S	176.9	134.8	134.8	0.0
		World Ocean		
	349.0	288.5		-

* Table from Broecker et al.(24) with corrections after Peng(48). All inventories estimated as of 1972.

\# The mean CO_2 invasion rate, I, is adjusted for each ocean so that input matches inventory.

The excesses and deficits of inventories over input evident from fig 4.8 and table 4.2b are in qualitative accord with prevailing oceanographic wisdom on invasion-rate variations and oceanic advection patterns. However, even with these patterns included, the model cannot account for the observed decline in atmosphere-borne fossil-fuel CO_2(48).

4.3.6 The Killough and Emanuel data assemblage

Killough and Emanuel(43) set out to compare the capabilities of several oceanographic models to account for the history of fossil-fuel transfer (as CO_2) to the oceans. In order to do this they

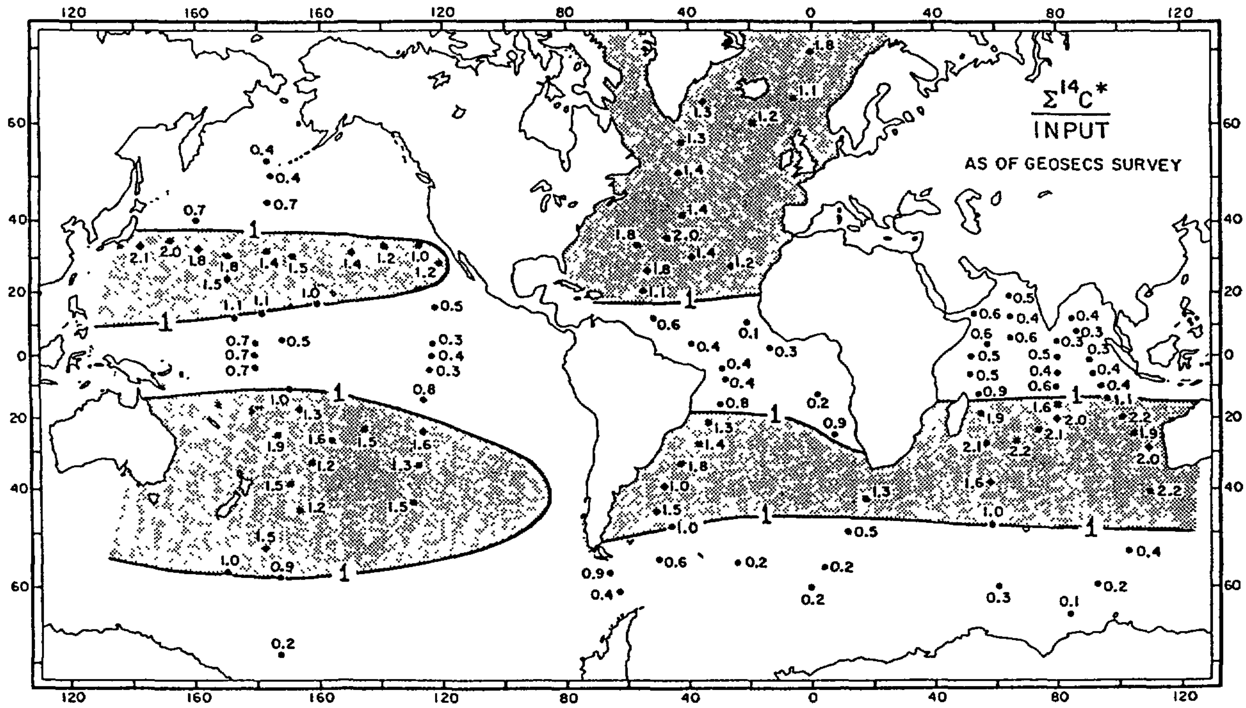

Fig. 4.8: Ratio of water column inventory to net input of bomb radiocarbon for the stations occupied during the GEOSECS program. The ratio averaged over each of the three main oceans is unity. (Taken from Broecker et al.(24)).

calibrated each model by insisting that its steady state reproduces the pre-industrial distribution of DI$O^{14}C$. The data base assembled for this calibration can also serve to supply an estimated inventory of that pre-industrial DI$O^{14}C$.

From the data reported by Broecker et al.(40) and by Bien et al.(44) Killough and Emanuel assembled data for depths below 200m data from 191 locations: 81 from the Atlantic, 62 from the Pacific, and 48 from the Indian Ocean. The depth cut-off at 200m was expected to exclude water likely to have been contaminated by bomb ${}^{14}C$. Killough and Emanuel extrapolate this data to a supposed $\Delta^{14}C^\circ = -40^\circ/_{\circ\circ}$ at the ocean surface.

Killough and Emanuel aggregated data into stratifications embodying the assumption that $\Delta^{14}C^\circ$ decreased with depth. Their data assemblage can be accumulated to estimate the pre-industrial inventory

$$\sum{}'_{KE}\ {}^{14}C = 1.90\mathrm{x}10^4\ \mathrm{RCU}.$$

The prime on $\sum$ denotes that ${}^{14}C/C$ ratios are fractionation corrected (see next section).

4.3.7 Oceanic inventories of cosmogenic and bomb-produced radiocarbon

The $^{14}C/C$ ratio, R, in a sample can be expressed in terms of $\delta^{14}C$ of (1.1) :

$$R = R_{std} \; (1 + 10^{-3}\delta^{14}C). \qquad (4.3)$$

The only assumption made is that there is no isotopic fractionation during the collection of a representative water sample and during its processing for isotopic determination. Thus, setting $R_{std} = 1.176\text{x}10^{-12}$,

$$[DIO^{14}C] = 1.176\text{x}10^{-12} \; (1{+}10^{-3}\delta^{14}C)([DIOC]), \qquad (4.4)$$

where the concentrations $[DIO^{14}C]$ and [DIOC] are in the same units (e.g. m.mole/litre). Integrating over the oceanic volume yields the the total $DIO^{14}C$ inventory.

Formula (4.4) contrasts with the formula usually adopted(2) to compute the $DIO^{14}C$ content of an oceanic reservoir. The latter has $\Delta^{14}C$ in place of $\delta^{14}C$, and takes the form

$$[DIO^{14}C]' = 1.176\text{x}10^{-12} \; (1{+}10^{-3}\Delta^{14}C)([DIOC]). \qquad (4.5)$$

Whichever of (4.4), (4.5) is the more appropriate depends upon the interpretation placed upon the result. Formula (4.4) correctly produces a reservoir inventory. Formula (4.5) is appropriate for considering carbon transfer processes for which ^{14}C is merely a tracer and for which isotope fractionation occurs (and is quantifiable from the $\delta^{13}C$ values). The latter formula would then measure an apparant or fractionation-corrected radiocarbon content.

Although $\delta^{14}C$ is a more fundamental experimental result than $\Delta^{14}C$ of equn (1.4), it is the latter which is the more extensively reported. It is thus useful to relate (4.4) and (4.5) :

$$[DIO^{14}C] = [DIO^{14}C]' \left(\frac{1 + 10^{-3}\delta^{13}C}{0.975}\right)^2, \qquad (4.6)$$

which effectively reconstructs $\delta^{14}C$ from $\Delta^{14}C$.

Fortunately, oceanic values for $\delta^{13}C$ do not vary by more than a few parts per mil - much less than variations in $\Delta^{14}C$. Kroopnick(49-50) presents extensive oceanic $\delta^{13}C$ measurements (including GEOSECS) which are largely confined to the range -1 to $2^{\circ}/_{oo}$, with most in the range 0 to $1^{\circ}/_{oo}$. Nozaki et al.(34) report pre-industrial $\delta^{13}C$ values (back to ca. 1770), as laid down in Bermuda corals, of between about -0.5 and $0^{\circ}/_{oo}$, falling towards $-0.8^{\circ}/_{oo}$ in modern times. Druffel(38) reports $\delta^{13}C$ values in the range -1.0 to $0.6^{\circ}/_{oo}$ in coral bands of the Florida Straits laid down since 1642. It is thus quite adequate to treat $\delta^{13}C$ as constant (and we take $\delta^{13}C = 0$) when volume-integrating $[DIO^{14}C]$ of equn (4.6) to form an oceanic inventory :

$$\sum{}^{14}C = \left(\sum{}'{}^{14}C\right)\left(\frac{1 + 10^{-3}\delta^{13}C}{0.975}\right)^2, \tag{4.7}$$

Here, $\Sigma^{14}C$ is $[DIO^{14}C]$ integrated over the oceanic volume, and $\sum{}'{}^{14}C$ is the analogous integration of $[DIO^{14}C]'$.

Inventories already available but computed from $\Delta^{14}C$ values can thus be corrected by means of equn (4.7). An inventory deduced from the Fairhall estimate (4.2) requires an extra factor :

$$[DIO^{14}C]_F = 0.96[DIO^{14}C]_F' \left(\frac{1 + 10^{-3}\delta^{13}C}{0.975}\right)^2 \tag{4.10}$$

The overall multiplier of $[DIO^{14}C]_F'$ is close to unity.

Table 4.3 summarises estimates of radiocarbon inventories, as detailed in preceding sections. Each is adjusted to "uncorrect" for fractionation, with the assumption $\delta^{13}C = 0$.

Table 4.3

Oceanic Radiocarbon Inventories

Source	Reference section	Inventory (10^{26} atoms)
Cosmogenic radiocarbon		
Fairhall et al.	4.3.4	1.93×10^4
Killough and Emanuel	4.3.6	1.99×10^4
Bomb carbon (as of ~1972)		
Stuiver et al. (extrapolated)	4.3.2	310
Broecker et al.	4.3.4	303

Although a consensus of both cosmogenic and bomb-carbon inventories seem evident from table 4.3, it should be noted that :

- both bomb-carbon inventories rely heavily on the GEOSECS data. Their close agreement is partly fortuitous because there are significant differences between the methodology of bomb-carbon segregation and between the distributions so deduced, particularly for the Atlantic Ocan. Furthermore, the inventory for the Stuivery et al. data depends on extrapolation to world oceans from the Atlantic and Pacific oceans in the 50°S - 50°N latitude range.

- the two cosmogenic inventories are obtained in different ways from independently compiled data. Their corroboration is thus reassuring.

In parenthesis, it is appropriate to comment on the correction for fractionation embodied in $\Delta^{14}C$ of (1.4). The inherent assumptions are that disparate $^{13}C/C$ ratios between intimate carbon reservoirs accurately portray fractionation in the nett inter-reservoir transfer of carbon. In fact there may be different causes of $\delta^{13}C$ disparities. For example, Kroopnick(49,51) has interpreted correlations between oceanic $\delta^{13}C$, dissolved oxygen and DIOC as indicating that a significant component of DIOC arises from oxidation of isotopically-lighter organic carbon ($\delta^{13}C = -23°/_{\circ\circ}$). A significant organic source of DIOC would thereby depress $\delta^{13}C$ values and render them unreliable for indicating fractionation during ocean-atmosphere exchange. This consideration does not affect ^{14}C inventories computed from actual or reconstructed $\delta^{14}C$ values.

4.4 Temporal Variations in Oceanic Radiocarbon

"Pre-industrial" and "cosmogenic" ^{14}C distributions have been used as interchangeable terms, reflecting the reality that, to a good approximation, the latter is perpuated largely unchanged in time; the former is therefore just a 19th century sampling of the latter. However, cosmogenic ^{14}C has been slightly redistributed during the industrial era due to isotopic dilution by fossil-fuel carbon.

The oceanic response to the bomb-carbon signal can be expected to follow the circulation pattern outlined in section 4.2. Thus bomb carbon will slowly invade abyssal waters via the N. Atlantic and Antarctic oceans over a time scale of order 500 yrs(23).

For most oceanic regions a comprehensive ^{14}C data set is available for only one isolated time (GEOSECS), precluding a time-series inventory. For certain regions a ^{14}C history is available, notably :

- in regions of banded coral growth (e.g. fig. 4.4).

- in extensive surface surveys over the period 1966-1981 by the Trondheim group(52). These show only a gradual decline, or no discernible decline at all, since the early 1970s, but do indicate strong seasonal effects - also noted by Broecker and Peng(53).

- in regions where NORPAX and TTO surveys have provided a follow-up to the GEOSECS surveys. Broecker *et al.*(24) in their fig 7 show the latitudinal variations in column inventories; their results are compatible with no significant lateral movement of bomb carbon over the period between the surveys.

4.5 Spatial Variations in Oceanic Radiocarbon

The spatial distribution of cosmogenic ^{14}C seems surprisingly uniform throughout the oceans at around $1.4\text{x}10^9$ atoms per litre, probably as a result of delivery rates closely matching the radioactive decay rate (section 4.3.4). Although this reported uniformity seems not to have been widely commented upon, some confirmation comes from noting that the resulting estimated inventory is in concordance with that deduced from Killough's and Emanuel's data (table 4.3).

Figs 4.7, 4.8 represent the best available portrayal (in 1972) of the gross bomb-carbon distribution. The spatial redistribution since that time is not known in detail, but can be qualitatively inferred from known ocean circulation patterns.

5. RADIOCARBON EXCHANGE BETWEEN ATMOSPHERE AND OCEAN

With the oceans representing a thin wafer on the earth's surface (vertical:horizontal dimensions approximately $1:10^4$), the ocean-air interface plays an important role in both carbon balance and marine chemistry. The uppermost (mixed) layer of ocean, averaging some 75m in depth, contains about the same amount of carbon as does the overlying atmosphere, and yet has only about 1.8% of all oceanic carbon(54).

This chapter addresses oceanic carbon dynamics including the atmospheric source. This involves an overview of carbon chemistry, of CO_2 exchange across the air-sea interface, and of models of carbon cycling.

5.1 Oceanic Carbon Chemistry

Only the briefest mention of the most salient aspects of oceanic carbon chemistry are mentioned here. More detail is beyond the scope of the overview, and may be found in refs (54-56) with a summary in ref(1).

Aqueous CO_2 or carbonic acid dissociates as

$$H_2O + CO_2 \rightleftarrows H_2CO_3 \rightarrow H^+ + HCO_3^- \rightleftarrows 2H^+ + CO_3^{2-}, \qquad (5.1)$$

and all components contribute to DIOC with HCO_3^- providing the dominant contribution. The dissociation constants which determine the relative abundance of these ions vary with temperature and pressure and thence with depth. Typical profiles of [DIOC] are displayed in figs 5.1 and 4.6. Table 5.1 records the estimated depth structure of DIOC.

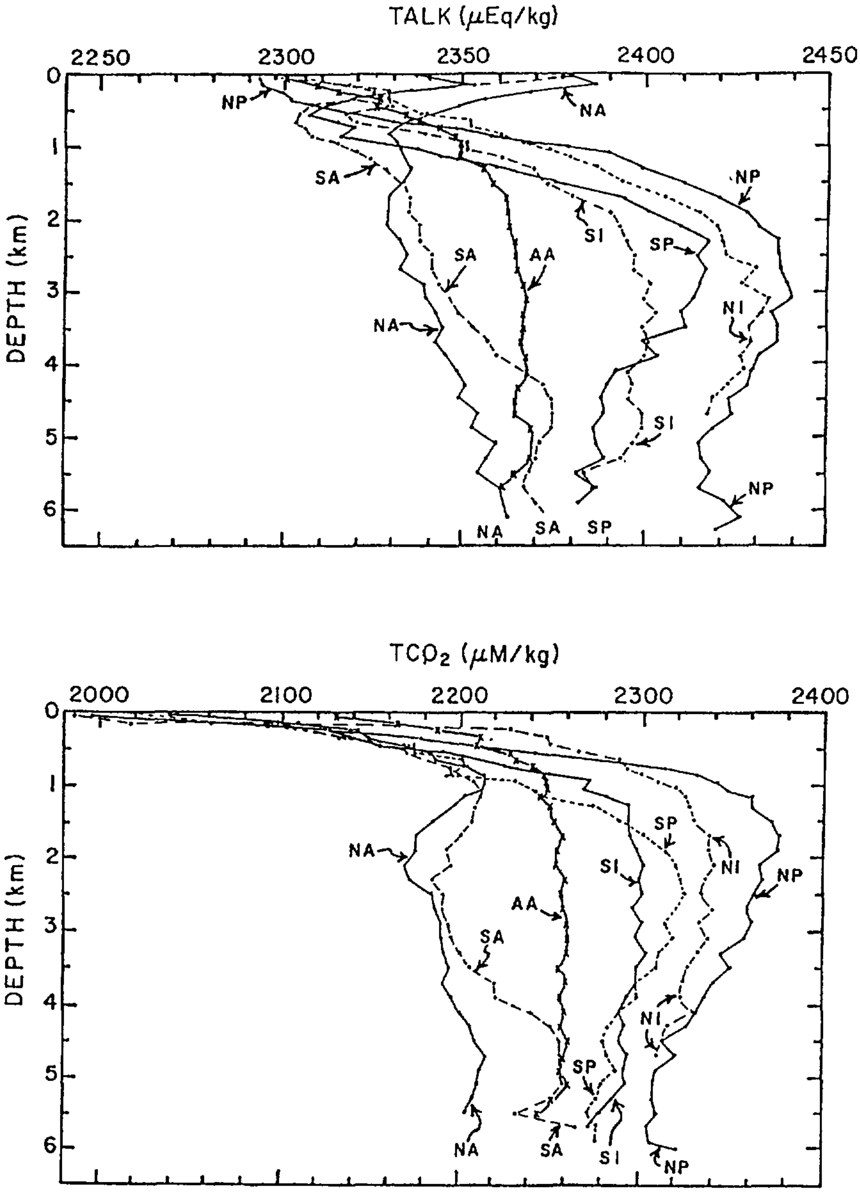

Fig. 5.1: The mean vertical distribution of (a) the alkalinity ("TALK") and (b) the total DIOC concentration ("TCO_2") in the seven regions of the world oceans. NA = North Atlantic, SA = South Atlantic, NP = North Pacific, SP = South Pacific, NI = North Indian, SI = South Indian, and AA = Antarctic region (south of 45°S). The mean values for each depth intervals in the first six oceanic regions are indicated by solid circles, and those in the Antarctic region are indicated by X's. (Taken from Takahashi et al.(58)).

Table 5.1

Oceanic inventory of DIOC*

Depth range (m)	[DIOC] m.mole/kg	Inventory 10^{15} mole (C)
0-50	2.012	37
50-1200	2.181	923
>1200	2.284	2206
whole ocean	2.254	3167

*Table adapted from Takahashi and Azevedo(54)

An important oceanic store of inorganic carbon is calcium carbonate ($CaCO_3$). This material is prevalent in ocean sediments in two forms, calcite and the more soluble aragonite, and its dissolution provides a source of carbonate ions:

$$CaCO_3 \rightleftarrows Ca^{2+} + CO_3{}^{2-}. \qquad (5.2)$$

The saturation concentration for each species at equilibrium is governed by the dissociation constant

$$K = [Ca^{2+}]\ [CO_3{}^{2-}]. \qquad (5.3)$$

The dependence of K upon temperature, pressure and salinity has been studied. The most significant dependence is the monotonic increase of K with pressure, for it ensures that calcite raining from the surface is all dissolved in deep waters. As a result of this, and the deep source of surface water (in the North Atlantic), surface waters are super-saturated in $CaCO_3$ and deep waters under-saturated. The skeletons and shells of many marine organisms are fabricated from the $CaCO_3$ which super-saturates the surface waters. Ultimately - after much surface recycling - the $CaCO_3$ rains downward as detritus to be redissolved at depth. This provides a mechanism for downward transferral of DIOC.

The carbonate and bicarbonate contributions to total oceanic anion concentration are small and are determined by the requirement of charge neutrality. Their contributions are known as the total alkalinity of the water: the alkalinity is the imbalance in ionic charges of strong electrolytes which is balanced only by dissociated weak acids, such as carbonic, boric, phosphoric acids. Alkalinity, denoted [Alk], is a concentration of "equivalents" (molarity weighted by ionic charge).

Thus, ignoring minor constituents,

$$[Alk] = [Na^+] + [K^+] + 2[Mg^{2+}] + 2[Ca^{2+}] - [Cl^-] - [Br^-] - 2[SO_4{}^{2-}] - [NO_3{}^-] \qquad (5.4a)$$

$$= [HCO_3{}^-] + 2[CO_3{}^{2-}]. \qquad (5.4b)$$

The alkalinity is directly measurable and provides a useful oceanic "tracer" (57-58). Typical alkalinity profiles are displayed in fig 5.1.

The partial pressure of CO_2 in the ocean surface, denoted pCO_2, is in equilibrium with the undissociated H_2CO_3; it is defined as the partial pressure of CO_2 in the overlying air under equilibrium conditions (no nett CO_2 transfer). Because most of the DIOC is dissociated a small fractional change in pCO_2 is not accompanied by the same fractional change in [DIOC]; such buffering is described by the "Revelle factor".

Nett CO_2 exchange depends upon the partial pressure difference ΔpCO_2 at the interface (ocean minus atmosphere), and therefore upon pCO_2 in surface waters (since atmospheric circulation homogenenises CO_2 partial pressures there); pCO_2 itself depends not only upon temperature (approximately 4% per °C) but also upon [DIOC] and [Alk]. This favours a nett eflux of CO_2 in regions of upwelling (particularly tropical waters) and nett influx in the downwelling gyre regions. Takahashi and Azevedo(54) have mapped contours of ΔpCO_2. Smethie et al.(59) have measured ΔpCO_2 in the winter tropical North Atlantic (to latitude 15°N), noting that ΔpCO_2 is positive equatorward of about 10° and negative north of that latitude.

It is apparent from fig 5.1 that both [DIOC] and [Alk] are relatively depleted at the surface (by about 10% and 4%, respectively). This depletion is significant in its effect upon pCO_2 and therefore upon CO_2 exchange with the atmosphere. Its origin is in biological activity.

Surface oceans are deficient in nutrients (indeed, are near phosphorus-free) and relatively rich in dissolved oxygen. These features together with depleted DIOC are a result of photosynthetic processes which consume dissolved CO_2 and nutrients and produce oxygen. Such processes deplete DIOC but increase alkalinity. The precipitation of $CaCO_3$ to produce shells and skeletons depletes both DIOC and alkalinity, more than offsetting the photosynthetic increase in the latter. At depth, [DIOC] and [Alk] and produced by the reverse process, producing also enhanced nutrient levels (phosphate and nitrate).

Baes et al.(55) review estimates of the downward flux of organic carbon necessary to account for the export of DIOC and alkalinity from the surface. These flux estimates are in accord with those estimated by Fairhall as necessary to maintain a constant ^{14}C concentration (see section 4.3.4) and with independent estimates by Suess(60).

5.2 Concepts of Atmosphere-Ocean CO_2 Exchange

The flux of CO_2 invading the ocean surface is proportional to the partial pressure of atmospheric CO_2; similarly the evading flux is proportional to pCO_2. These opposing fluxes (also called the invasion and evasion rates) are denoted I and E. But what are the mechanisms by which CO_2 traverses the air-sea interface? At least two conceptual models have been put forward as contending descriptors of such gas exchange. The "stagnant film model" and "film replacement model" are discussed below. A more detailed description of each with quantitative support is supplied by Broecker and Peng(56) [p113ff]. An intercomparison of contending models (with their ancestry) is presented by Holmen and Liss(61).

5.2.1 Stagnant film model

Gas transfer across an air-sea interface is presumed to be limited by molecular diffusion, the limiting step being diffusion across the uppermost millimetre or less of sea surface.

According to the stagnant film model (SFM) a thin film of sea surface of thickness z separates two reservoirs of CO_2, each of uniform but distinct CO_2 partial pressures. The film has a uniform partial pressure gradient across it which drives CO_2 exchange by molecular diffusion. If D is the molecular diffusivity of the gas in sea water then the model predicts evasion and invasion rates proportional to D/z, termed the piston velocity or transfer velocity.

Broecker, Peng and coworkers (56, 62-65) have applied the SFM in a novel manner which they term the "radon method". This method assumes that the SFM describes the evasion of radon gas (radioisotope ^{222}Rn with half-life 3.8 day, a daughter of ^{226}Ra indigenous to all marine waters) with *only* D (but not z) dependent upon the diffusing species. In comparing Rn and CO_2 evasion allowance must also be made for marine dissociation of the latter.

The idea of the radon method is that for seas not severly disturbed over the radon lifetime (about 6 day), marine Rn concentrations reflect equilibrium with the parent. The exception is near the surface (typically throughout the mixed layer and upper thermocline) where Rn concentration is deficient due to its evasion into the overlying air (usually devoid of Rn). The deficit can be determined from Ra and Rn profiles, and the SFM used to infer the equivalent film thickness. This thickness can be used in conjunction with CO_2 diffusivities to estimate CO_2 exchange rates.

Broecker and Peng(66) list values of z for radon evasion at 31 West Atlantic GEOSECS stations between 69°N and 55°S; z ranges from 26 μm to 126 μm, with a tendency for thicker films in equatorial regions. Smethie et al.(59) catalogue recent (TTO) values for D/z and z, calculated by the radon method.

Stuiver(6) analysed radiocarbon data from GEOSECS Atlantic stations, and deduced CO_2 exchange rates from which film thicknesses z are inferred. The latter ranged from 12-66 μm, averaging 19 μm (which corresponds to $E = 27.0$ mole/yr/m^2) for the western Atlantic; and from 15-138 μm, averaging 28 μm ($E = 18.7$ mole/yr/m^2) for the eastern Atlantic. These estimates of z do not employ the radon method, and seem systematically smaller that the radon-deduced z.

Broecker et al.(67) list CO_2 evasion rates calculated using the radon method as well as from bomb-^{14}C and cosmogenic-^{14}C distributions. Whilst agreement is favourable, radon-method evasion rates tend to be smaller. This is attributed either (a) to inadequacies in the SFM, and/or (b) on the basis of the short residency times for radon in the mixed layer compared to the many years for CO_2 including periods of inclement weather. The latter is pertinent because I and E increase significantly with wind speed (perhaps with its square).

5.2.2 Film replacement and other models

There are several alternatives to the SFM of gas transfer, and of these the film replacement model (FRM) has a 50-yr history (mainly in the engineering literature); for a summary of its lineage, see Holmen and Liss(61) and/or Smethie et al.(59). As with the SFM this model envisions molecular diffusion through a film of sea as the rate-limiting process. According to the FRM the film of surface water is periodically replaced by water from the interior which bring with it the properties of the bulk liquid. The rate-determining parameter is the film replacement time, analogous to the SFM's film thickness. The main empirical distinction between the SFM and FRM is that the piston velocities are proportional to D and to $D^{1/2}$ respectively (D being the molecular diffusivity).

Holmen and Liss(61) assumed a piston velocity proportional to D^n and sought an optimal value for n to fit data from their laboratory-tank experiments. They deduced that $n=0.57\pm0.15$, which would support the FRM and appear to rule out the SFM.

Smethie et al.(59) discussed models of gas exchange, noting that the Holmen and Liss result did not preclude $n=2/3$ which has some empirical support. Smethie et al. analysed the radon evasion data using both a D and a $D^{1/2}$ dependence. Both piston velocities and nett CO_2 fluxes are smaller with the $D^{1/2}$ assumption.

5.3 Radiocarbon Balance and the CO_2 Exchange Rate

The dominant source of $DIO^{14}C$ is atmospheric, and the dominant sink is by radioactive decay. This section discusses the balance between the two, particularly in the light of perturbing injections of bomb-carbon into the atmosphere.

The term "exchange rate" refers to either an invasion or evasion rate. In a global steady state nett exchange is nil and the term therefore unambiguous.

Local exchange rates are expected to be proportional to the partial pressure in the departing medium. Invasion rates will therefore have increased by 15% over the industrial era concomitantly with the increased CO_2 partial pressure.

5.3.1 Equilibrium in the pre-bomb era

If one assumes a pre-industrial steady state distribution of ^{14}C, a back-of-envelope calculation can indicate the invading CO_2 flux from the atmosphere necessary to maintain this equilibrium.

Assume a pre-industrial oceanic inventory of 19600 RCU (table 4.3), of which a fraction 1/8267 decay every year - a radioactive loss of 6.6×10^{11} atoms/yr/m^2 of ocean surface. At steady state the average exchanging CO_2 fluxes (I and E) exactly cancel, but because of an imbalance in $^{14}C/^{12}C$ ratio there is a nett air-to-sea ^{14}C flux of

$$1.176 \times 10^{-12} \times 1.052 \times 10^{-3} \times \{\Delta^{14}C(\mathrm{air}) - \Delta^{14}C(\mathrm{surface})\} \times E\ N_0,$$

where N_0 is Avagadro's number and E is in mole(CO_2)/yr/m^2. The factor 1.052 accounts for fractionation(6). Equating source and sink and setting interfacial $\Delta^{14}C$ increment at 45°/₀₀ provides E = 19.7 mole/yr/m^2. This can be expressed in terms of the average residence time for atmospheric CO_2 with respect to oceanic invasion, which for the pre-industrial atmosphere is 7.2 yr. The major uncertainty in this estimate is in the $\Delta^{14}C$ increment at the air-sea interface.

5.3.2 Response of the bomb-carbon signal

The invading bomb carbon provides a rare opportunity to study the dynamics of CO_2 exchange at the ocean-air interface. This opportunity has two caveats. First, time-series data of the invasion is not available, save one exception, because extensive surveys have yet to follow up GEOSECS. The exception is the excellent data documented by Nydal et al.(52), so far incompletely analysed. The second caveat is the caution by Broecker et al.(68) that equilibration times for isotopes of carbon are longer by an order of magnitude than CO_2 residence times in the surface ocean and atmosphere. Hence the distribution of DIO^{14}C among the chemical species in the ocean (bicarbonate and carbonate ions) may not mirror the distribution of DIOC. Such a consideration would affect estimated evasion rates more than invasion rates, and even then not in the pre-1972 era with large atmospheric $\Delta^{14}C$ values.

Some exchange rate estimates from bomb-carbon distributions are outlined below; others deduced by the radon method are reported in section 5.2.1.

(a) Munnich and Roether(69):

This early calculation based on a 1965 Atlantic survey assumes that bomb-carbon and tritium have penetrated to the same depths. An average CO_2 piston velocity of 2.1 km/yr (5.8 m/day) is indicated, corresponding to an exchange rate of 23 mole/yr/m^2.

(b) Stuiver(70):

Stuiver summarises available estimates of atmospheric residence times based on early bomb-carbon data. These range from 4 to 10 yrs.

(c) Stuiver(6):

The GEOSECS Atlantic results are analysed separately for the eastern and western Atlantic. The bomb component $\Delta^{14}C^*$ is segregated from measured values by assuming a pre-industrial surface value -40°/₀₀ and an equal penetration of bomb carbon and tritium. By assuming that the bomb-carbon column inventories are derived solely from the overlying air of known history (i.e. none is by lateral advection), an exchange rate can be estimated. The average exchange rate for the western and eastern Atlantic is 27.0 and 18.7 mole/yr/m^2, with an Atlantic average of 23 mole/yr/m^2. This corresponds to an atmospheric residence time of 6.8 yr.

(d) Quay and Stuiver(22):

Quay and Stuiver put forward a model of vertical transport to intepret GEOSECS data in and overlying the thermocline. Bomb carbon invades the sea surface and is mixed by vertical advection and diffusion. The CO_2 exchange rate matches input to inventory; the area-weighted rate for the Atlantic is then 21 mole/yr/m^2, and that for the Pacific (with sparser data) is about 25 mole/yr/m^2. However, unlike the Atlantic, the Pacific bomb-carbon distribution is not well explained by vertical transport alone.

(e) Stuiver _et al_.(25):

A preliminary analysis as in (c) above of mid-Atlantic and mid-Pacific GEOSECS results provide average exchange rates in the tropical and gyre regions as reported in table 4.1.

(f) Broecker _et al_.(24):

Broecker _et al_. deduce ocean-average CO_2 exchange rates separately for the Atlantic, Pacific and Indian Oceans of 22.3, 19.4 and 19.2 mole/yr/m^2; see section 4.3.5 for details. The area-weighted average for world oceans is 20.1 mole/yr/m^2 which, by design, matches the bomb-carbon inventory to accumulated input. The temperate regions together with the North Atlantic show an inventory excess over apportioned input (fig 4.9b). This is explainable in part by local exchange rates depending upon average wind speeds. For reported wind speed dependences, Broecker _et al_. reassessed local invasion rates, which are reported in table 5.2. This partly accounts for the "Inventory-Input" variations in table 4.2b.

Table 5.2

Regional invasion rates corrected for average wind speeds*

Region #	Invasion Rate (mole/yr/m^2)	Inventory/Input+
Atlantic Ocean	22.3	1.0
Northern	26	1.0
N. temperate	17	1.9
Tropical	15	0.6
S. temperate	22	1.2
Antarctic	≈26	0.3
Indian Ocean	19.4	1.0
Tropical	13	0.9
S. temperate	22	1.8
Antarctic	≈26	0.5
Pacific Ocean	19.2	1.0
Northern	23	0.5
N. temperate	19	1.4
Tropical	14	0.9
S. temperate	22	1.3
Antarctic	≈26	0.5

* Table from Broecker et al.(24)

\# The latitude bands are those of table 4.2b

\+ The inventories are those of table 4.2b; the inputs are recalculated from table 4.2b using a wind-corrected invasion rate.

(g) Smethie et al.(59):

Smethie et al. report extensive winter measurements of pCO_2 and of radon profiles in the tropical Atlantic surface (the TTO survey). The piston velocities, estimated by the radon method, increase with wind speed. The differential in CO_2 partial pressure at the sea-air interface (ΔpCO_2) vary between -35 μatm and 64 μatm, with null value at near 10°N. North of this latitude the nett CO_2 flux into the ocean peaked at 1.4 mole/yr/m^2 (at 15°N), and the tropical peak outflow was 2.7 mole/yr/m^2 at 8°S. The separate invading and evading fluxes were not estimated.

5.3.3 Sources of CO_2-exchange variation

Nett CO_2 fluxes from atmosphere to ocean are known to depend upon marine conditions (temperature, salinity, pCO_2) and upon the prevailing wind speed. The mechanism for the latter seems not to be well understood, conceptually or empirically. In laboratory experiments Kanwisher(71) has deduced empirically an exchange rate varying with the square of wind speed; however such a dependence seems to be only roughly applicable to oceanic conditions(59, 65-67, 72).

5.4 Models of the Global Carbon Cycle

It is unnecessary to undertake detailed carbon-cycle modelling in order to estimate radiocarbon inventories and dynamics in the oceans. However, it is useful to appraise the overall consistency of apportioned carbon and radiocarbon. Accordingly, only the main features and developments of global carbon models are mentioned, including their successes and shortcomings in respect of radiocarbon and oceanic DIOC.

5.4.1 Model structures

The major model developments are outlined below. For more detail, see the review by Emanuel et al.(73).

The simplest models contain a few simply-connected boxes, with the oceans usually represented by two boxes, surface and deep ocean; only the surface ocean box is connected to the atmosphere box in such models. Craig (74) and Rafter and O'Brien(9) used such a model structure to estimate turnover times. Such models have simplicity as their major virtue, but do not well represent DIOC transfer to deep water. If carefully selected, a two-box ocean model remains a useful interpretive tool(75).

The "Oeschger model" (76) has become the benchmark model for oceanic carbon transport - although some reparameterisations have been investigated (45, 47, 77). The deep ocean is given vertical structure by means of a reservoir whose carbon is transported diffusively. The Oeschger model has 5 reservoirs: atmosphere, long-term biosphere (with 60-yr lag), well-mixed surface ocean, eddy-diffusive deep ocean. The surface ocean is chosen as 75m deep (the accepted average depth of the mixed layer). Other parameters are selected to provide a best fit to the steady-state cosmogenic ^{14}C distribution: including a vertical diffusivity of 1.26 cm^2/s. Diffusion-controlled deep mixing seems to introduce a superior simulation of DIOC distribution without sacrificing structural simplicity. Such diffusion is seen as an empirical mimic of carbon transport to depth rather than as a conceptual portrayal. Siegenthaler and Oeschger (77) use the model to predict the response to fossil-fuel combustion; some predictions depend markedly upon assumed values for the Revelle factor and for the poorly-known biospheric growth rate. Broecker et al.(47) propose a retuning of parameters and note that tritium-penetration data favour a larger diffusivity (~1.7 cm^2/s). Broecker et al.(45) note that increasing the depth of the surface-ocean box can improve the model's fit to fossil-fuel data.

Several investigators have modified the Oeschger model in attempts to inject more realism into its structure or to test if model shortcomings were responsible for imperfect simulations. Broecker et al.(47) and Hoffert et al.(78) independently introduced vertical advection into the model, with separate upwelling and downwelling regions. Hoffert et al. also included effects of a marine biosphere. Crane(79) and Siegenthaler(80) added boxes to represent polar regions which outcrop to make contact with the atmosphere.

In numerically implementing box-diffusion models to study carbon dynamics, Oeschger found it appropriate to stratify the diffusive reservoir into a finite number (viz, 42) of layers. Several recent models have explicitly incorporated a multi-box structure in an attempt to better simulate real oceanic processes. Bjorkstrom(81) designed a model with a "cold" and "warm" surface ocean, a stratified (2 layers) intermediate ocean simulating the thermocline, and a deep ocean stratified into 8 boxes. Enting and Pearman(13) developed a similar but more complex compartmental structure. Some other variations are developed for comparative purposes (see succeeding section).

5.4.2 Model intercomparisons

Two recent studies(43, 82) compare the predictive prowesses of differently structured carbon cycle models. Both studies select representative models and calibrate them consistently (so that differences in their simulations are truly indicative of differences in their structures); the calibration requires that the models reproduce the steady-state pre-industrial distribution of radiocarbon (as well as it is known). Both intercomparisons have as objective a study of model responses to fossil-fuel combustion scenarios. A summary of these intercomparisons follows; more detail is provided by Emanuel et al.(73).

Bacastow and Bjorkstrom(82) present and compare four structures of oceanic models. A major interest was in the forecasted "airborne fraction" of combusted fossil fuel following scenarios of fossil-fuel combustion proceeding into the next century. For near-term forecasts an Oeschger model seems satisfactory but with a higher diffusivity than originally proposed. For longer term forecasting, a retuned Oeschger model should be appropriate; this would feature a deeper surface ocean (201m) and diffusivity $K = 1.437\ cm^2/s$, selected to simulate deep ^{14}C.

Killough and Emanuel(43) compare five unidimensional models. For model calibration the cosmogenic ^{14}C data base discussed in section 4.3.6 was carefully assembled. The performances of the models in predicting uptake of excess (fossil fuel) CO_2 are compared. All of the models underpredict the current airborne fraction of combusted fossil fuels, the underproduction being substantial in the case of simple 2-box oceanic models. The most successful models seem to include some direct vertical transport (upwelling/downwelling) to give deep oceans access to surface DIOC levels.

5.4.3 Model overview

Emanuel et al(73) summarise the two intercomparison studies from the viewpoint of understanding the fate of atmospheric CO_2. They suggest that the important features of a model which govern how quickly excess CO_2 is taken up by the oceans is the effective size of those reservoirs which are coupled with short time constants to the atmosphere. The important interplay between model structure and calibration is stressed. Diffusion can be entirely replaced by advective transport among stratified levels and, suitably calibrated, both can provide equally good accounts of steady state distributions. But responses on the different time scales can differ. It is not apparent that model response on the fossil-fuel time scale (several decades) necessarily reflects model response to the sharp bomb carbon signal; however, the relative responses of the Killough and Emanuel models to both inputs are similar.

The CO_2 exchange rate is not pinned down by these analyses. There is a close coupling between that rate and parameters which describe the rate of transfer to the deep ocean. For example, both Oeschger et al.(76) and Broecker et al.(47) demonstrate the coupling between the atmospheric exchange rate and the deep-ocean diffusivity. Thus it seems impossible to specify an average exchange (or invasion) rate to better than 25% (typically, 19±5 mole/yr/m^2); for modelling applications its selection will be in tandem with those of other rate constants. The invasion rate should be expected to have increased by ~15% since the mid 19th century due to the increased atmospheric CO_2 content. Hence a pre-industrial invasion rate of 19 mole/yr/m ^{2} is equivalent to perhaps 22 mole/yr/m^2 for the modern atmosphere (i.e. to 0.065 mole/yr/m^2/μatm). The evasion rate will have undergone a similar (but slightly retarded) increase due to the close coupling and comparable sizing of the exchanging reservoirs.

5.5 Post-1972 Bomb-Carbon Inventory : An Update

The bomb-carbon inventory of table 4.3 is as of 1972. Since that time atmospheric bomb carbon has continued to invade the oceans. We can estimate the post-1972 invasion in a manner similar to Stuiver(6).

The nett flux of bomb ^{14}C from air to surface ocean is

$$(1.176\times10^{-12})(1.052)(10^{-3})\{I\times\Delta^{14}C^*_{air} - E\times\Delta^{14}C^*_{surf}\}$$

mole/yr/m^2. The factor 1.052 is due to relating fractionation to a normalised value of $\delta^{13}C = -25°/_{oo}$. We assume that the globally-averaged invasion and evasion rates (I and E) balance each other. This incurs slight error in that each is increasing due to the increasing CO_2 partial pressures in the departing medium. To select a representative invasion rate, we update the area-weighted average 20.1 mole/yr/m^2 from the Broecker et al. analysis (section 5.3.2(f)): a 2.5% enhancement to I = E = 20.6 mole/yr/m^2 acknowledges the 5% increase in CO_2 partial pressures over 1972-85.

The cumulative nett input of ^{14}C to world oceans since 1972 is then

$$5.53\times10^{24} \int_{1972}^{1985} \{\Delta^{14}C^*_{air}(t) - \Delta^{14}C^*_{surf}(t)\}dt \text{ atoms.}$$

The integral over $\Delta^{14}C^*_{air}$ is determined by noting that the post-1972 atmospheric record (fig. 3.1) is closely (and universally) approximated by an exponential decline from 470°/₀₀ at a rate of 6.1% p.a. This integral then equates to 4219°/₀₀-yr (equivalent to a 1972-85 average $\Delta^{14}C^*_{air}$ = 325°/₀₀).

Globally-averaged $\Delta^{14}C^*_{surf}$ values are more elusive. Broecker et al.(24) have reported empirical regional averages; area-weighting these provides a global average $\Delta^{14}C^*_{surf}$ (1972) = 157°/₀₀. The coral record adopted by Broecker et al. shows surface $\Delta^{14}{}^*C$ values decreasing almost linearly since 1972 to 80% of that 1972 value by 1985. Accepting such a linear decline, the time-integral over $\Delta^{14}C^*_{surf}(t)$ is 1840°/₀₀-yr (equivalent to an average value of 141°/₀₀).

With the above values the cumulative nett input of ^{14}C during 1972-85 is 132 RCU. Thus, supplementing the 1972 estimated inventory (table 4.3) the bomb-carbon inventory in the oceans as of 1985 is estimated to be 435 RCU.

It is of interest to check the atmospheric inventory change for consistency. It is immediately clear from fig. 3.1 and table 3.1 that estimates of total atmospheric (tropospheric + stratospheric) inventory require extrapolation of the pre-1969 stratospheric estimates. At best, such extraplation can be guided by model forecasts. At worst it is complicated by the slow intra-stratospheric mixing which means that even the pre-1969 measurements on which table 3.1 is based may not typify the entire stratosphere (see section 3.3). Thus the atmospheric inventories of Levin et al.(14) and Manning et al.(15) reported in table 3.1 are subject to perhaps 25% uncertainty, and are more likely to be under-estimates than overestimates. The 1972-1985 inventory decline is thus 101±25 RCU. Such uncertainty remains small compared with the cosmogenic inventory of 400 RCU.

Atmospheric input during 1972-1985 consisted of 9 RCU from nuclear detonations (at 1.2 RCU per Mt yield). The nett atmospheric exodus during this period was thus 110±25 RCU of bomb ^{14}C; this should be compared with the estimated nett input of 132 RCU to the oceans, leaving an atmospheric input of 22±25 RCU unaccounted for. Part of this discrepancy is due to exchange with the terrestial biosphere. While the magnitude of such exchange (even its sign) has not been reported, we note that Broecker et al.(47) estimated bomb-^{14}C inventories for 1965 and 1969 in the various reservoirs and that these inventories required a nett eflux from biosphere to atmosphere. A continuation of this trend is concordant with the above estimates for 1972-1985.

6. SUMMARY

From the information reviewed above, the oceanic inventory for natural ^{14}C is 19,600 RCU (table 4.3) and this inventory is similar to estimates predicted by other methods. The natural production rate of ^{14}C in the atmosphere is currently about 3 RCU/yr. By 1950 the natural atmospheric ^{14}C specific activity had declined by about 3% as a result of the burning of fossil fuels, a decline commencing in the 19th century (Suess effect).

The 1972 oceanic inventory of bomb-produced ^{14}C is estimated at 303 RCU (table 4.3), some 55% of a total production of about 550 RCU. At that time at least 181 RCU was airborne (table 3.1) and presumably up to 65 RCU had entered the terrestrial biosphere. Between 1972 and 1985 a further 132 RCU has been added to the oceanic burden (secion 5.5) to provide a 1985 inventory there of 435 RCU. Some 90 RCU still remain in the 1985 atmosphere, declining at about 6.1% p.a.

However there will be an increasing input of man-made ^{14}C discharged into the atmosphere from the nuclear power industry. Current production is about 0.5 RCU/yr, some 17% of the natural ^{14}C production rate. Although small compared with current bomb ^{14}C inventories, it will assume more significance in the future if the nuclear power industry expands significantly.

The exchange rate of radiocarbon between the atmosphere and the ocean can be inferred from models of cosmogenic ^{14}C redistribution or from empirical studies of CO_2 exchange at the interface, Both approaches are consistent with an atmospheric residence time of about 7 yr and an average invasion rate to the oceans of 20 mole/m^2/yr. The current atmospheric inventory decline for bomb produced ^{14}C suggests an oceanic input of 6 RU/yr.

As can be seen from fig. 4.7, the oceanic distribution of bomb ^{14}C in surface water is far from uniform. Generally concentrations are higher in low to mid latitudes, particularly in those areas with a relatively shallow thermocline. Concentrations are lower in areas of upwelling and where there is strong convective mixing with deep ocean waters as in the oceans surrounding Antarctica. Figs 4.5 and 4.6 indicate that although bomb ^{14}C had mixed into the surface ocean and into the thermocline regions by 1970, much of the deep ocean remained uncontaminated.

Over the next few centuries present variations in oceanic ^{14}C distribution will become smooth and will ultimately reflect the age structure of ocean waters. The present concentration of bomb-carbon in the atmosphere and surface oceans will decline until the bomb-carbon signal is reduced to a global 2.5% elevation in natural ^{14}C level before being lost to statistical precision.

The GEOSECS survey of oceanic ^{14}C gave a very good snapshot of its distribution in the 1970s. There is a need for further surveys in the future to verify the time evolution of ^{14}C in the world's oceans. A continuing international effort is also needed to monitor ^{14}C in the atmosphere in order to verify future releases from the nuclear power industry and in this respect monitoring the difference between the northern and southern hemispheres is particularly relevant.

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REFERENCES

1 Lassey, K.R., Manning, M.R. and O'Brien, B.J., " An assessment of the inventory of carbon-14 in the oceans", Institute of Nuclear Sciences (NZ) Report INS-R--348, 1986.

2 Stuiver, M., "Workshop on ^{14}C reporting", Radiocarbon, 22, 964-966, 1980.

3 Stuiver, M. and Polach, H.A., "Reporting of ^{14}C data", Radiocarbon, 19, 355-363, 1977.

4 Bolin, B., Bjorkstrom, A., Keeling, C.D., Bacastow, R. and Siegenthaler, U., "Carbon Cycle Modelling", in Carbon Cycle Modelling (SCOPE 16), edited by B. Bolin, pp1-28, John Wiley, New York, 1981.

5 O'Brien, K., "Secular variations in the production of cosmogenic isotopes in the earth's atmosphere", J. Geophys. Res. 84, 422-431, 1979.

6 Stuiver, M., "^{14}C distribution in the Atlantic Ocean", J. Geophys. Res., 85, 2711-2718, 1980.

7 Stuiver, M. and Quay, P.D., "Atmospheric ^{14}C changes resulting from fossil fuel CO_2 release and cosmic ray flux variability", Earth Planet. Sci. Lett. 53, 349-362, 1981.

8 Telegadas, K., "The seasonal atmospheric distribution of excess carbon 14 from March 1955 to July 1969", in Health and Safety Lab. Rep. HASL-243, USAEC, 1971.

9 Rafter, T.A. and O'Brien, B.J., "Exchange rates between the atmosphere and the ocean as shown by recent ^{14}C measurements in the South Pacific", in Radiocarbon Variations and Absolute Chronology (Nobel Symp. 12), edited by I.U. Olsson, pp355-377, John Wiley, New York, 1970.

10 Glasstone, S. and Dolan, P.J., "The effects of nuclear weapons", Castle House Publications, Tunbridge Wells, England, 653pp, 1977.

11 Machta, L., List, R.J., and Telegadas, K., "Meteorology of fallout from 1961-1962 nuclear test", Congress of the United States, Hearing before subcommittee in Research, Development and Radiation of the joint committee of Atomic Energy, 88th Congress 46-61, June 1963.

12 UNSCEAR, "Sources and Effects of Ionizing radiation, report of the United Nations Scientific Committee on the Effects of Atomic Radiation, p119, United Nations, New York, 1977.

13 Enting, I.G. and Pearman, G.I., "Refinements to a one-dimensional carbon cycle model", Div. Atmos. Res., (Technical Paper No. 3), CSIRO, Canberra, Australia, 1983.

14 Levin, I., Kromer, B., Schoch-Fischer, H., Bruns, M., Munnich, M., Berdau, D., Vogel, J.C. and Munnich, K.O., "25 years of tropospheric ^{14}C observations in central Europe", Radiocarbon, 27, 1-19, 1985.

[15] Manning, M.R., Jansen, H.S., McGill, R.C., and Burr, M.K., "A summary of atmospheric carbon 14 measurements made in the South Pacific from 1954 to 1985", Institute of Nuclear Sciences, DSIR, New Zealand, (in preparation) 1986.

[16] UNSCEAR, "Ionizing Radiation Sources and Biological Effects", report of the United Nations Scientific Committee on the Effects of Atomic Radiation, p259, United Nations, New York, 1982.

[17] Stuiver, M. and Ostlund, H.G., "GEOSECS Atlantic Radiocarbon", Radiocarbon, 22, 1-24, 1980.

[18] Ostlund, H.G. and Stuiver, M., "GEOSECS Pacific Radiocarbon", Radiocarbon, 22, 25-53, 1980.

[19] Stuiver, M. and Ostlund, H.G., "GEOSECS Indian Ocean and Mediterranean Radiocarbon", Radiocarbon, 25, 1-29, 1983.

[20] Broecker, W.S., Peng, T.-H. and Stuiver, M., "An estimate of the upwelling rate in the equatorial Atlantic based on the distribution of bomb radiocarbon", J. Geophys. Res., 83, 6179-6186, 1978.

[21] Wunsch, C., "An estimate of the upwelling rate in the equatorial Atlantic based on the distribution of bomb radiocarbon and quasi-geostrophic dynamics", J. Geophys. Res., 89, 7971-7978, 1984.

[22] Quay, P. D. and Stuiver, M., "Vertical advection-diffusion rates in the oceanic thermocline determined from ^{14}C distributions", Radiocarbon, 22, 607-625, 1980.

[23] Stuiver, M., Quay, P.D. and Ostlund, H.G., "Abyssal water carbon-14 distribution and the age of the world oceans", Science, 219, 849-851, 1983.

[24] Broecker, W.S., Peng, T.-H., Ostlund, G. and Stuiver, M., "The distribution of bomb radiocarbon in the ocean", J. Geophys. Res., 90, 6953-6970, 1985.

[25] Stuiver, M., Ostlund, H.G. and McConnaughey, T.A., "GEOSECS Atlantic and Pacific ^{14}C distribution", in *Carbon Cycle Modelling* (SCOPE 16), edited by B. Bolin, pp201-221, John Wiley, New York, 1981.

[26] Quay, P. D., Stuiver, M. and Broecker, W.S., "Upwelling rates for the equatorial Pacific Ocean derived from the bomb ^{14}C distribution", J. Mar. Res., 41, 769-792, 1983.

[27] Fairhall, A.W., Young, A.W. and Bradford, P.A., "Radiocarbon in the sea", in proc. *8th Int. Conf. on Radiocarbon Dating*, Wellington N.Z., pp C2-C42, Royal Soc. of N.Z., 1972.

[28] Fairhall, A.W., "Radiocarbon in the sea", Prog. Rep. 15/Aug/68-1/May/71, University of Washington Rep. RLO-2225-T20-3 (avail. from NTIS), 1971.

[29] Fairhall, A.W., "Radiocarbon in the sea", Prog. Rep. 15/Aug/68-1/May/74, University of Washington Rep. RLO-2225-T20-10 (avail. from NTIS), 1974.

[30] Fairhall, A.W., "Radiocarbon in the sea", Final Rep., University of Washington Rep. RLO-2225-T20-12 (avail. from NTIS), 1975.

[31] Fairhall, A.W. and Young, A.W., "Historical ^{14}C measurements for the Atlantic, Pacific and Indian Oceans", Radiocarbon, 27, 473-507, 1985.

[32] Broecker, W.S. and Peng, T.-H., "A strategy for the development of an improved model for the uptake of fossil fuel CO_2 by the ocean", in *Carbon Cycle Modelling* (SCOPE 16), edited by B. Bolin, pp223-226 John Wiley, New York, 1981.

[33] Gordon, A.L., "Interocean exchange of thermocline water", J. Geophys. Res., 91, 5037-5046, 1986.

[34] Nozaki, Y., Rye, D.M., Turekian, K.K. and Dodge, R.E., "A 200 year record of carbon-13 and carbon-14 variations in a Bermuda coral", Geophys. Res. Lett., 5, 825-828, 1978.

[35] Druffel, E.M. and Linick, T.W., "Radiocarbon in annual coral rings of Florida", Geophys. Res. Lett., 5, 913-916, 1978.

[36] Druffel, E.M., "Radiocarbon in annual coral rings of Belize and Florida", Radiocarbon, 22, 363-371, 1980

[37] Druffel, E.M., "Radiocarbon in annual coral rings from the eastern tropical Pacific Ocean", Geophys. Res. Lett., 8, 59-62, 1981.

[38] Druffel, E.M., "Banded corals: changes in oceanic carbon-14 during the little ice-age", Science, 218, 13-19, 1982.

[39] Druffel, E.M. and Suess, H.E., "On the radiocarbon record in banded corals: exchange parameters and net transport of $^{14}CO_2$ between atmosphere and surface ocean", J. Geophys. Res., 88, 1271-1280, 1983.

[40] Broecker, W.S., Gerard, R., Ewing, M., and Heezen, B.C., "Natural radiocarbon in the Atlantic Ocean", J. Geophys. Res., 65, 2903-2931, 1960.

[41] Bien, G. and Suess, H., "Transfer and exchange of ^{14}C between the atmosphere and the surface water of the Pacific Ocean", in Radioactive Dating and Methods of Low-level Counting, pp105-115, IAEA, Vienna, 1967.

[42] Linick, T.W., "La Jolla measurements of radiocarbon in the oceans", Radiocarbon, 20, 333-359, 1978.

[43] Killough, G.G. and Emanuel, W.R., "A comparison of several models of carbon turnover in the ocean with respect to their distributions of transit time and age, and responses to atmospheric CO_2 and ^{14}C", Tellus, 33, 274-290, 1981.

[44] Broecker, W.S. and Li, Y.-H., "Interchange of water between the major oceans", J. Geophys. Res., 75, 3545-3552, 1970.

[45] Broecker, W.S., Takahashi, T., Simpson, H.J. and Peng, T.-H., "Fate of fossil fuel carbon dioxide and the global carbon budget", Science, 206, 409-418, 1979.

[46] Craig, H., "Abyssal carbon and radiocarbon in the Pacific", J. Geophys. Res., 74, 5491-5506, 1969.

[47] Broecker, W.S., Peng, T.-H. and Enge, R., "Modeling the carbon system", Radiocarbon, 22, 565-598, 1980.

[48] Peng, T.-H., "Uptake of anthropogenic CO_2 by lateral transport models of the ocean based on the distribution of bomb-produced ^{14}C", Radiocarbon, 28, 363-375, 1986.

[49] Kroopnick, P., "Correlations between ^{13}C and CO_2 in surface waters and atmospheric CO_2", Earth Planet. Sci. Lett., 22, 397-403, 1974.

[50] Kroopnick, P., "The distribution of ^{13}C in the Atlantic Ocean", Earth Planet. Sci. Lett., 49, 469-484, 1980.

[51] Kroopnick, P., "The dissolved O_2-CO_2-^{13}C system in the eastern equatorial Pacific", Deep-Sea Res., 21, 211-227, 1974.

[52] Nydal, R., Gulliksen, S., Lovseth, K. and Skogseth, F.H., "Bomb ^{14}C in the ocean surface 1966-1981", Radiocarbon, 26, 7-45, 1984.

[53] Broecker, W.S. and Peng, T.-H., "Seasonal variability in the $^{14}C/^{12}C$ ratio for surface ocean water", Geophys. Res. Lett., 7, 1020-1022, 1980.

54 Takahashi, T. and Azevedo, A.E.G., "The oceans as a CO_2 reservoir", in Interpretation of Climate and Photochemical Models, Ozone and Temperature, edited by R.A. Peck and J.R. Hummel, pp83-109, Amer. Inst. Physics conf. proc. no. 82, 1982.

55 Baes, C.F., Bjorkstrom, A. and Mulholland, P.J., "Uptake of carbon dioxide by the oceans", in Atmospheric Carbon Dioxide and the Global Carbon Cycle, edited by J.R. Trabalka, pp81-111, USDoE Rep. DOE/ER-0239, 1985.

56 Bien, G. and Suess, H., "Transfer and exchange of ^{14}C between the atmosphere and the surface water of the Pacific Ocean", in Radioactive Dating and Methods of Low-level Counting, pp105-115, IAEA, Vienna, 1967.

57 Fiadeiro, M., "The alkalinity of the deep Pacific", Earth Planet. Sci. Lett. 49, 499-505, 1980.

58 Takahashi, T., Broecker, W.S. and Bainbridge, A.E., "The alkalinity and total carbon dioxide concentration in the world oceans", in Carbon Cycle Modelling (SCOPE 16), edited by B. Bolin, pp271-286, John Wiley, New York, 1981.

59 Smethie, W.M., Takahashi, T. and Chipman, D.W., "Gas exchange and CO_2 flux in the tropical Atlantic Ocean determined from ^{222}Rn and pCO_2 measurements", J. Geophys. Res., 90, 7005-7022, 1985.

60 Suess, E., "Particulate organic carbon flux in the oceans - surface productivity and oxygen utilization", Nature, 288, 260-263, 1980.

61 Holmen, K. and Liss, P., "Models for air-water gas transfer: an experimental investigation", Tellus, 36B, 92-100, 1984.

62 Broecker, W.S. Li, Y.H. and Cromwell, J., "Radium-226 and Radon-222: concentrations in Atlantic and Pacific Oceans", Science, 158, 1307-1310, 1967.

63 Broecker, W.S. and Peng, T.-H., "The vertical distribution of radon in the BOMEX area", Earth Planet. Sci. Lett., 11, 99-108, 1971.

64 Peng, T.-H., Takahashi, T. and Broecker, W.S., "Surface radon measurements in the North Pacific Ocean station PAPA", J. Geophys. Res., 79, 1772-1780, 1974.

65 Peng, T.-H., Broecker, W.S., Mathieu, G.G. and Li, Y.-H., "Radon evasion rates in the Atlantic and Pacific Oceans as determined during the GEOSECS program", J. Geophys. Res., 84, 2471-2486, 1979.

66 Broecker, W.S. and Peng, T.-H., "Gas exchange rates between air and sea", Tellus, 26, 21-35, 1974.

67 Broecker, W.S., Peng, T.-H., Mathieu, G., Hesslein, R. and Torgersen, T., "Gas exchange rate measurements in natural systems", Radiocarbon, 22, 676-683, 1980.

68 Broecker, W.S., Peng, T.-H. and Takahashi, T., "A strategy for the use of bomb-produced radiocarbon as a tracer for the transport of fossil fuel CO_2 into the deep-sea source regions", Earth Planet. Sci. Lett., 49, 463-468, 1980.

69 Munnich, K.O. and Roether, W., "Transfer of bomb ^{14}C and tritium from the atmosphere to the ocean. Internal mixing of the ocean on the basis of tritium and ^{14}C profiles", in Radioactive Dating and Methods of Low-level Counting, pp93-104, IAEA, Vienna, 1967.

70 Stuiver, M., "The ^{14}C cycle and its implications for mixing rates in the ocean-atmosphere system", in Carbon and the Biosphere, edited by G.M. Woodwell and E.V. Pecan, pp6-20, USAEC Rep. CONF-720510.

[71] Kanwisher, J., "On the exchange of gases between the atmosphere and the sea", Deep-Sea Res., 10, 195-207, 1963.

[72] Wannikhof, R., Ledwell, J. and Broecker, W.S., "Gas exchange - wind speed relation measured with sulfur hexafluoride on a lake", Science, 1224-1226, 1985.

[73] Emanuel, W.R., Fung, I.Y.-S., Killough, G.G., Moore, B. and Peng, T.-H., "Modeling the global carbon cycle and changes in the atmospheric carbon dioxide levels", in Atmospheric Carbon Dioxide and the Global Carbon Cycle, edited by J.R. Trabalka, pp141-173, USDoE Rep. DOE/ER-0239, 1985.

[74] Craig, H., "The natural distribution of radiocarbon and the exchange time of carbon dioxide between atmosphere and sea", Tellus, 9, 1-17, 1957.

[75] Bacastow, R.B. and Keeling C.D., "Models to predict future atmospheric CO_2 concentrations" in proc. Workshop on the Global Effects of Carbon Dioxide from Fossil Fuels, edited by W.P. Elliot and L. Machta, pp72-90, USDoE Rep. CONF-770385, 1979.

[76] Oeschger, H., Siegenthaler, U., Schotterer, U. and Gugelmann, A., "A box diffusion model to study the carbon dioxide exchange in nature", Tellus, 27, 168-192, 1975.

[77] Siegenthaler, U. and Oeschger, H., "Predicting future atmospheric carbon dioxide levels", Science, 199, 388-395, 1978.

[78] Hoffert, M.I., Callegari, A.J. and Hsieu, C.-T., "A box-diffusion carbon cycle model with upwelling, polar bottom water formation and a marine biosphere", in Carbon Cycle Modelling (SCOPE 16), edited by B. Bolin, pp287-305, John Wiley, New York, 1981.

[79] Crane, A.J., "The partitioning of excess CO2 in a five-reservoir atmosphere-ocean model", Tellus, 34, 398-401, 1982.

[80] Siegenthaler, U., "Uptake of excess CO_2 by an outcrop-diffusion model of the ocean", J. Geophys. Res., 88, 3599-3608, 1983.

[81] Bjorkstrom, A., "A model of CO_2 interaction between atmosphere, oceans, and land biota", in The Global Carbon Cycle (SCOPE 13), edited by B. Bolin, E.T. Degens, S. Kempe and P. Ketner, pp403-457, John Wiley, New York, 1979.

[82] Bacastow, R.B. and Bjorkstrom, A., "Comparison of ocean models for the carbon cycle", in Carbon Cycle Modelling (SCOPE 16), edited by B. Bolin, pp29-79, John Wiley, New York, 1981.

INVENTORY OF ^{137}Cs AND ^{90}Sr IN THE WORLD'S OCEANS

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Abstract

This paper estimates ^{137}Cs inventories in the oceans of the world, at 1972-1974 and attempts in some cases a rough material balance for individual oceans. The balance is found rather poor for the Arctic which contains larger amounts than predicted. The total world inventory is 637±70 PBq and 30 PBq are in the sediments. The total is in statistical accord with the estimated fallout deposition of 637 PBq excluding estimated close-in fallout of 125-195 PBq. The estimated ^{90}Sr inventory for the oceans is 439±48 PBq.

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1. Introduction

^{137}Cs and ^{90}Sr have been added to oceans mostly through fallout from nuclear weapons testing, though an important contribution in parts of the Northern Hemisphere is additions from fuel reprocessing plants. For accurate inventory determination it is necessary to take samples at many vertical depths but coverage of the oceans with adequate profiles is not uniform and there are very few profiles of any type for some oceans. There is sometimes no intercalibration and dubious reliability of results. To minimize these problems and to gain a synoptic view, the results of the GEOSECS expeditions (GEOSECS(16)) have been used extensively.

Later input from the Chernobyl accident is not considered.

Ocean areas are taken from the compilation of Baumgartner and Reichel(3).

An important principle used often in the calculations which follow is the known and constant ratio of ^{137}Cs to ^{90}Sr in fallout which should be taken as 1.45 as shown by Bowen et al.(7). Thus under many oceanographic conditions given the quantity of one radionuclide it is possible to calculate the other.

^{90}Sr analyses are much less common now than the easier ^{137}Cs analyses. Therefore we use the ^{137}Cs results and calculate the ^{90}Sr results from them.

In what follows the input from runoff is assumed to be 3% of fallout input(15, 25, 27) unless specific information is available (e.g. the Baltic(45)).

Information on dumped waste is quite imprecise. An attempt is made in what follows to estimate ^{137}Cs additions, using the known composition of wastes dumped in the N. Atlantic published in NEA/OECD(42) and apply it to wastes dumped in the Pacific also(21). The error associated with that calculation is not known but could be large.

A subsidiary purpose of the exercise was consideration of anomalies in the inventories compared with the inputs and outputs. The exercise is not complete, but given in more detail for interesting anomalies.

2. North Atlantic

Information for this region was surveyed well by Kupferman *et al.*(27) Excluded were the Caribbean, Mediterranean, North Sea, and Norwegian and Greenland seas, which are considered below.

Water column inventory	119 PBq (PBq=10^{15}Bq)
Sediment inventory	2.1 PBq
Total	121(+3.1-12.7) PBq

Fallout (coastal stations)	109±27 PBq
Currents (net addition)	8.9±4.5 PBq
Sellafield addition	5 PBq
Total soluble additions	123±27 PBq

No significant discrepancy is noted. Fallout data here and subsequently are derived from the decay corrected ^{90}Sr results in Volchok and Toonkel(55). An estimated 4 PBq of ^{137}Cs was dumped as waste, but is probably still in insoluble form.

3. S. Atlantic

The GEOSECS(16) data are the only ones used, other data increasing the variance, and not consistent among themselves.

Table 1. S. Atlantic ^{137}Cs inventories by latitude band.

Latitude band	Geosecs inventory (PBq)	Fallout inventory (PBq)
0-10°S	2.2	2.8
10-20°S	3.2	2.5
20-30°S	8.8	5.0
30-40°S	8.0	6.4
40-50°S	4.6	7.4
50-60°S	1.2	3.7
60-70°S	0.31	1.7
70-80°S	0.078	0.27

Total inventory 28.4±5.4 PBq
Sediments: estimated 3.6 PBq

The fallout input is 29.5 PBq and estimated runoff 0.8 PBq. No discrepancy is found. However individual 10° bands to the extreme south show severe deficiencies compared with fallout. The current patterns carry the ^{137}Cs north into temperate zones. This pattern can be expected in other oceans also.

4. Caribbean

No GEOSECS(16) points are in this sea. Data from Vdovenko(53) from 1967 is rather early, and rather variable but gives an inventory of 9.6±0.4 PBq. Using the data for the appropriate GEOSECS(16) latitude bands and areas gives an inventory of 10.5±0.6 PBq which is also consistent with the first estimate, and preferable since more recent. An estimate of sediment inventories is 0.3 PBq. Fallout is 6.4 PBq and runoff 0.2 PBq. According to Kupferman et al.(27) based on recent unpublished data there appears to be a balance between the inventory and the fallout data.

5. Mediterranean

The case of the Mediterranean is summarised in Whitehead et al. (submitted: also available as internal laboratory document). A variety of profiles is available although GEOSECS(16) did not cover this area. Work used was by Thommeret et al.(47), Kautsky(22), Murray and Fukai(37), Livingston et al.(30), Fukai et al.(14) and Ballestra et al.(2). Special allowance was made for the different depths present in the Mediterranean, because the body of water is much shallower than the oceans discussed thus far, and the waters are much better mixed vertically than normal. The calculations yield an inventory of 10.2±1.2 PBq and available sediment inventories (Livingston et al.(30)) give 0.5 PBq.

Input from fallout	10±2 PBq
River input	0.3±0.1 PBq
From Black Sea	0.3±0.1 PBq
From Atlantic	3.5±0.7 PBq

Output to Atlantic	0.7±0.2 PBq
To Black Sea	0.1±0.1 PBq

Net total input and theoretical inventory 13.3±2.3 PBq

The effects of the Straits of Gibraltar are very significant but theoretical and actual inventories balance within error.

6. Black Sea

Vinogradova et al.(54) (1976) quote an inventory of 1.8 PBq. A later estimate of Vakulovskii et al.(51) (1980) is 1.4±0.3 which is the same within statistical error. These should be compared with a calculated fallout input of 1.6 PBq which is not significantly different. Calculated sediment content is 0.5 PBq.

7. N. Pacific

There are extensive other data available than those from GEOSECS(16). Analysis shows that there is a tendency to increase the variance if they are included, however the areas not covered by GEOSECS are so large that other data are necessary and those used are shown in Figure 1. The Bering Sea is considered in the next section.

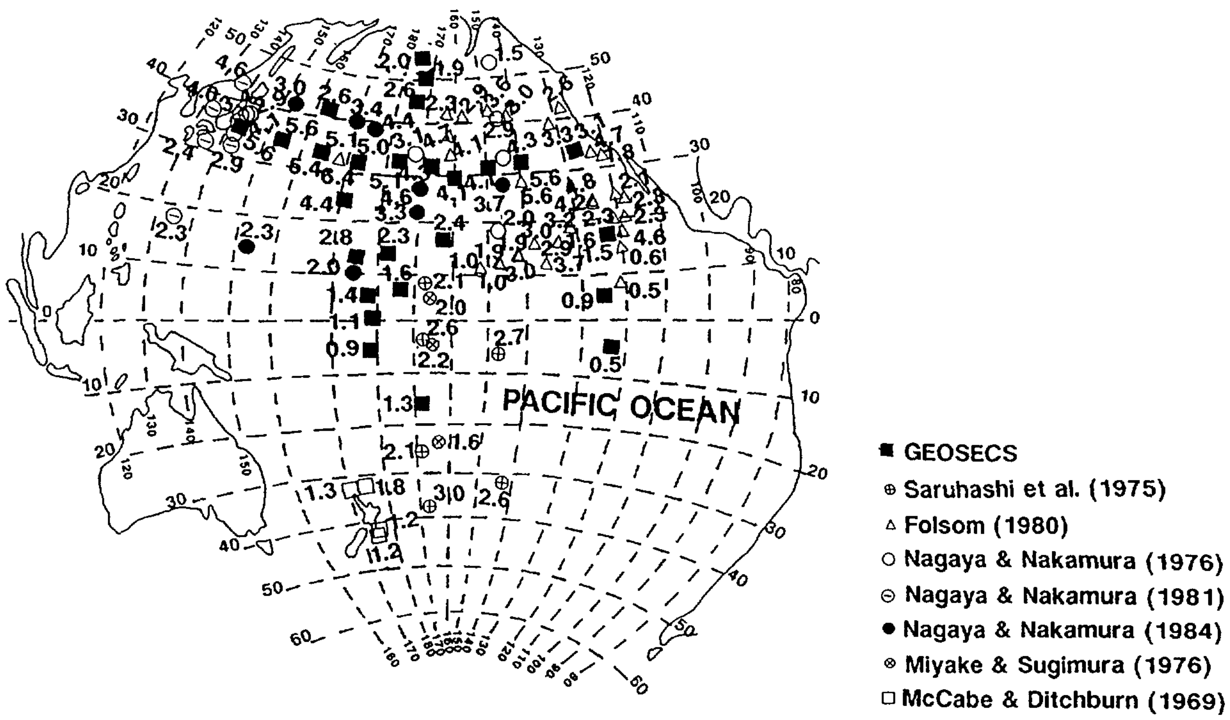

FIG.1. ^{137}Cs inventories (GBq km^{-2}).

Table 2. Inventories of ^{137}Cs in N. Pacific latitude bands.

Latitude band	GEOSECS & other data inventory (PBq)	Fallout inventory (PBq)
0-10°N	29 ±13	17.6
10-20°N	43 ±18	22.2
20-30°N	52 ±20	26.8
30-40°N	49 ±11	28.8
40-50°N	26.1±5.7	28.9
50-60°N	10.3±1.4	18.2
Total	210±32 PBq	144 PBq

Fallout input	144 PBq.
Close-in fallout (decay corrected)	125 PBq
Runoff	4.3 PBq
Loss to sediment (estimation)	4.8 PBq
Loss to Indian Ocean	20 PBq
Loss to Arctic Ocean	4 PBq
Theoretical inventory	240 PBq

Overall the found and theoretical inventories are the same within statistical error.

An estimated 0.06 PBq of ^{137}Cs were also dumped as waste in the N. Pacific.

The excess inventories in the temperate regions are attributed to close-in fallout from weapons testing (Bowen et al.(6)). The deficits in the region 40-60°N are 10.7 PBq and rather large to attribute solely to transfer to the Arctic. Some has probably been transferred by currents to more temperate latitudes.

Sediment inventories or losses to sediment are estimated here and subsequently unless specific data are available, from the relationship between depth and ^{137}Cs inventory for a large collection of data worldwide (Bowen(4), Livingston and Bowen(29), Vakulovskii et al.(52), Noshkin and Bowen(43), Aarkrog(1), Miyamoto et al.(35) , Koivulehto et al.(26), Salo(45), Bowen et al.(5), Nakamura and Nagaya(41), Feldt et al.(12)) which gave a regression relationship

$$\text{Log BM} = -0.5772 \text{ Log D} + 3.7589$$

Where BM is the inventory in Bq Km^{-2} and D is the depth in meters. Loss to the Indian Ocean was estimated using Indonesian archipelago bathymetric data and the maximum literature value of 14E6 m^3 sec^{-1} water transfer.

8. Bering Sea

This sea was not included in the data for the N. Pacific. It contains one GEOSECS(16) station, and the calculated inventory is 4.6 PBq. The sediment inventory is estimated at 0.2 PBq. Fallout for the Bering Sea would be 7.2 PBq and there is a deficit, probably accounted for by the constant flow into the Arctic Ocean. If the 4 PBq calculated earlier has really passed through the area of the sea, then it is clear input and output by currents affect this sea a great deal.

9. S. Pacific

Very few data points are available for this ocean.

Table 3. S. Pacific ^{137}Cs inventories by latitude bands

Latitude band	Minimum Inventory	Data source	Maximum Inventory	Data source	Fallout
0-10°S	32.9	(1)	32.9	(1)	8.2
10-20°S	22.6	(1)	22.6	(1)	6.9
20-30°S	9.4	(2)	28.4	(3)	11.1
30-40°S	8.5	(2)	31.0	(3)	11.6
40-50°S	7.4	(2)	18.3	(3)	12
50-60°S	1.79	(4)	5.8	(5)	5.8
60-70°S	0.52	(4)	2.9	(5)	2.9
70-80°S	0.16	(4)	0.45	(5)	0.45

(1) Measurements in S. Pacific. (2) Calculated from 40-50°S GEOSECS(16) measurements in S. Atlantic. (3) Measurements in S. Pacific (McCabe and Ditchburn(33), Saruhashi et al.(46)). (4) Calculated from corresponding GEOSECS(16) latitude data in S. Atlantic. (5) Estimation from fallout.

Inventory 63-142 PBq: best estimate 102 PBq
Estimate of sediment inventory 4 PBq

The range assumes the possibility of deficits of fallout due to current transfers as seen for the S. Atlantic, but allows for the possibility that the rather high non-GEOSECS measurements reported might be correct. The GEOSECS cruise accumulated samples from this region but analysis awaits funding.

10. Indian Ocean

Data for this ocean are very rare. GEOSECS did not take samples for ^{137}Cs analysis. Russian data(11) for 1975 gave an inventory of 480±300 PBq but a more precise estimate is available from analyses of banded corals etc. as follows.

Inventory estimate assuming S. Atlantic GEOSECS(16) data at 40-50°S apply to 30-50°S in Indian Ocean, and fallout data apply

further north;	44 PBq
Inventory assuming fallout figures only;	56 PBq
Inventory applying world-wide coral data;	155 PBq
Inventory applying local coral data;	86 (+17-9) PBq

Range 44-155 PBq best estimate 86 PBq.

The coral calculations rely on the work of Toggweiler and Trumbore(48) which showed the ^{90}Sr in banded corals corresponded very well with the surface ^{90}Sr in sea-water (and hence indirectly with the ^{137}Cs).

Using their data and GEOSECS(16) data it may be shown that the summed values for ^{90}Sr in their coral bands have a strong relationship to GEOSECS inventories in the water column at nearby points. The regression relationship is

$$\text{Log } C = 0.00855\ I + 0.833$$

where C is the mean concentration of ^{90}Sr in the corals during the period 1959-1979 in dpm/100g and I is the GEOSECS(16) inventory in mCi km^{-2}. In this calculation the Galapagos data point for coral was rejected because there were no adequate GEOSECS stations. Applying this relationship to the mean coral ^{90}Sr figures for the Indian Ocean including one extra coral point from Moore and Krishnaswami(36) gives an oceanic inventory of 155 PBq. However when some local comparisons of Indian Ocean coral data with known historical ^{90}Sr data in surface water of the Indian Ocean are made the relationship is significantly different and the calculated inventory becomes 86(+17-9) PBq. This is taken as the best estimate, though it is not clear why the relationship between ^{90}Sr in water and corals in the Indian Ocean should be different from elsewhere.

Calculated fallout input;	56 PBq
Runoff;	1.7 PBq
Carried from Pacific by currents;	20 PBq
Added as close-in fallout from British tests;	<2 PBq
Estimated lost to sediments;	3.7 PBq
Theoretical inventory;	87 PBq

No clear discrepancy with the best estimate of 86 PBq is demonstrated, but the range is large and agreement is fortuitous.

11. North Sea

This sea was not covered in the previously cited paper on the N. Atlantic, but is particularly important because it is greatly affected by the effluents from the Sellafield and Cap La Hague reprocessing plants and is a contributor to other seas further north. The inventory question is covered in great detail in the accompanying paper in this volume by Kautsky(25). Inventories are given for several years e.g. 0.85 PBq for 1974, but there is constant change. Because of the transit times northwards, effluents from Cap La Hague affect seas to the north very little in 1974.

It is claimed by Jeffries et al.(20) that the Irish and North Seas combined may contain in the sediments as much as 10% of the ^{137}Cs emitted from the Sellafield works. If that is so then the sediment content at the end of 1974 would be 1.0 PBq which is very high, but the result is included until better data become available.

12. Baltic Sea

This very shallow sea is one of those not covered by the previously mentioned paper which surveyed the North Atlantic. It has received a great deal of attention and data are available for inventories in the water column and in the sediment column for the years 1962-1981, as described in Salo(45) et al. (1982).

Table 4. Yearly ^{137}Cs inventories in the Baltic

Year	1960	61	62	63	64	65	66	67	68	69	70	71	72	73	74	1975	76	77	78	79	80	81
TBq	198	302	379	629	747	596	744	757	714	672	645	547	494	492	492	463	384	431	354	362	364	346

The amount in the sediment column at the end of the '70s is given as 49.1 TBQ. Data are also tabulated for ^{90}Sr.

Inflow and outflow are also important for the Baltic. The exact state of balance for any given year in the cited range may be found in the reference.

13. Norwegian Sea

Stations 17, 18 and 19 of GEOSECS(16) were in this region. From these and detailed work of Kautsky(24) giving a total of 31 available profiles the mean and standard deviation is 4.88±0.52 E9

Bq km^{-2}, which with an area of 1.383E6 km^{2} gives an inventory of 6.75±0.72 PBq.

Fallout should have contributed 2.5 PBq. The amount of Sellafield ^{137}Cs reaching the sea can be judged by consideration of the work of Dahlgaard _et al._ (10) and Kautsky(25) who considered ^{134}Cs/^{137}Cs ratios and after relating them to those in the Sellafield effluent, derived transit times, which were about 5 years to the Norwegian sea. The cumulative ^{137}Cs output from Sellafield up to 5 years before the sampling date was 0.54 PBq which is therefore a minor contribution. Estimated sediment content is 0.1 PBq. In principle the transit times as far as the Norwegian Sea should be fairly accurate because it was possible not only to use the ^{134}Cs/^{137}Cs isotope ratio but also follow the isolines of various ^{137}Cs concentrations.

Many more data are available from the 1981 TTO project (Casso and Livingston(9)). Seven profiles yield a total inventory of 9.5±3.2 PBq which is possibly a significant increase compared with 1972-1974. However the calculated Sellafield contribution was larger - 24.5 PBq, and it is obvious much had passed through into other seas.

14. Greenland Sea

Five profiles from Kautsky(23, 24) give a mean and standard deviation of 3.93±0.69 E9 Bq km^{-2}, and since the area of the Greenland sea is 1.205 E6 km^{2} the resultant inventory is 4.74±0.83 PBq. Fallout is 2.2 PBq and with an estimated transit time of 7 years cumulative Sellafield contribution is 0.21 PBq. The sediment column is estimated to contain 0.1 PBq.

For both the Norwegian and Greenland seas, the maximum possible contribution from Sellafield is still small compared with fallout levels in 1972.

1981 data derived from the Transient Tracers in the Ocean Programme (Livingston(28)) are equivalent to an inventory of 8.2 PBq and 10.7 PBq are the calculated Sellafield emissions, some of which probably has passed through to other seas. Later data, from 1984 (Livingston and Casso pers. comm) are equivalent to 8.7±2.0 PBq, which is statistically the same.

15. Arctic Ocean

Up until the GEOSECS expeditions the only data apparently available for the Arctic were ^{90}Sr results for the Chukchi sea by Bowen and Sugimura(8) (unfortunately their result may not be translatable to a ^{137}Cs value) and some results for the Barents Sea by Kautsky[(24)]. It is possible to take the latter and attempt to calculate an inventory for the whole Arctic but we shall show shortly that this is probably quite misleading. The six profiles available yield a mean and standard deviation of 2.03±0.76 E9 Bq km^{-2} and an inventory of 17.3±6.4 PBq. That should be compared with an input from fallout of 5.4 PBq and assuming a transit time of 11 years (Livingston _et al._[(31)]), a Sellafield input of 0.2 PBq. There seems to be a slight excess ^{137}Cs content even as measured around the Barents Sea.

The 1979 LOREX expedition[(31)] obtained two profiles through the ice directly into the Arctic Ocean yielding inventory mean and standard deviation for the Arctic of 75±32 PBq which is considerably higher. (The inventories for Pu isotopes were also much higher than expected). Later data includes a profile in the region just north of Svalbard in 1980 (Holm _et al._)[(19)]. It was not a continuous profile, but combined from various depths and locations. $^{239+240}$Pu/^{137}Cs ratios were consistent with a fallout origin. Incorporation of other unpublished data (Livingson and Casso pers. comm.: Cesar Station, 1983, 86°N, 130°W, Aiwex-3, 1985, 74°N, 144°W) each with ^{137}Cs/^{90}Sr ratios typical of fallout gives a mean and standard deviation of 5.6±4.0 GBq km^{-2} or an inventory of 47±34 PBq. Both inventory and variance are high. Subsequent discussion depends on the assumption the above profiles are moderately representative for the Arctic.

The high LOREX inventories were partially attributed to effluent from Sellafield but analyses failed to show any measurable ^{134}Cs. It may be estimated that a transit time of 10 years or more is needed to produce such an indetectably low ratio. That means cumulative Sellafield output prior to 1969 is responsible which unfortunately is only 1.6 PBq. When we remember that fallout accounts for 5.4 PBq it seems there is a definite difficulty in accounting for the 75 PBq present. To make the situation worse, the ^{90}Sr/^{137}Cs ratios in the LOREX samples are rather higher than expected from fallout. That implies a passage over relatively shallow sediments and adsorption of ^{137}Cs, but an even higher original ^{137}Cs inventory. Such higher ratios are also found at depth in a number of the GEOSECS[(16)] samples in the Greenland sea and Irminger sea GEOSECS samples.

It must be pointed out that there is some uncertainty in the literature about the transit times from Sellafield to the ice and under it in the Arctic. The estimates of Kautsky[(25)] and Dahlgaard et al.(10) do not quite coincide for the vicinity of Svalbard, that of Kautsky being longer, though relying on more data points. However even somewhat shorter transit times than used in this paper would still present a problem due to severe inventory imbalance.

In this context it is worth noting that even in 1972 the inventory calculations suggest excesses present in the Norwegian and Greenland seas.

What could be the origin of such a large amount of ^{137}Cs in the Arctic? Consideration of input from normal fallout in the N. Atlantic carried north by currents gives a contribution of less than 0.05 PBq and therefore cannot be responsible.

One possible origin to be investigated is the alleged 1957 disaster in the Ural mountains in which ^{137}Cs and ^{90}Sr were supposed to be released to the environment in large quantities. Since the effluents would reach the Arctic via the Ob river this source must be considered. The best quantitative estimates, due to Trabalka et al.(49)(1980) give a decay-corrected range of 1.8-18 PBq for ^{90}Sr and about 10% of those figures for ^{137}Cs. However this would create a ratio of the two radionuclides in the Arctic water which is not seen, and also the maximum calculated addition of 1.8 PBq of ^{137}Cs cannot possibly account for the inventory observed. This supposed source probably has a negligible contribution as has other runoff.

According to Bowen and Sugimura(8) the very large weapons tests in Novaya Zemlya were air bursts which do not produce much close-in fallout and should therefore not be contributors. However examination of the original reference (Machta et al.(32)) says only there was no close-in fallout "in so far as can be determined", leaving the question rather open. Close-in fallout would not be detected by the world-wide chain of monitoring stations operated for many years by the EML laboratory in New York to detect ^{90}Sr. The data in Machta et al.(32) also show that almost 50% of the supposed ^{90}Sr produced given the size of the explosions cannot be found in the atmosphere. A significant fraction may have been close-in fallout.

There are a few more small pieces of evidence that there could have been some close-in fallout from the Arctic tests. Cumulative surface deposition on Svalbard of ^{137}Cs (Holm et al.(19)) was found to be 2.2±0.3 E9 Bq km^{-2}, three times the expected value (Volchok and Toonkel(55)) of 0.76 E9 Bq km^{-2}. Also oceanic sediments from that area(19) have ^{137}Cs content 6 times higher than would be expected from the world-wide regression given earlier and translate to a very high sediment inventory for the Arctic of 5.2 PBq. Also persuasive are the Plutonium isotope ratios reported in mosses and lichens from extreme North-east Greenland (Holm *et al.*(18)) which contained amounts of Plutonium isotopes up to 100 times normal, but corresponding to fallout ratios rather than Sellafield addition. In addition, workers from Sweden (Persson *et al.*(44)) reported around 1961-1962 a surprising increase in the quantity of ^{134}Cs in the biosphere which is normally only produced from surface bursts, though stable ^{133}Cs might have been added to a weapon for tracing purposes. Close-in fallout is suggested.

From fission yields of the Novaya Zemlya tests (UNSCEAR(50) 1982) the amounts producing close-in fallout could range between 16 and 100 MT depending on the explosions affected, which following the figure of 28% close-in fallout in Glasstone(17) and known yields of radionuclides could yield between 11 and 70 PBq of ^{137}Cs.

These calculations yield the following:

Inventory found: 47±34 PBq

Losses to sediments:	5.2 PBq
Added from runoff:	0.2 PBq
Added from Sellafield:	1.6 PBq
Added from fallout:	6.9 PBq
Added from N. Pacific:	4 PBq
Added from N. Atlantic:	<0.05 PBq
Added supposed release in Ural Mountains:	0.2-1.8 PBq
Added possible close-in fallout:	11-70 PBq

Of these, only the last seems likely to close the balance, but more work is necessary to make this suggestion less speculative.

16. Summary

The following are the oceans and their estimated inventories, and inventories in the sediment column, as at 1974. Ranges are in brackets, other errors being standard deviations.

Table 5. ^{137}Cs Inventories for all world oceans.

Ocean	Inventory PBq	Sediment inventory PBq	Areal inventory GBq km^{-2}
N. Atlantic	119(+3.1-12.7)	2.1+4 (dumping)	3.43
S. Atlantic	28.2±5.4	3.6	0.43
Caribbean	10.5±0.6	0.6	2.41
Mediterranean	10.2±1.2	0.5±0.2	4.1
Black sea	1.8	0.2	3.91
N. Pacific	210±32	4.8	2.66
Bering sea	4.6	0.2	2.0
S. Pacific	102(63-142)	4	1.07
Indian	86(44-155)	3.7	1.11
North sea	0.85	1.0	1.48
Baltic sea	0.49	0.049	1.28
Norwegian sea	6.75±0.72	0.1	4.88
Greenland sea	4.74±0.83	0.1	3.93
Arctic (1979ff)	47±34	5.2	5.55
Totals	637±70 PBq	30 PBq	

Most of the variance in the result is attributed to the large errors on the estimates for the Indian and S. Pacific Oceans.

We expect a decay-corrected fallout deposition worldwide(55) in the oceans of 637 PBq and an additional 19 PBq from runoff which gives a total the same as the above inventory within errors, even if close-in fallout of 125-195 is added, but does not allow for the possible underestimation of fallout in the sea(27).

Using the known oceanic ratio for ^{90}Sr and ^{137}Cs it may also be calculated that the inventory of ^{90}Sr in the water column is 439±48 PBq.

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REFERENCES

(1) Aarkrog, A., Dahlgaard, H. and Nilsson, K. (1982). Studies on the Distribution of Transuranics in the Baltic Sea, the Danish Belts, the Kattegat and the North Sea. in "Transuranic Cycling Behaviour in the Marine Environment" IAEA-TECDOC-265, I.A.E.A, Vienna, 23-32.

(2) Ballestra, S., Bojanowski, R., Fukai, R. and Vas, D. (1984) Behaviour of selected radionuclides in the Northwestern Mediterranean basin influenced by river discharge. in "International Symposium on the behaviour of long lived radionuclides in the Marine Environment". C.E.C., Luxembourg, 215-232.

(3) Baumgartner, A. and Reichel, E. (1975). "The World Water Balance" R.Oldenburg Verlag, Munich, Vienna, p143.

(4) Bowen, V.T. (1982). Transuranic Behavior in Marine Environment. in "Transuranic Cycling Behaviour in the Marine Environment" IAEA-TECDOC-265, I.A.E.A, Vienna, 129-155.

(5) Bowen, V.T., Livingston, H.D. and Burke, J.C. (1976). Distributions of transuranium Nuclides in Sediment and Biota of the North Atlantic Ocean. in "Transuranium Nuclides in the Environment" I.A.E.A., Vienna, 107-120.

(6) Bowen, V.T., Noshkin, V.E., Livingston, H.D. and Volchok, H.L. (1980). Fallout Radionuclides in the Pacific Ocean: Vertical and Horizontal Distributions largely from GEOSECS Stations. Earth Pl. Sci. Lett. 49, 411-434.

(7) Bowen, V.T., Noshkin, V.E., Volchok, H.L., Livingston, H.D and Wong, K.M. (1974). Cesium 137 to strontium 90 ratios in the Atlantic Ocean 1966 through 1972. Limnol. Oceanol 19, 670-681.

(8) Bowen, V.T. and Sugimura, T.T. (1964). Fission product concentrations in the Chukchi Sea. Arctic 17, 198-203.

(9) Casso, S.A. and Livingston H.D. (1984). Radiocesium and other nuclides in the Norwegian-Greenland Seas 1981-1982. Woods Hole Oceanographic Institute, Woods Hole, Mass. Rep WHOI-84-40. 101pp.

(10) Dahlgaard.H., Aarkrog, A., Hallstadius, L., Holm, E. and Rioseco, J. (1986). Radiocaesium transport from the Irish Sea via the North Sea and the Norwegian Coastal Current to East Greenland. Rapp. P.-v. Reun. Cons. Int. Explor. Mer. 186, 70-79.

(11) Domanov, M.M. and Nelepo, B.A. (1975). Study of the structure of cesium 137 distribution in the equatorial zone of the Indian Ocean. Morsk. Gidrofiz. Issled. 1, 191-197

(12) Feldt, W., Kanisch, G., Kanisch, M. and Vobach, M. (1985). Radioecological Studies of Sites in the Northeast Atlantic used for Dumping of low-level radioactive Wastes - Results of the research Cruises of FRV "Walther Herwig" 1980-1984. Arch. FischWiss. 35, 91-195.

(13) Folsom, T.R. (1980). Some measurements of Global Fallout suggesting Characteristics of the N.Pacific controlling dispersion rates of certain surface pollutants. in "Isotope Marine Chemistry". Geochem. Res. Ass. Tokyo, 51-117.

(14) Fukai, R., Ballestra, S. and Thein, M. (1981). Vertical distribution of transuranic nuclides in the Eastern Mediterranean Sea. in "Techniques for Identifying Transuranic Speciation in Aquatic Environments", I.A.E.A., Vienna, 79-87.

(15) Fukai, R., Ballestra, S., Thein, M. and Guion, J. (1981). Input of transuranic elements through rivers into the Mediterranean Sea, in "Impacts of Radionuclide Releases into the Marine Environment" I.A.E.A., Vienna, 3-14.

(16) GEOSECS Executive Committee (1987). "Atlantic, Pacific and Indian Ocean Expeditions, Volume 7, Shorebased data and graphics" National Science Foundation, Washington, D.C. 200pp.

(17) Glasstone, S. (1962). The effects of Nuclear testing. U.S.A.E.C.

(18) Holm, E., Aarkrog, A., Ballestra, S. and Dahlgaard, H. (1986). Origin and isotopic ratios of plutonium in the Barents and Greenland Seas. Earth. Pl. Sci. Lett. 79, 27-32.

(19) Holm, E., Persson, B.R.R., Hallstadius, L., Aarkrog, A. Dahlgaard, H. (1983). Radio-cesium and transuranium elements in the Greenland and Barents Seas. Oceanol. Acta 6, 457-462.

(20) Jeffries, D.F., Steele, A.K. and Preston, A. (1982). Further studies on the distribution of ^{137}Cs in British coastal waters. I. Irish Sea. Deep-sea Res. 29, 713-738.

(21) Joseph, A.B., Gustafson, P.F., Russell, I.R., Schuert, E.A., Volchok, H.L. and Tamplin, A. (1971). Sources of Radioactivity and their characteristics. in "Radioactivity in the Marine Environment" National Academy of Sciences, Washington, D.C. 6-41.

(22) Kautsky, H. (1977). The vertical distribution of radioactive fallout products in the Western Mediterranean in 1970 and 1974. Deut. Hyd. Zeit. 30, 175-184.

(23) Kautsky, H. (1977). Distribution of radioactive fallout produts in Atlantic water between 10°S and 81°N during the years 1969 and 1972. Deut. Hyd. Zeit. 30, 216-227.

(24) Kautsky, H. (1980). Distribution of radioactive fallout products in the water of the North Atlantic and the Barents Sea during the year 1972. in "Isotope Marine Chemistry" Geochem. Res. Ass. Tokyo. 9-23.

(25) Kautsky, H. (1988). Artificial radioactivity in the North Sea and the Northern North Atlantic during the years 1977 to 1986. This volume, 31pp.

(26) Koivulehto, M. Saxen, R., and Kristiina, S. (1979). Radionuclides in aquatic Environments. in "Stud. Environ. Radioact. Finl." Sateilyturvallisuuslaitos STL-A26, 65-83.

(27) Kupferman, S.L., Livingston, H.D. and Bowen, V.T. (1979). A mass balance for ^{137}Cs and ^{90}Sr in the North Atlantic Ocean. J. Mar. Res. 37, 157-199.

(28) Livingston, H.D. (1985). Anthropogenic radiotracer evolution in the Central Greenland Sea. Rit Fiskideildar 9, 43-54.

(29) Livingston, H.D. and Bowen, V.T. (1979). Pu and ^{137}Cs in coastal sediments. Earth. Pl. Sci. Lett. 49, 29-45.

(30) Livingston, H.D., Casso, S.A., Bowen, V.T. and Burke, J.C. (1979). Soluble and particle-associated fallout radionuclides in Mediterranean water and sediments. Rapp. Comm. Int. Mer. Medit. 25/26, 71-74.

(31) Livingston, H.D., Kupferman. S.L., Bowen, V.T. and Moore, R.M. (1984). Vertical profile of artificial radionuclide concentrations in the Central Arctic Ocean. Geochim. Cosmochim. Acta. 48, 2195-2203.

(32) Machta, L., List, R.J. and Telegadas, K. (1964). Inventories of selected long-lived radioisotopes produced during nuclear testing. U.S.A.E.C. Health and Safety Laboratory, New York, Rep. HASL-142, 244-271.

(33) McCabe, W.J. and Ditchburn, R.G. (1969). The investigations of the distribution of radionuclides in a series of ocean water profiles and surface water stations in the South Pacific Ocean. Institute of Nuclear Sciences, Lower Hutt, New Zealand Rep. INS-R-61. 14pp.

(34) Miyake, Y. and Sugimura, Y. (1976). The Plutonium content of Pacific Ocean waters. in "Transuranium nuclides in the environment" I. A. E. A., Vienna, 91-101.

(35) Miyamoto, T., Hishida, M., Shibayama, N., and Shiozaki, M. (1986). Marine Behaviour of Long-lived Radionuclides (Fall-out) at the proposed Disposal Site of Radioactive Wastes in Western North Pacific. in "Behaviour of Long-lived Radionuclides associated with deep-sea Disposal of Radioactive Wastes." IAEA-TECDOC-368, I. A. E. A., Vienna, 73-80.

(36) Moore, W. S. and Krishnaswami, S. (1974). Correlation of X-radiography revealed banding in corals with radiometric growth rates. Proc. 2nd Int. Coral reef Symp. 2, 269-276.

(37) Murray, C. N. and Fukai, R. (1978). Measurement of $^{239+240}$Pu in the North-western Mediterranean. Est. Coast. Mar. Sci. 6, 145-151.

(38) Nagaya, Y. and Nakamura, K. (1976). ^{90}Sr and ^{137}Cs contents in the surface waters of the adjacent seas of Japan and the North Pacific during 1969 to 1973. J. Oceanogr. Soc. Jap. 32, 228-234.

(39) Nagaya, Y. and Nakamura, K. (1981). Artificial radionuclides in the western Northwest Pacific. (I) ^{90}Sr and ^{137}Cs in the deep waters. J. Oceanogr. Soc. Jap. 37, 135-144.

(40) Nagaya, Y. and Nakamura, K. (1984). ^{239,240}Pu, ^{137}Cs and ^{90}Sr in the Central North Pacific, J. Oceanogr. Soc. Jap. 40, 416-424.

(41) Nakamura, K. and Nagaya, Y. (1985). Accumulation of Cs-137 and Pu-239, 240 in sediments of the Coastal Sea and the North Pacific. in "Marine and Estuarine Geochemistry", Lewis Publishers, Chelsea, Mich. 171-180.

(42) NEA/OECD (1980). Review of the continued suitability of the dumping site for radioactive waste in the north-east Atlantic. NEA/OECD, Paris, 100 pp.

(43) Noshkin, V. E. and Bowen, V. T. (1973). Concentrations and Distributions of Long-lived fallout radionuclides in open Ocean Sediments. in "Radioactive Contamination of the Marine Environment" IAEA-SM-158/45, I. A. E. A., Vienna, 671-86.

(44) Persson, R. B. R. (1968). ^{134}Cs/^{137}Cs activity ratio in the biosphere from 1956 until 1966. Health Phys. 14, 241-250.

(45) Salo, A., Tuomainen, K. and Voipio, A. (1982). The inventories of certain long-lived radionuclides in the Baltic Sea. in "Transuranic cycling behaviour in the marine environment" IAEA-TECDOC-265, I.A.E.A., Vienna, 53-62.

(46) Saruhashi, K., Katsuragi, Y., Kanazawa. T., Sugimura, Y. and Miyake, Y. (1975). ^{90}Sr and ^{137}Cs in the Pacific waters. Rec. Oceanogr. Works Jap. 13, 1-15.

(47) Thommeret, J., Thommeret, Y. and Hugues, G. (1976). Several measurements of Carbon-14 and Caesium-137 in the Ligurian Sea. Rapp. Comm. Int. Mer Medit. 23, 173-176.

(48) Toggweiler, J.R. and Trumbore, S. (1985). Bomb-test ^{90}Sr in Pacific and Indian Ocean surface water as recorded by banded corals. Earth. Pl. Sci. Lett. 74, 306-314.

(49) Trabalka, J.R., Eyman, L.D. and Auerbach, S.I. (1980). Analysis of the 1957-1958 Soviet Nuclear Accident. Sci. 209, 345-357.

(50) UNSCEAR (1982). Ionizing radiation: Sources and biological effects. United Nations, New York, p 227.

(51) Vakulovskii, S.M., Katrich, I.Yu., Krasnopevtsev, Yu. V., Nikitin, A.I., Chumichev, V.B. and Shkuro, V.N. (1980). Spatial distribution and balance of ^{3}H and ^{137}Cs in the Black Sea during 1977. Atomn. Energ. 49, 105-108.

(52) Vakulovskii, S.M., Krasnopevtsev, Yu.V., Nikitin, A.I. and Chumichev, V.B. (1982). Distribution of ^{137}Cs and ^{90}Sr between water and bottom sediments in the Black Sea, 1977. Oceanology 22, 712-715.

(53) Vdovenko, V.M., Kolesnikov, A.G., Spitsyn, V.I., Bernovskaya, R.N., Gedeonov, L.I., Gromov, V.V., Ivanova, L.M., Nelepo, B.A. Tikhomirrov, V.N. and Trusov, A.G. (1971). Radioactivity of Ocean Waters and Behaviour of some Fission Products. At. Energ. 31, 409-422.

(54) Vinogradova V.K., Rosyanov, S.P. and Gedeonov, L.I. (1976). Deposition of Global ^{137}Cs fallout in the Black Sea. Oceanology 15, 43-5.

(55) Volchok, H.L. and Toonkel (1974). Worldwide deposition of ^{90}Sr through 1973. U.S.A.E.C. Health Safety Lab. Fallout Program Quart. Summ. Rep. HASL-286, pp I-17-I-35.

ARTIFICIAL RADIOACTIVITY IN THE NORTH SEA AND THE NORTHERN NORTH ATLANTIC DURING THE YEARS 1977 TO 1986

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Abstract

Data are presented for ^{134,137}Cs, ^{90}Sr, ^{239,240}Pu, ^{238}Pu, ^{241}Am and ^{3}H in the North Sea during 1977-1985. ^{3}H distribution tends to differ from that of the other radionuclides. Seasonal effects are noted. The $^{137+134}$Cs inventory in 1984 was 2400 TBq. Transport times calculated are; 3 years from Sellafield to the Pentland Firth, 6-7 to Spitsbergen and 7-9 to East Greenland. Some data from analyses following the Chernobyl accident are given.

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1. INTRODUCTION

In this report only the purely radiological questions of the content and distribution of artificial radio nuclides in the North Sea and in the northern North Atlantic are dealt with. The oceanographical, biological, and sedimentological questions have already been dealt with - as far as possible - in the Report of 1981 (1).

The distribution of the radio nuclides in water, offers a basis for the recognition of the transport routes. The existing isotope ratios $^{137+134}$Cs:^{90}Sr resp. ^{134}Cs:^{137}Cs, and their temporal changes are utilized for estimation of transport times. The basis for the studies presented are essentially the radiological measurement results gained in the years 1977 to 1986 in the Deutsches Hydrographisches Institut in Hamburg (data available until November 1986). In several points, they are supplemented by corresponding results which originate from Aarkrog and his collaborators (2),(3).

2. GENERAL

One can look upon the nuclear fuel reprocessing plants Sellafield Works on the Irish Sea and La Hague on the English Channel as being the principal sources of the artificial radio nuclides present today in the North Sea and in the adjoining regions of the Atlantic towards the North. Thereby, the discharge from La Hague is slight compared with that of Sellafield Works. The actual quantities discharged by both plants are always quite clearly below the quantities they are permitted.

Only in a closely limited sea area in the immediate vicinity of the discharge from Sellafield Works can clearly higher activity concentrations of ^{137}Cs be measured in the sea-water. According to the last English measurements published (4), these values lay at about 5 kBq$\cdot$m^{-3} in 1984. That indicated a further constant decrease of the ^{137}Cs activity concentrations in this region compared with previous years, with around 100 kBq$\cdot$m^{-3} in 1977, around 50 kBq$\cdot$m^{-3} in 1978, around 25 kBq.m^{-3} in 1979, and around 10 kBq$\cdot$m^{-3} in 1982.

In the interest of safety, a decrease of this discharge from Sellafield Works - as has constantly taken place for several years - is to be welcomed. The effect of the long time influence of low supplementary radiation doses upon the biosphere, however, is not exactly known to date. However, one should be able to assume that, so long as this additional radiation remains far below the radiation dose range present in nature, then no measurable influence upon the biosphere is to be expected. The activity concentrations of the artificial radio nuclides present in the water of the North Sea - especially those of ^{137}Cs and ^{90}Sr, as well as those of ^{106}Ru and ^{125}Sb insofar as they are capable of being measured - lie at not more than 1 % of the natural radio nuclides present in seawater. The activity concentrations of other artificial nuclides, such as for example the Transuranics, lie even further below (at about the factor 1000) and lie, in the majority of cases, near the limits of detection of our working methods.

The Chernobyl accident had only a temporally limited effect upon the North Sea because, owing to the constant water exchange with the Atlantic, the radio nuclides brought in, during a short period of time, via the atmospheric fallout were relatively quickly distributed and transported away.

3. DISTRIBUTION AND CONTENT OF $^{137+134}Cs$ IN THE WATER OF THE NORTH SEA

At all points of the station grid in different years, surface water samples were taken. In addition, 1982, at all points, water samples were taken at depth North of 53°30' N and North of 53° N in 1984 (11).

Deep water samples were also taken in the years 1978 to 1981, only in the northern regions of the North Sea, from about 80m depth between 57° N as far as - in part - 62° N, as well as in the Norwegian Deep and in the Skagerrak. In the two latter regions, there are always layers recognizable which are to be traced back to the different water bodies lying over one another (Baltic Sea, North Sea, and Atlantic waters).

In each case, from 50 l of water the Caesium was precitated as Hexanitrocobaltate. After chemical conversion of the precipitate over several stages into Iodobismuthate the activity of the sample was determined in an anti-coincidence β -counter.

A few γ-spectroscopical measurements, carried out during the years 1983 to 1984 using water samples of about 2000 l, proved that about 2 to 3 % of the total amount of the radio caesium present in the water of the North Sea are ^{134}Cs (influence Sellafield Works). Aarkrog (personnel communication) found in 1983, at 5 stations in the region immediately West of Jutland, a mean value of about 10.2 % ^{134}Cs (between 8 to 14 %) (influence La Hague).

The general picture of the distribution of the radio caesium in the North Sea, found in the previous years, is also to be met with in years 1982 to 1985. As the result of the much smaller discharges of radio caesium from La Hague into the English Channel, compared with that of the Sellafield Works into the Irish Sea, in the southeastern North Sea along the mainland coast from Dover as far as in the region of northernmost Jutland (circa 56° N) clearly lower concentrations of $^{137+134}Cs$ are to be found than in the western and central North Sea (Fig.4 - 6).

In April 1977, the beginning of an influx appeared of larger quantities of ^{137}Cs into the North Sea in the region of the Pentland Firth in a clearly recognizable way (5). The activity concentration values rose to over 400 $Bq \cdot m^{-3}$ in the surface water compared to 170 to 190 $Bq \cdot m^{-3}$ in September 1976 (6).

In contrast, the rest of the North Sea was still marked by comparatively low activity concentration values of about 50 to 100 $Bq \cdot m^{-3}$.

The ^{137}Cs found in the northwestern North Sea and also westwards of the Orkney and Shetland Isles - apart from slight fallout quantities still present - originated with certainty from the Sellafield Works (6),(7),(8),(9).(10).

Nearly all of the Caesium flowing through the Minch to the North enters the North Sea in the region of the Orkney and Shetland Isles. A direct transport into the Norwegian Sea in the region westward of the Shetland Islands takes place - if at all - to only a small extent.

The large-scale investigations of surface and deep water in the years 1982 and 1984 offer, for the first time, the possibility of undertaking a comparison of the horizontal distribution of the Cs in different water layers of the North Sea for the years 1982 and 1984 (11).

The large-scale distributed measurements permit the calculation of balances of the total content of different radio nuclides in the water of the North Sea for different years. For $^{137+134}$Cs this is possible from 1978 onwards (in partial regions, from 1975 already) (5).

With the comparison of the content of $^{137+134}$Cs (Table 5a) in the different years, a distinct maximum is indicated in 1978. This is to be traced back to the particularly strong input of the year 1977 in the region of the Orkney Islands. By a comparison of different regions of the North Sea one can recognize that the maximum content occurred in its western parts in 1978; in the eastern parts as far as 58°30' N as well as in the Central North Sea, in 1979; and in the northeastern part as far as 61° N, in 1980. That reflects a particular, although not very exact, picture concerning the transport routes and transport times. The flushing time of the North Sea between the Pentland Firth through the Central and Southern North Sea as far as into the Norwegian Deep, according to these values, could lie within the range of about 2 to 3 years.

In total, the content of $^{137+134}$Cs in the North Sea - after the unprecedented particular strong input in 1977 - has again reduced to around half of that between the years 1979 and 1981.

4. TRANSPORT PATTERN

4.1 North Sea

During the years 1977 to 1985 an annual cruise was undertaken for the investigation of the distribution of artificial radio-nuclides in the water of the North Sea. The general distribaution picture of $^{137+134}$Cs (Fig.1) is largely consistent with the results of earlier investigations (6).

The water flowing into the North Sea in the region of the Pentland Firth and the Orkney Islands moves along the British East coast towards the South. In the course of this route, one can recognize in three regions - at circa 57° N to 58° N, 55° N to 56° N and 53° N to 54° N - transport currents directed towards the East resp. Northeast.

The water near Dover, flowing into the North Sea from the South, moves along the mainland coast, in a relatively narrow strip, towards the North. The activity concentrations of $^{137+134}$Cs contained therein, and originating from La Hague, are essentially lower than in the water coming from the North. Thereby, a steep concentration gradient of the Cs exists between this two water bodies.

The two water masses, that coming from the North and that coming from the South, flow between about 52°20' N 2° E and 56° N 6°30' E, alongside but separate to one another.

One can also well distinguish the two water bodies from their ratio of ^{137}Cs:^{90}Sr. Westwards of Jutland - between 54°30' N and 56° N - where the boundary lies mostly at 6°30' E, one can recognize the strong concentration drop from West to East. The ratio figures Cs:Sr are also very different between 55°30' N 6° E and 55°30' N 7° E (Fig.3).

There is much more Cs than Sr in the discharge from the Sellafield Works (about nine to thirteen times more) whereas, in the discharge from La Hague normally the Sr clearly predominates over the Cs (up to 5 times more) (Table 4) (12).

A mixing of these two water bodies, and that of the water masses on the northernmost transport route towards the East, first takes place at about 57° N and particularly in the region of the Skagerrak. The further transport then takes place mainly along the Norwegian coast towards the North; small quantities reach the Baltic Sea (1)(2)(7)(13).

The degree of variation with time of the large scale transport routes is relatively slight in the Central and Southern North Sea. Conversely, in the North rather more complex ratios are encountered.

4.2 Northern North Atlantic

Almost the whole of the Caesium introduced into the sea with the waste waters of the Sellafield Works is transported into the northern North Atlantic over the North Sea alongside the Norwegian coast (1)(5)(6)(11).

The vertical distribution of the radio caesium, originating from the Sellafield Works, in the northern North Atlantic speaks for the fact that the water transport also over these wide stretches predominantly takes place in the surface layer (about down to 200m depth). The radio caesium found in the greater sea depths (from about 500m) should essentially originate from the earlier atmospheric atomic bomb tests - which were practically discontinued in 1962. Thereby, activity concentration values of in part below 1 $Bq{\cdot}m^{-3}$ lie even more clearly below the values to be found today in the surface water of around 3 $Bq{\cdot}m^{-3}$ for radio caesium from that source.

In the horizontal distribution pattern of the Cs, there appears very clearly in outline the different current routes in the northern North Atlantic given by Trangeled.

a) Along the Norwegian coast, the Norwegian Atlantic current
b) The branch of that current branching-off at about 64°N towards the Northwest
c) The North Cape Current
d) The branching-off West Spitsbergen Current at about 70°N towards the North
e) The Bear Island Current coming from the Northeast (18)

Also in the data of 1982 (Fig.13) these transport routes appear in outlines.

In addition to that, in 1982 - especially in connection with the data of Aarkrog (19) - one can recognize very well the water transport running from NNE towards SSW between about 78° N 3° W and 72° N 18° W eastwards of Greenland (East Greenland Current).

The transport route branching off at 64° N towards the Northwest runs as far as the 0° meridian, which it reaches at about between 69° N and 72° N. With regard to its further course, we are unable to make any statements from the data available to date.

5. TRANSPORT TIMES

5.1 North Sea

The content of $^{137+134}Cs$ in the water of the North Sea can be correlated in the years observed, with the discharge from the Sellafield Works, which took place around 2 years previously (Table 1a).

In comparison, the content of ^{90}Sr in 1979 - the only year from which sufficient ^{90}Sr data were available for the compilation of a total assessment of the ^{90}Sr content present in the North Sea - lay at around double that of the quantity discharged from the Sellafield Works two years previously (Table 1b).

The comparison of the ratio figures of Cs:Sr in the discharge of the Sellafield Works and in the region of the Pentland Firth and the Central North Sea (Fig.2) clearly indicates an unexpectedly strong shift in favour of ^{90}Sr. The shorter half-life of the ^{134}Cs - of around 2 years - plays no rôle worth mentioning thereby (Fig.2).

Our own estimation for the region between La Hague and the German Bight as well as calculations carried out by other scientists for the Irish Sea (7)(14)(15) indicate a loss of ^{137}Cs from the seawater by absorption onto particulate matter or bottom sediments to be in the range of no more than about 5 to 10 %. Therefore, the over-proportional great change of the Cs:Sr ratio observed should be attributed to the influence of the worldwide fallout, the runnoff from land, and the water exchange with the Atlantic.

In Table 2 rough estimate of the yearly input of ^{137}Cs and ^{90}Sr into the North Sea from sources other than the nuclear facilities is presented. This estimate clearly shows that - in relation to the quantities of ^{137}Cs and ^{90}Sr already present in the North Sea and in the main originating from the outlet of the Sellafield Works - substantially more ^{90}Sr (about 11 %) compared with ^{137}Cs (about 2 to 2.5 %) is introduced yearly into the North Sea from sources other than nuclear facilities. One can assume that the corresponding conditions in the Irish Sea, in principle, are largely commensurable with those in the North Sea. The outlet of La Hague has no influence upon the radiological conditions of the Irish Sea or the western North Sea.

The course of the two curves of the Cs:Sr ratio in the discharge of the Sellafield Works and in the region of the Pentland Firth indicates a transport time of about three years from the Sellafield Works as far as the Pentland Firth (Fig.2). If one compares further the corresponding ratio figures westwards of Jutland at 55°30' N and 6° E, this then results in a practically parallel curve, but offset by about a further year.

For the estimation of the transport times one can also use the temporal change of the ^{134}Cs:^{137}Cs isotope ratio - dependent upon the different half-life times - referred to the isotopes mixture discharged from the Sellafield Works.

In Table 6, the discharge of the caesium isotopes and their ratio in the waste waters for the Sellafield Works (17) and for La Hague (12) are given. In Table 7, the decay corrected ^{134}Cs:^{137}Cs ratio figures - referred to the different discharge years - are summarized for the years 1982 to 1985.

If one considers the temporal changes of the ratios of the ^{134}Cs:^{137}Cs present in the water on its way from the Sellafield Works through the North Sea, then one attains transport times which are comparable with those which have been calculated before in another manner (Table 8).

5.2 Northern North Atlantic

For the investigation of the water transport times over greater distances, the temporal change of the isotope ratios of the isotopes ^{134}Cs and ^{137}Cs, which enter the sea with the waste waters, is suitable. The exactitude of these methods depends naturally upon the relevance of the measurement data. In practice, only 2 factors should be viewed as being actually essential: the statistical counting error by the determination of the activity concentrations and the input of Cs isotopes from other sources. Thereby, practically only two sources come into consideration:

a) The waste water from La Hague
b) The worldwide fallout.

An exact calculation of the influence of La Hague upon the ^{134}Cs:^{137}Cs ratio is not possible. The diecharge from Sellafield Works (17) is many times that of La Hague (Table 6).

The water coming from the West which contains the caesium from the Sellafield Works, the water streaming along the coast of Jutland towards the North with the caesium from La Hague, and the water flowing out of the Baltic Sea in which practically only ^{137}Cs is contained, mix in the Skagerrak. An exact mixing ratio of these 3 water bodies could only be calculated if the water quantities of the respective inflow would be known exactly. That is not the case.

During the years 1982 to 1985 annually at certain positions in the northern North Atlantic, water samples of 500 l to 2000 l were taken. The separation of the Cs took place over ion exchangers (21)(22). The exchanger was measured gamma spectroscopically in a Marinelli beaker with a semi-conductor detector (Ge(Li)) for 250 hours.

The transport time calculation of the water from the ratio of the activity concentrations of the ^{134}Cs:^{137}Cs, bases upon the following suppositions:

a) The only source for radio caesium worthy of mention are the Sellafield Works in the Irish Sea.
b) In the surface water of the North Atlantic, there are still around 3 Bq.m^{-3} of ^{137}Cs present from the fallout of the atmospheric atomic bomb experiments (19)(20)(26). The original ^{134}Cs present from that source has decayed in the meantime.

The results for the individual years are summarized in Tables 9 to 11. Table 12 gives a comparative summary of all the data, and Fig.14 their spatial distribution. If one considers the data from the different regions then one can see the following mean transport times from the Sellafield Works to the respective regions:

Southern Norway	4 to 5 years
Middle Norway (Lofoten)	5 years
Northern Norway (North Cape)	5 to 6 years
Bear Island until the middle of Spitsbergen	6 to 7 years
North of Spitsbergen	7 to 8 years
Eastwards of Greenland	7 to 9 years
SSE of the southernmost point of Spitsbergen	8 to 9 years
70° N 0°	4 years

The transport time of 4 years calculated in the central Norwegian Sea in 1985 at 70° N 0° is certainly somewhat too short. In general, the transport time values found in 1985 - when compared with the values of the previous years - appear to be somewhat shorter. This could be due to the fact that the background for fallout ^{137}Cs of 3 $Bq \cdot m^{-3}$ determined in 1982, in the meantime has receded further and is only now conditionally applicable as correction factor.

6. DISTRIBUTION OF ^{90}Sr

6.1 North Sea

In comparison to ^{137}Cs the activity concentrations of ^{90}Sr observable in the North Sea are distinctly lower (Figs.7, 8). The main source is, as for Cs^{137}, the Sellafield Works near the Irish Sea. In addition, since 1983, the region of the southeastern North Sea along the continental coast is clearly influenced by the ^{90}Sr released into the English Channel with the waste waters of La Hague.

Until 1982, the distribution of the ^{90}Sr in the North Sea widely corresponds to that of the ^{137}Cs. Here also, the highest activity is observed mostly off the English Coast in the region between 55° N and 58° N. Since 1983, the highest values can be found in the Channel and the southeastern North Sea between La Hague and about 56°30' N.

In parallel to ^{137}Cs, the highest ^{90}Sr values occurred in the North Sea during 1978. Between 1978 and 1981, in the western North Sea, they lay at about 15% of the ^{137}Cs values.

1979 was the only year for which sufficient measurement data of ^{90}Sr was available, in order to be able to calculate the total content of the ^{90}Sr in the water of the North Sea (Table 5b). It amounted, with 913 TBq (24 688 Ci), to around 17.5 % of the ^{137}Cs present in the North Sea.

6.2 Northern North Atlantic

Following the water which leaves the North Sea at 61° N one can clearly observe in 1979 (Fig.15) the transport of ^{90}Sr to the North within a narrow strip alongside the Norwegian coast. Along this path the activity concentrations of ^{90}Sr diminish from 26 $Bq \cdot m^{-3}$ at 61° N to about 10 $Bq \cdot m^{-3}$ at 71°N. Westwards of this strip, towards the open Atlantic, as well as northwards to the Polar Sea, much lower activity concentrations of ^{90}Sr are to be found.

The ratio of $^{137}Cs:^{90}Sr$ originating from the atomic bomb tests lies at about 1.4:1. On the other hand, this ratio amounts to 4.6 in the mean (4.2 to 5.1) in the surface water of the region between 65° N and 72° N. This clearly indicates an additional content of ^{137}Cs in the water of this region originating mainly from Sellafield Works. A ratio of 2.4 can still be observed at 80°49' N 9°E.

At the few stations in the Barents Sea where water samples have been taken from the surface and from 100m depth, the ratio values of $^{137}Cs:^{90}Sr$ of about 3.6 (3.3 to 3.9) found in the deep water lie distinctly below those observed in the surface water of about 4.5 (4.3 to 4.6).

In 1982, at 73°50' N 5°20' E, water samples were taken at different depths down to near bottom. The activity concentrations of the ^{90}Sr contained therein decrease between the surface and the near bottom region from 5.7 $Bq \cdot m^{-3}$ to 0.5 $Bq \cdot m^{-3}$. Only at 50m depth an anomalous low value can be observed (in parallel to ^{137}Cs). Having 2.6 in the mean (2.2 to 3.1) the ratio of $^{137}Cs:^{90}Sr$ between the surface (2.3) and 750m depth (2.2) lies clearly above the ratio of about 1.4 in fallout. Further down, at 1000m depth, a ratio of 1.5 can be observed, This might indicate that part of the ^{137}Cs originating from nuclear facilities has spread over the water column down to 750m. The number of ^{90}Sr data available is not sufficient to carry out transport time clculations using the ratio $^{137}Cs:^{90}Sr$.

7. DISTRIBUTION OF TRANSURANIC NUCLIDES

The activity concentrations of the transuranic nuclides lie in the region of $mBq \cdot m^{-3}$ and thereby fundamentally 3 orders of magnitude below those of the ^{137}Cs ($Bq \cdot m^{-3}$). For that reason, their detection requires very large-scale and time-consuming analysis methods, so that only relatively few measurement data of these nuclides are available.

Their distribution pattern in the North Sea (Fig.9) largely corresponds to that of the ^{137}Cs. The highest measurement values were normally found in the region of the Pentland Firth and alongside the British East coast.

Data of the activity concentrations of $^{239+240}Pu$ covering the North sea between 51° N and 60° N only exist from the year 1980 (23) from a coarse measurement network. This data material was utilized in order to undertake a rough estimation of the total content of $^{239+240}Pu$ in the North Sea (Table 5c). The total quantity of $^{239+240}Pu$, with 1.5 TBq (40.6 Ci) in that year was only 0.03 % of the $^{137+134}Cs$ present in the North Sea.

In 1982 (Fig.9) the distribution picture of the $^{239+240}Pu$ is similar to that of 1980, with the highest measurement values in the region of the Pentland Firth and along the British East coast. In all cases, the activity concentrations of the ^{238}Pu lie clearly below those of the $^{239+240}Pu$.

The highest activity concentration values of the ^{241}Am (Fig.10), in general , lie 1982 in total somewhat lower than in 1981. The highest values are also here to be found in the region of the Pentland Firth.

The foregoing reproduced results indicate a widely unchanged extremely slight burdening of the North Sea by transuranic nuclides.

8. DISTRIBUTION OF TRITIUM

A sufficient quantity of data for the calculation of the whole of the Tritium content in the North Sea (Fig.11) is only available for the year 1981. Compared with other nuclides , such as ^{137}Cs or ^{90}Sr, the distribution of Tritium (Table 5d) is quite different. In 1981, its content lay - with 25766 TBq (696 397 Ci) - at around 9 times that of the ^{137}Cs. The highest concentrations are to be met with in the southeastern North Sea and in the Norwegian Deep; that is, in the region with the highest fresh water input (24). Its highest concentrations occur in the North Sea in those regions with lower salinity. The measurement of Tritium carried out in the years 1980 to 1985 show, especially in the region of the German Bight, a constant decrease of the Tritium activity concentration in surface water.

The few data of Tritium gained during 1979 in the northern North Atlantic are presented in Figure 15.

9. FIRST MEASUREMENTS AFTER THE ACCIDENT AT CHERNOBYL (25)

Via the fallout and the precipitation, artificial radionuclides - which were emitted into the armosphere from the accident at Chernobyl - entered the North Sea surface water and especially the Baltic Sea surface water.

In the North Sea, circa 1 month after the main input, especially in the region of the German Bight and along the coast of Jutland, clearly increased activity concentrations of ^{137}Cs (Fig.16) are indicated - compared with the situation before Chernobyl. Nevertheless, the values occurring still lie below the values which could be measured in the previous years in the northwestern and central North Sea as the result of the discharges from the Sellafield Works (Fig.4).

Besides the Cs-isotopes in the fallout there were also a whole series of other radionuclides such as ^{131}I (half life 8.02 d), ^{103}Ru (39.27 d), ^{106}Ru (372 d), ^{141}Ce (32.5 d), ^{140}Ba (12.75 d), ^{132}Te (3.2 d), and of the transuranic nuclides besides $^{239+240}$Pu (24 400 a); ^{238}Pu (86.4 a), ^{241}Am (458 a), and particularly the ^{242}Cm (163 d) were to be met with in amounts worth mentioning. The introduction of ^{90}Sr and Tritium in this event was low.

10. SUMMARY

The measured concentrations of ^{137}Cs in the water of the North Sea gained during the years 1977 to 1985 largely confirm the results found in the preceeding years (6)(16).

During ths years 1982 and 1984, besides surface water samples, water samples were also taken at depth for the first time along the entire station grid northward of 53°30' N (1982) and 53° N (1984).

The fundamental distribution pattern of the $^{137+134}$Cs in the North Sea as well as the transport routes - within the framework of a certaim variation width - can be viewed as being largely constant (Figs. 1, 4 to 6).

In the South, and westwards of Jutland, on the basis of steep activity concentration gradients, one can clearly recognize the boundary between the water coming out of the Channel from the South and the water coming from the North along the English coast. In the surface water, this boundary between the two water masses lies between 6° E and 7° E (Figs. 4, 5).

However, in deep water westwards of Jutland, clear differences of the distribution are observed between the years 1982 and 1984. In August/September 1982, a clear front in the entire water column between surface and bottom is present. A comparable pattern, in principle, is indicated by the salinity distribution. In comparison with it, in May/June 1984, a clearly stronger advance of the deep water (on the basis of the activity concentration differences) in the direction of Jutland compared with the surface water is recognizable (Figs. 5, 6). Here also this effect appeared in outline to a certain extent in the salinity distribution.

On the way from the Sellafield Works near the Irish Sea as far as into the North Sea, a strong shift of the ^{137}Cs:^{90}Sr ratio in favor of the ^{90}Sr can be observed.By following the temporal course of that ratio at different measurement points , a transport time can be deduced of around 3 years between the Sellafield Works and the Pentland Firth; a further year would be needed to reach into the southeastern North Sea (Fig. 2). Measurements of the ^{134}Cs:^{137}Cs ratio lead to corresponding transport times (Table 8).

The ratio Cs:Sr furthermore, in the southeastern North Sea, permits a good distinction between the water masses coming from the North into the North Sea with respect to those that come through the Channel from the South (Fig. 3). These masses practically flow side by side until about the entrance to the Skagerrak.

The vertical distribution of the ^{137}Cs shows, as is to be expected, seasonally conditioned differences. However, effects also occur, which - on the basis of the data available to date - do not yet permit a clear explanation.

After a distinct maximum in 1978 - brought about by a particular strong influx during 1977 - the total quantity of ^{137}Cs present in the North Sea has receded to around a little more than one third of these peak values by the year 1984 (Table 5a). in 1984, it totalled- approximately 2400 TBq $^{137+134}Cs$ - only 0.5 % of the circa 481 000 TBq ^{40}K present in nature in the North Sea.

The ^{90}Sr (Figs. 7, 8) as well as the $^{239+240}Pu$ (Figure 9), present in the North Sea show an analogous distribution scheme compared with that of the ^{137}Cs. Their concentrations lie far below that of the ^{137}Cs (^{90}Sr in 1979 about 17.5 %; $^{239+240}Pu$ in 1980 about 0.03 %).

Only in limited regions of the southeastern North Sea, along the mainland coast, as far as into the Skagerrak, water bodies with temporally severely changing activity concentration values of ^{90}Sr (up to around the factor 3) were observed. This is particularly the case in the years 1983, 1984, and 1985 and may be traced back to an irregular emission of ^{90}Sr with the waste waters of the nuclear fuel reprocessing plant La Hague.

The transuranic nuclides $^{239+240}Pu$, ^{238}Pu, and ^{241}Am are present in the North sea in only very slight activity concentrations. Their distribution picture largely covers that of the ^{137}Cs (Figs. 9, 10).

Tritium (Fig. 11), on the other hand - with essentially higher concentrations - shows a completely different distribution, because it is principally introduced via the freshwater flowing in from land.

The distribution of the $^{137+134}Cs$ in the northern North Atlantic (Figs. 12, 13) illustrates well the previously known current picture in this region.

The ratio ^{134}Cs:^{137}Cs permits the estimation of the transport times from the Sellafield Works as far as into the northern North Atlantic. As far as Sptsbergen, the transport times lie at 6 to 7 years; as far as into the region eastwards of Greenland at 7 to 9 years (Fig.14).

The input of artificial radionuclides in May 1986 from the accident at Chernobyl via the fallout into the sea - apart from the possible occurring enrichment of individual radionuclides in the bottom sediments - may be without essential influence upon the conditions in the North Sea (Fig. 16) because owing to the constant water exchange - a relativeely quick transport into the North Atlantic takes place.

In the Baltic Sea, with its lesser water exchange, of course, the high activity concentrations in the surface water at the beginning will fall quickly to lower values owing to vertical mixing; on the other hand, the total quantity of the radionuclides introduced - regardless of their radioactive decay - will only very slowly decrease. For that reason, further measurements in this region are a matter of urgent necessity.

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REFERENCES

(1) KAUTSKY,H., C.N.MURRAY, 1981 :
Artificial radioactivity in the North Sea
Atomic Energy Rev. Suppl.No.2, 63 -105

(2) AARKROG,A., L.BØTTER JENSEN, H.DAHLGAARD, H.HANSEN, J.LIPPERT, S.P.NIELSEN, K.NILSSON, 1982 :
Environmental radioactivity in Denmark 1981, Risø-Rep. No.469

(3) AARKROG,A., S.BOELSKIFTE, E.BUCH, G.C. CHRISTENSEN, H.DAHLGAARD, L.HALLSTADIUS, H.HANSEN, E.HOLM, 1984 :
Environmental radioactivity in the North Atlantic Region. The Fare Islands and Greenland included
Risø-Rep. No.528

(4) HUNT,G.T., 1985 :
Radioactivity in surface and coastal waters of the British Isles.
Aquatic Environment Monitoring Report
MAFF Directorate of Fisheries Research Lowestoft. See also the same reports of former years.

(5) KAUTSKY,H. 1985
Distribution and content of different artificial radionuclides in the water of the North Sea during the years 1977 to 1981 (complemented with some results from 1982 to 1984
Dt.hydrogr.Z. 38, 193 - 224

(6) KAUTSKY,H., D.F.JEFFERIES, A.K.STEELE 1980 :
Results of the radiological North Sea Programme RANOSP 1974 to 1976
Dt.hydrogr.Z. 38, 152-157

(7) JEFFERIES,D.F., A.K.STEELE, A.PRESTON 1982 :
Further studies on the distribution of Cs137 in British coastal waters: Irish Sea. Deep-Sea Res.29, 713-738

(8) McKAY,W.A., M.S.BAXTER, 1985 :
Water transport from the North-east Irish Sea to western Scottish waters: Further observations from time-trend matching of Sellafield radiocaesium.
Estuar.coast.Shelf Sci. 21, 471-480

(9) McKAY,W.A., M.S.BAXTER, D.J.ELLETT, D.T.MELDRIN, 1987 :
Radiocaesium and circulation patterns west of Scotland
Continent.Shelf Res. (in press)

(10) McKINLEY,I.G., M.S.BAXTER, D.J.ELLETT, W.JACK, 1981 :
Tracer application of radiocaesium in the sea of the Hebrides
Estuar.coast.Shelf Sci. 13, 69-82

(11) KAUTSKY,H. 1986 :
Distribution and content of 137+134Cs and 90Sr in the water of the North Sea during the years 1982 to 1984
Dt.hydrogr.Z. 39, 139-159

(12) CALMET,D., P.GUEGUENIAT, 1985 :
Les rejets d'effluents liquides radioactifs du centre de traitement des combustibles irradiés de La Hague (France) et l'évolution radiologique du domaine marin.
Int.Atomic Energy Agency Vienna
TECDOC - 329

(13) KAUTSKY,H., 1981 :
Radiological investigations in the Western Baltic Sea including Kattegat during the years 1975 to 1980
Dt.hydrogr.Z. 34, 125-149

(14) LIVINGSTON,H.D., V.T.BOWEN, S.L. KUPFERMAN, 1981 :
Radionuclides from Windscale discharges. 1: Nonequilibrium tracer experiments in high latitude oceanography.
J.mar.Res. 40, 253-272

(15) LIVINGSTON,H.D., V.T.BOWEN, 1977 :
Windscale effluent in the water and sediments of the Minch.
Nature 169, 586-588

(16) KAUTSKY,H. 1973 :
The distribution of the radionuclide 137Cs as an indicator for North Sea watermass transport.
Dt.hydrogr.Z. 26, 241-246

(17) CAMBRAY,R.S., 1982 :
Annual discharge of certain long-lived radionuclides to the sea and the atmosphere from Sellafield Works, Cumbria 1957 to 1981
Harwell: United Kingdom Atomic Energy Authority. AERE-M 3269

(18) TRANGELED,S., 1974 :
Oceanography of the Norwegian and Greenland Seas and adjacent areas.
NATO SACLANTCEN Memorandum SM-47

(19) AARKROG,A., L.BØTTER-JENSEN, H.DAHLGAARD, H.HANSEN, J.LIPPERT, S.P.VILSON 1983 :
Environmental radioactivity in Denmark in 1982. Risø National Laboratory, Risø Report R-487

(20) AARKROG,A., H.DAHLGAARD, L.HALLSTADIUS, H.HANSEN, E.HOLM, 1983 :
Radiocaesium from Sellafield effluents in Greeland waters.
Nature 304, 49-51

(21) BONI,A.L., 1966 :
Rapid ion exchange analysis of radiocaesium in milk, wine, seawater, and environmental samples.
Analytic.Chem. 38, 89

(22) EICKE, H.-F., 1974 :
Eine Methode zur Anreicherung und Bestimmung sehr geringer Gehalte von 137Cs in ozeanischem Tiefenwasser
Dt.hydrogr.Z. 27, 20-26

(23) EICKE,H.-F., 1981 :
Distribution of transuranic isotopes in the surface water of the North Sea and adjacent regions.
in: Techniques for identifying transuranic speciation in aquatic environment, IAEA Vienna, 73-78

(24) WEDEKIND,CH., 1982 :
Tritium distribution and spreading in the North Sea and the Baltic Sea in 1980/81, as well as in the surface water of the North Atlantic in 1979.
Dt.hydrogr.Z. 35, 177-186

(25) DEUTSCHES HYDROGRAPHISCHES INSTITUT HAMBURG, 1987, Nr. 2149/34 :
Die Auswirkungen des Kernkraftwerksunfalls von Tschernobyl auf Nord- und Ostsee, August 1986, überarbeitet und ergänzt Januar 1987.
Meereskundl.Beobacht.und Ergeb. Nr.62

(26) KAUTSKY, H., 1987 :
Investigations on the distribution of 137Cs, 134Cs and 90Sr and the water mass transport times in the northern North Atlantic and the North Sea.
Dt.hydrogr.Z. 40, (in press)

Table 1a

Comparison of the content of $^{137+134}$Cs in the water of the North Sea with the discharge rates of the Sellafield Works (Cambray [1982])

Year	Discharge rate of $^{137+134}$Cs from Sellafield Works		North Sea content of $^{137+134}$Cs		Month of measurement
	TBq	Ci	TBq	Ci	
1974	5058	136 698	—	—	
1975	6312	170 538	—	—	
1976	5028	135 879	—	—	
1977	5073	137 098	—	—	
1978	4492	121 392	5836	157 727	Febr /March
1979	2798	75 618	5209	140 785	Sept /Oct
1980	3205	86 627	4621	124 885	August
1981	2524	68 225	2775	75 008	November
1982	—	—	3034	81 906	Aug /Sept

Table 1b

Comparison of the content of ^{90}Sr in the water of the North Sea with the discharge rates of the Sellafield Works

Year	Discharge rate of ^{90}Sr from Sellafield Works		North Sea content of ^{90}Sr		Month of measurement
	TBq	Ci	TBq	Ci	
1975	467	12 635	—	—	
1976	383	10 344	—	—	
1977	427	11 534	—	—	
1978	598	16 160	—	—	
1979	252	6 820	913	24 688	Sept /Oct
1980	352	9 500	—	—	

Table 2

Estimation of the yearly influx of ^{137}Cs and ^{90}Sr into the North Sea from other sources than nuclear facilities **(Kautsky and Murray [1981], Kautsky [1983])**

Assumptions for calculation

Medium	Average activity concentration			
	^{137}Cs		^{90}Sr	
	mBq/l	pCi/l	mBq/l	pCi/l
Atlantic water	4 4	0 12	3 7	0 1
Baltic Sea water	22	0 6	22	0 6
Runoff	1 9	0 05	19	0 5

Inflowing water		Content of			
		^{137}Cs		^{90}Sr	
	km^3	TBq	Ci	TBq	Ci
a) whole North Sea					
Runoff incl rivers	290	0 55	15	5 3	145
Atlantic from NW	9000	40	1080	33	900
Atlantic from N	8700	39	1044	32	870
near Dover	4500	20	540	17	450
Baltic Sea	500	11	300	11	300
Fallout		5 5	150	5 5	150
Total	22 990	116	3129	104	2815
b) western and central North Sea*					
Runoff**	51	0 11	3	0 92	25
Atlantic from NW	9000	40	1080	33	900
Atlantic from N	8700	39	1044	32	870
Fallout***		3 7	100	3 7	100
Total	17 751	83	2227	70	1895

* relevant to the ratio Cs Sr in the western and central North Sea

** runoff from the British Island only

*** two thirds of the total fallout guessed as relevant to the Cs Sr ratio

Table 3

Transport velocity along the British East coast, calculated from the position of the isolines of the ^{137}Cs concentration during the years 1977 to 1978

Isolines	Date	Coordinates	Distance	Days	Velocity
Bq m^{-3}			sm		sm/d
400	16 04 1977 24 02 1978	58° 15' N 2° 40' W 54° 40 N 0°	240	314	0 76
300	18 04 1977 28 02 1978	57° 40' N 1° 30' W 53° 10' N 1° E	290	316	0 91
200	18 04 1977 28 02 1978	56° 45' N 1° W 53° N 2° E	250	316	0 79
150	21 04 1977 28 02 1978	56° 30. N 1° W 52° 50 N 2° E	250	313	0 80

Table 4

Ratio of activity concentration c of $^{137+134}$Cs to activity concentration c of ^{90}Sr in the waste waters of La Hague and the Sellafield Works

Year	Ratio c_{Cs}'c_{Sr}	
	Sellafield Works	La Hague
1974	12 84	0 62
1975	13 50	0 51
1976	13 13	1 03
1977	11 88	0 82
1978	7 51	0 33
1979	11 10	0 22
1980	9 10	0 52
1981	9 09	1 19

Table 5a

Content of $^{137+134}Cs$ in the water of the North Sea during different years

Region	Year	Volume considered km^3	$^{137+134}Cs$ Activity found TBq	Ci	Mean activity concentration mBq/l	pCi/l
North Sea 51°N to 61°N 4°W to 9°E in the region of the Norwegian Deep is only the upper 100m layer included	1978	33 719	5 836	157 727	174	4.7
	1979		5 209	140 785	155	4.2
	1980		3 950	106 745	117	3.2
	1981		2 797	75 008	81	2.2
	1982		3 034	81 900	90	2.4
	1984		2 219	59 986	62	1.7
North Sea 51°N to 61°N 4°W to 9°E total water column	1982	42 444	3 424	92 436	81	2.2
	1984		2 621	70 866	62	1.7
58°30'N to 61°N westward of 3°E	1978	8 379	908	24 527	106	2.9
	1979		608	16 419	74	2.0
	1980		405	10 938	48	1.3
	1981		284	7 675	34	0.92
	1982		353	9 550	41	1.1
	1984		216	5 834	26	0.69
56°N to 58°30'N westward of 3°E	1978	8 063	2 527	68 306	315	8.5
	1979		1 881	50 823	233	6.3
	1980		1 094	29 556	137	3.7
	1981		717	19 381	89	2.4
	1982		1 119	30 213	138	3.7
	1984		646	17 452	78	2.1
51°N to 56°N westward of 3°E	1978	4 710	1 460	39 457	311	8.4
	1979		1 104	29 834	233	6.3
	1980		981	26 510	207	5.6
	1981		579	15 634	122	3.3
	1982		591	16 146	127	3.4
	1983		619	16 721	130	3.5
	1984		556	15 030	115	3.1
	1985		289	7 803	63	1.7
51°N to 56°N eastward of 3°E	1978	3 739	360	9 737	96	2.6
	1979		564	15 260	152	4.1
	1980		571	15 423	152	4.1
	1981		429	11 614	115	3.1
	1982		363	9 801	97	2.6
	1983		363	9 809	97	2.6
	1984		282	7 618	74	2.0
	1985		213	5 757	57	1.5
56°N to 58°30'N 3°E to 9°E upper 100m in the region of the Norwegian Deep	1978	5 799	530	14 313	93	2.5
	1979		837	22 632	144	3.9
	1980		622	16 821	107	2.9
	1981		505	13 636	89	2.4
	1982		486	13 122	84	2.3
	1984		392	10 594	67	1.8
56°N to 58°30'N 3°E to 9°E total water column	1982	9 984	723	19 521	74	2.0
	1984		621	16 788	62	1.7
58°30'N to 61°N eastward of 3°E upper 100m layer	1978	3 030	199	5 388	67	1.8
	1979		215	5 818	70	1.9
	1980		277	7 497	93	2.5
	1981		262	7 068	85	2.3
	1982		114	3 078	37	1.0
	1984		134	3 625	44	1.2
58°30'N to 61°N eastward of 3°E total water column	1982	7 569	267	7 209	35	0.95
	1984		301	8 144	40	1.1

Table 5b
Content of ^{90}Sr in the water of the North Sea during 1979

Region	Volume considered	^{90}Sr Activity found		mean activity concentration	
	km^3	TBq	Ci	mBq/l	pCi/l
51° 30′ – 61° N 4° W – 9° E	33 508	913	24 688	28	0.74
56° – 58° 30′ N west of 3° E	8 063	305	8 238	37	1.0
51° 30′ – 56° N west of 3° E	4 589	178	4 807	37	1.0
51° – 56° N east of 3° E	3 739	117	3 175	31	0.84
56° – 58° 30′ N 3° – 9° E	5 799	157	4 239	27	0.73
58° 30′ – 61° N east of 3° E	3 030	46	1 251	15	0.41

Table 5c
Content of $^{239+240}$Pu in the water of the North Sea during 1980

Region	Volume considered	$^{239+240}$Pu Activity found		mean activity concentration	
	km^3	TBq	Ci	µBq/l	fCi/l
51° – 61° N 4° W – 9° E	33 719	1.515	40.59	44	1.2
56° – 58° 30′ N west of 3° E	8 063	0.620	16.77	78	2.1
51° – 56° N west of 3° E	4 710	0.265	7.16	55	1.5
51° – 56° N east of 3° E	3 739	0.108	2.92	29	0.78
56° – 58° 30′ N 3° – 9° E	5 799	0.139	3.77	24	0.64
58° 30′ – 61° N east of 3° E	3 030	0.060	1.61	20	0.53

Table 5d
Content of Tritium in the water of the North Sea during 1981

Region	Volume considered	Tritium activity found		mean activity concentration	
	km^3	TBq	Ci	mBq/l	pCi/l
51° – 61° N 4° W – 9° E	33 719	25 766	696 379	760	20.6
56° – 58° 30′ N west of 3° E	8 063	5 083	137 386	630	17.0
51° – 56° N west of 3° E	4 710	3 825	103 372	810	21.9
51° – 56° N east of 3° E	3 739	4 086	110 438	945	25.5
56° – 58° 30′ N 3° – 9° E	5 799	4 832	130 596	830	22.5
58° 30′ – 61° N east of 3° E	3 030	2 799	75 651	930	25.0

Table 6 Annual discharges of ^{134}Cs and ^{137}Cs (TBq • y^{-1}) from the Sellafield Works and from La Hague.

	Sellafield Works			La Hague		
Year	^{134}Cs	^{137}Cs	$^{134}Cs : ^{137}Cs$	^{134}Cs	^{137}Cs	$^{134}Cs : ^{137}Cs$
1970	251	1 154	0.22	14	89	0.16
1971	236	1 325	0.18	48	242	0.20
1972	215	1 285	0.17	6.1	33	0.19
1973	166	768	0.22	8.4	69	0.12
1974	997	4 061	0.25	9.0	56	0.16
1975	1 081	5 231	0.21	4.3	34	0.12
1976	738	4 289	0.17	6.5	35	0.19
1977	594	4 478	0.13	9.5	51	0.19
1978	404	4 088	0.099	7.8	39	0.20
1979	235	2 562	0.091	3.6	23	0.16
1980	239	2 966	0.080	3.9	27	0.15
1981	168	2 357	0.071	6.0	39	0.15
1982	138	2 000	0.070	8.4	51	0.17
1983	89	1 200	0.074			
1984	35	434	0.080			

Table 7 Annual ratios of ^{134}Cs and ^{137}Cs in the discharges of the Sellafield Works and La Hague, decay corrected to the years 1982, 1983, 1984 and 1985. Half lifes of ^{134}Cs and ^{137}Cs are 2.07 and 30.02 years.

Release year	$^{134}Cs : ^{137}Cs$ Sellafield Works				$^{134}Cs : ^{137}Cs$ La Hague			
	1982	1983	1984	1985	1982	1983	1984	1985
1970	0.00514	0.00377	0.00276	0.00201				
1971	0.00577	0.00421	0.00299	0.00225				
1972	0.00738	0.00541	0.00396	0.00291				
1973	0.013	0.00955	0.00699	0.00512				
1974	0.020	0.015	0.011	0.0079	0.0113	0.0097	0.0071	0.0052
1975	0.023	0.017	0.012	0.0091	0.0139	0.0101	0.0074	0.0054
1976	0.026	0.019	0.014	0.010	0.029	0.021	0.015	0.011
1977	0.027	0.020	0.014	0.010	0.039	0.029	0.021	0.015
1978	0.028	0.021	0.015	0.011	0.058	0.042	0.031	0.023
1979	0.035	0.026	0.019	0.014	0.062	0.045	0.033	0.024
1980	0.042	0.031	0.022	0.016	0.080	0.058	0.043	0.031
1981	0.052	0.038	0.028	0.020	0.112	0.082	0.060	0.044
1982	0.070	0.051	0.037	0.027	0.174	0.127	0.093	0.068

Table 8 Activity concentrations of ^{134}Cs and ^{137}Cs and their ratios at three positions of the North Sea in the years 1983 and 1984

Date	Coordinates	Water depth m	Sampling depth m	Sample size l	Counting time h	^{137}Cs Bq·m^{-3} ±%1σ	^{134}Cs Bq·m^{-3} ±%1σ	Ratio ^{134}Cs:^{137}Cs	related to discharge year of Sellafield
1983									
1.12.	58°30'N 2°30'W	74	5	1800	250	204.6 ±0.1	6.66 ±0.5	0.033	1980
26.11.	53°30'N 1°00'E	27	5	2100	160	150.6 ±0.1	3.70 ±0.6	0.025	1979
5.12.	57°00'N 7°00'E	36	5	1880	125	105.1 ±0.1	2.37 ±1.0	0.023	1978/79
1984									
18.5.	58°30'N 2°30'W	72	5	2185	250	106.2 ±0.1	3.03 ±0.7	0.029	1981
28.4.	53°30'N 1°00'E	24	5	2200	250	132.5 ±0.1	3.15 ±0.6	0.024	1980
11.5.	57°00'N 7°00'E	34	5	2226	250	69.2 ±0.1	1.48 ±0.9	0.021	1980

Table 9 Activity concentrations of ^{134}Cs and ^{137}Cs and their ratios in the water of the *northern North Atlantic during 1982 (Counting time 250 hours).*

Date 1982	Coordinates	Water depth m	Sampling depth m	Sample size l	^{137}Cs measured Bq·m^{-3}±%1σ	^{137}Cs corr. Bq·m^{-3}	^{134}Cs measured Bq·m^{-3}±%1σ	Ratio ^{134}Cs:^{137}Cs	related to discharge year of Sellafield
9.6.	73°18'N 14°07'E	1074	5	441	27.9 ± 0.4	24.9	0.57 ± 6.2	0.023	1975
			100	1039	16.1 ± 0.4	(13.1)	0.33 ± 4.6	0.025	(1976)
22.6.	77°42'N 10°30'E	525	5	1009	11.5 ± 0.4	8.5	0.19 ± 7.1	0.022	1975
18.6.	75°52'N 7°50'W	2170	5	1016	8.58 ± 0.5	5.58	0.12 ± 11	0.021	1974/75
			100	1037	7.51 ± 0.5	(4.51)	0.12 ± 12	0.026	(1976)
5.6.	74°58'N 11°44'W	2170	5	871	7.62 ± 0.6	4.62	0.074 ± 20	0.016	1973/74
			100	1056	5.96 ± 0.6	(2.96)	0.067 ± 19	0.023	(1975)
1.6.	71°55'N 4°15'W	1400	5	924	7.07 ± 0.6	4.07	0.085 ± 16	0.021	1974/75

Table 10 Activity concentrations of ^{134}Cs and ^{137}Cs and their ratios in the water of the northern North Atlantic during 1983 (Sampling depth 10m, Counting time 250 hours)

Date 1983	Coordinates	Water depth m	Sample size l	^{137}Cs measured Bq·m^{-3} ± % 1σ	^{137}Cs corr. Bq·m^{-3}	^{134}Cs measured Bq·m^{-3} ± % 1σ	Ratio ^{134}Cs:^{137}Cs	related to discharge year of Sellafield
2.7.	68°00'N 12°11'E	225	504	55.5 ± 0.2	52.5	1.15 ± 3.0	0.022	1978
3.7.	71°32'N 25°04'E	300	498	41.1 ± 0.3	38.1	0.78 ± 3.8	0.020	1977
5.7.	76°15'N 16°52'E	280	465	12.4 ± 0.5	9.4	0.15 ± 23	0.016	1974/75
19.7.	79°30'N 10°10'E	100	496	11.5 ± 0.5	8.5	0.15 ± 18	0.018	1975/76
23.7.	81°06'N 17°50'E	450	495	11.1 ± 0.5	8.1	0.15 ± 17	0.018	1975/76
7.7.	81°31'N 05°42'E	800	490	11.5 ± 0.6	8.5	0.15 ± 17	0.018	1975/76
15.7.	79°45'N 12°00'W	220	507	6.3 ± 0.8	-	n.d.	-	-
16.7.	79°45'N 04°17'W	2000	499	6.3 ± 0.8	-	n.d.	-	-

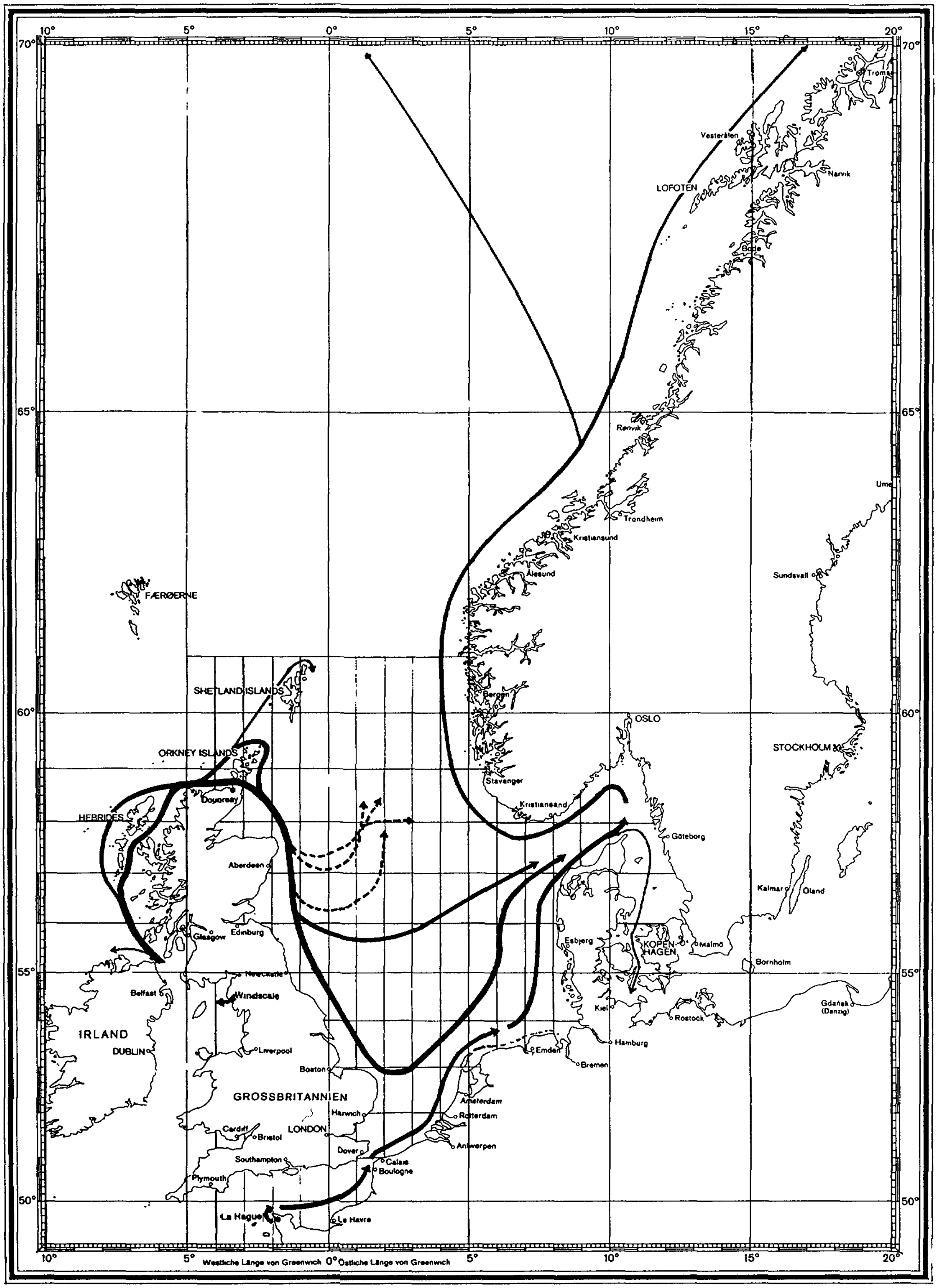
n.d. = not detectable

Table 11 Activity concentrations of ^{134}Cs and ^{137}Cs and their ratios in the water of the northern North Atlantic during 1984 and 1985 (Sampling depth 5m, Counting time 250 hours)

Date	Coordinates	Water depth m	Sample size l	^{137}Cs measured Bq·m^{-3} ± % 1σ	^{137}Cs corr. Bq·m^{-3}	^{134}Cs measured Bq·m^{-3} ± % 1σ	Ratio ^{134}Cs:^{137}Cs	related to discharge year of Sellafield
1984								
17.6.	62°00'N 04°01'E	223	2030	62.2 ± 0.1	59.2	1.26 ± 1.1	0.021	1979/80
19.6.	67°40'N 11°30'E	157	1910	45.9 ± 0.1	42.9	0.85 ± 1.5	0.020	1979
7.8.	70°55'N 18°55'E	280	2000	25.2 ± 0.2	22.2	0.41 ± 2.1	0.018	1979
21.6.	76°00'N 15°01'E	356	1957	11.5 ± 0.2	8.5	0.15 ± 4.5	0.0176	1978/79
1985								
16.6.	61°00'N 04°00'E	348	1719	61.3 ± 0.1	58.3	1.12 ± 2.7	0.019	1981
30.6.	67°00'N 10°59'E	262	1765	45.9 ± 0.1	42.9	0.75 ± 3.1	0.018	1980/81
10.7.	70°00'N 00°00'	3095	1980	7.58± 0.5	4.6	0.094 ± 22.3	0.020	1981
17.7.	71°00'N 22°00'E	285	1764	30.7 ± 0.1	27.7	0.49 ± 5.3	0.018	1980/81
31.7.	73°30'N 14°00'W	2480	1814	6.54± 0.6	3.5	0.043 ± 32.1	0.012	1976/79
5.8.	78°00'N 00°01'W	3080	1799	7.03± 0.5	4.0	0.081 ± 21.4	0.020	1981
11.8.	77°00'N 12°56'E	265	1698	10.9 ± 0.4	7.9	0.10 ± 27.8	0.013	1978/79

Table 12 Approximate transport times from Sellafield to different regions of the northern North Atlantic

Year	Coordinates		Region	discharge year related to Cs ratio	approx. transport time a	^{134}Cs counting error %
1982	73°18'	14°07'E	SW of Bear Island	1975	7	6
	77°42'	10°30'E	W of Spitsbergen	1975	7	7
	75°52'	07°50'W	E of Greenland	1974/75	7-8	11
	74°58'	11°44'W	E of Greenland	1973/74	8-9	20
	71°55'	04°15'W	Central Basin	1974/75	7-8	16
1983	68°00'	12°11'E	near Lofoten	1978	5	3
	71°32'	25°04'E	N of North Cape	1977	6	4
	76°15'	16°52'E	SSE of southern edge of Spitsbergen	1974/75	8-9	23
	79°30'	10°10'E	W northern Spitsberg.	1975/76	7-8	18
	81°06'	17°50'E	N of Spitsbergen	1975/76	7-8	17
	81°31'	05°42'E	NW of Spitsbergen	1975/76	7-8	17
1984	62°00'	04°01'E	W of southern Norway	1979/80	4-5	1
	67°40'	11°30'E	near Lofoten	1979	5	1.5
	70°55'	18°55'E	W of North Cape	1979	5	2
	76°00'	15°01'E	SSW of southern edge of Spitsbergen	1978/79	5-6	4.5
1985	61°00'	04°00'E	W of southern Norway	1981	4	2.7
	67°00'	10°59'E	near Lofoten	1980/81	4-5	3.1
	70°00'	00°00'	Central Norwegian Sea	1981	4	22.3
	71°00'	22°00'E	W of North Cape	1980/81	4-5	5.3
	77°00'	12°56'E	W southern Spitsbergen	1978/79	6-7	27.8
	78°00'	00°01'W	Central Greenland Sea	1981	4	21.4
	73°30'	14°00'W	E of Greenland	1978/79	6-7	32.1



Herausgegeben vom DEUTSCHEN HYDROGRAPHISCHEN INSTITUT, Hamburg

Transport routes of ^{137}Cs deduced from the measurements of the activity concentration distribution in the years 1971 to 1984. Dotted lines indicate temporal different transport routes.

Fig. 1.

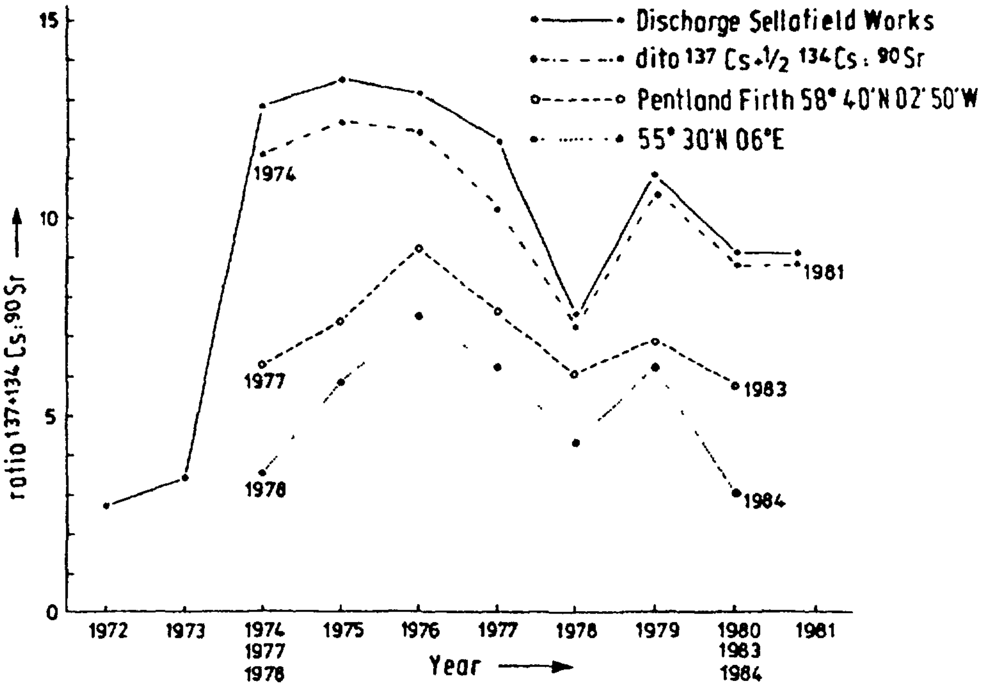


Fig 2 Comparison of the ratio of activity concentration of $^{137+134}$Cs to activity concentration of ^{90}Sr in the discharge from the Sellafield Works and at 2 positions in the North Sea

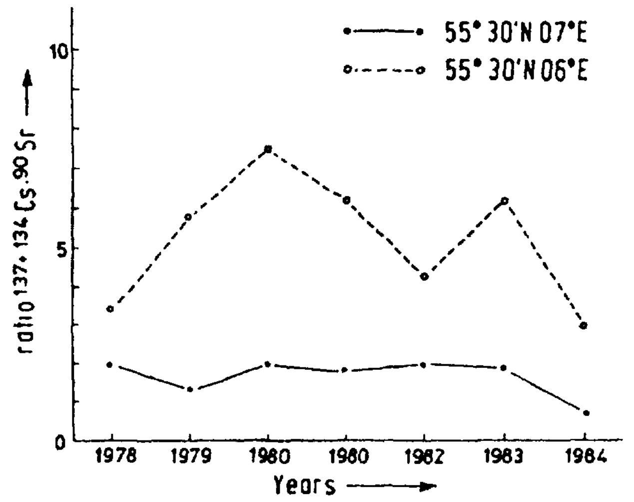


Fig. 3 Comparison of the ratio of activity concentration of ^{137}Cs to activity concentration of ^{90}Sr at 2 positions westwards of Jutland

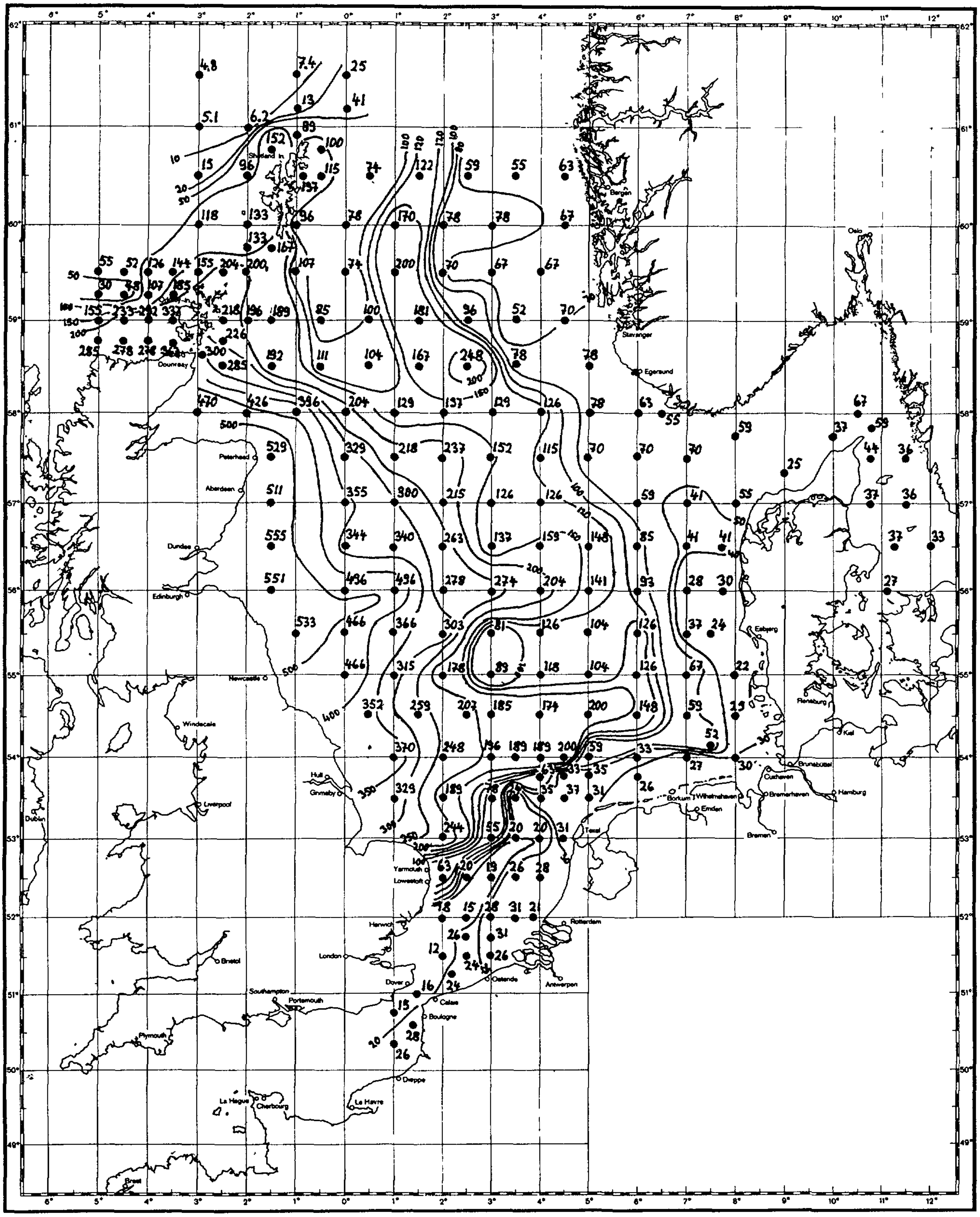


Fig.4

$^{137+134}$Cs in surface water

February/March 1978

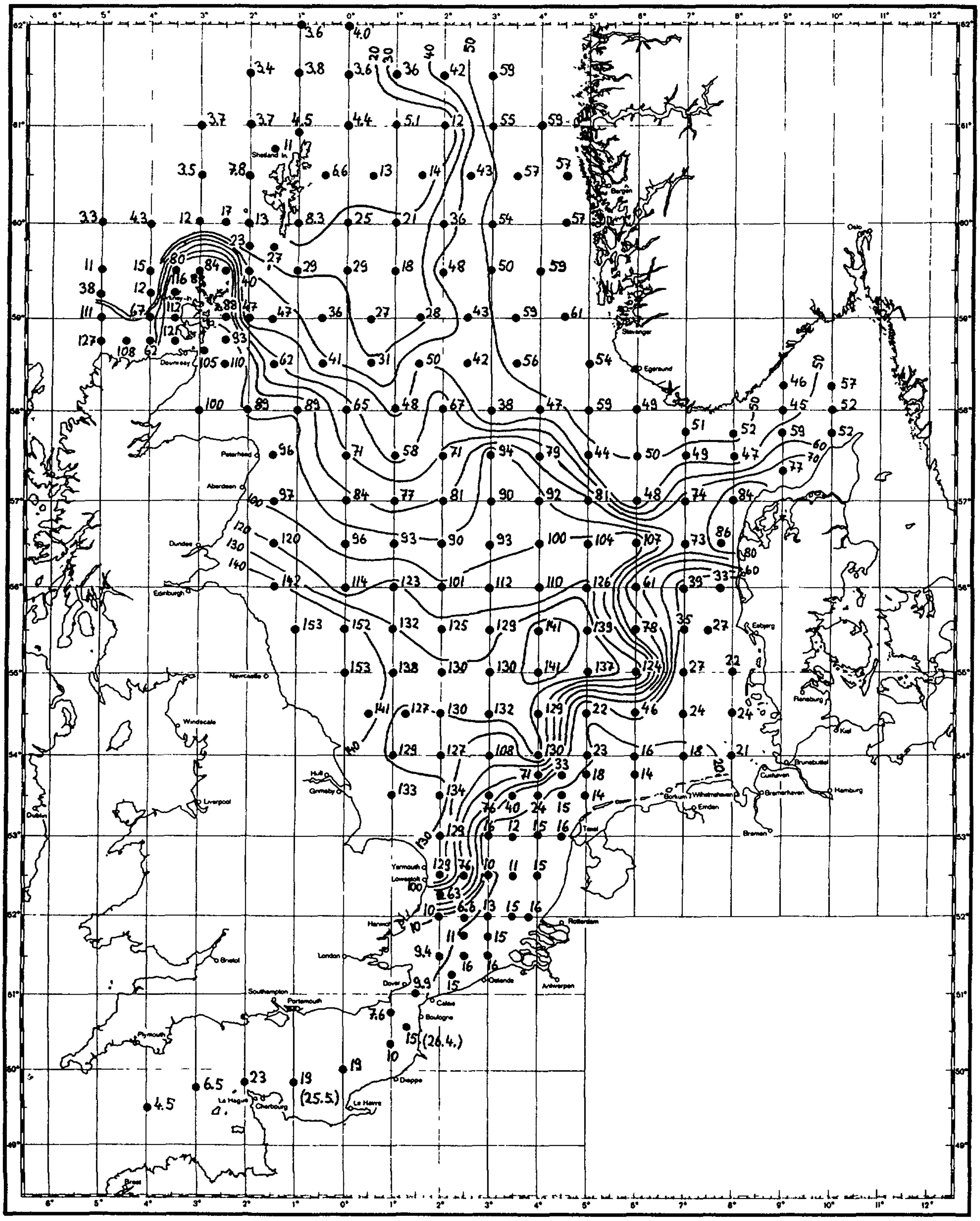


Fig. 5

$^{137+134}$Cs in surface water

April/June 1984

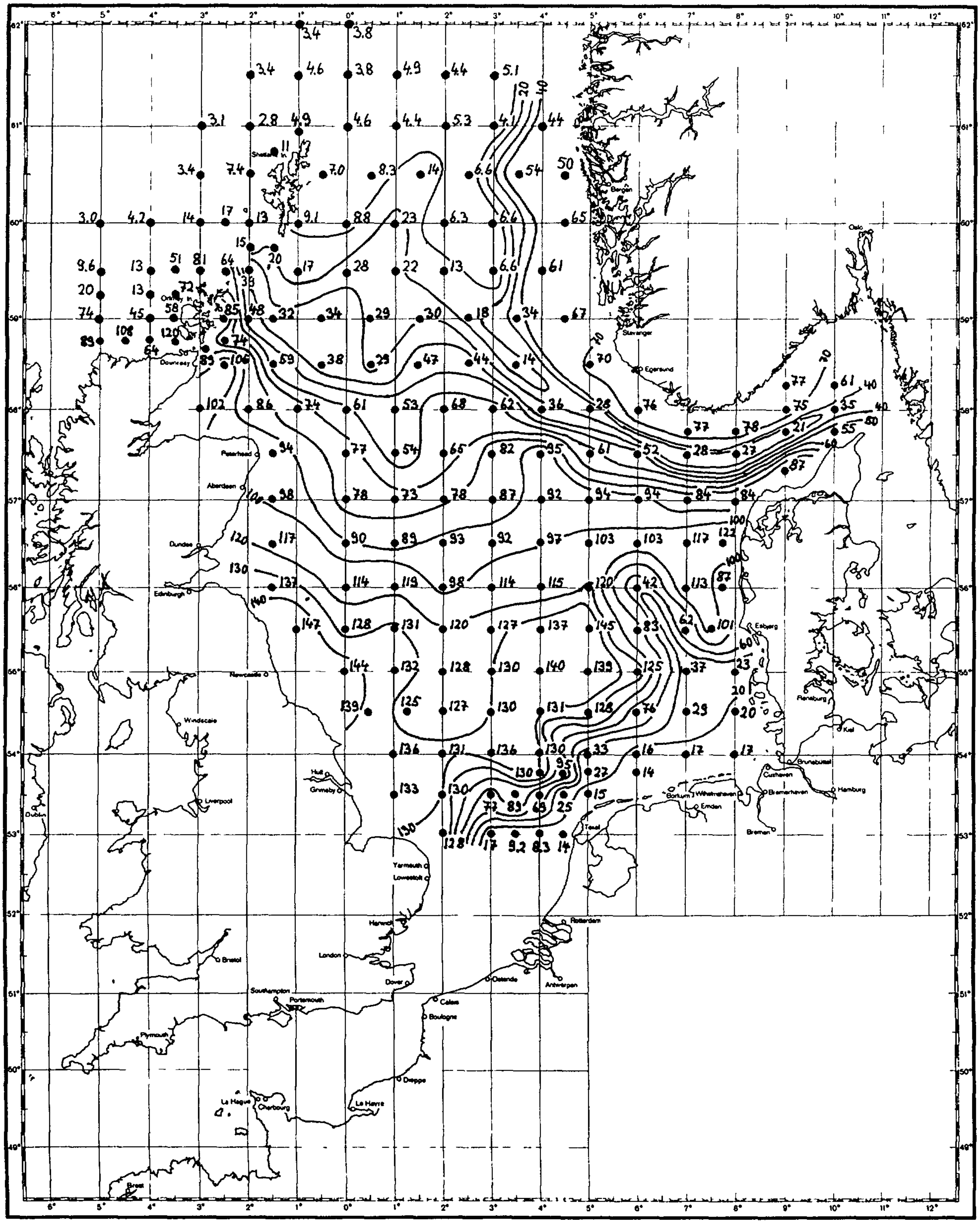


Fig. 6

$^{137+134}$Cs in 50m depth or near bottom down to 85m depth April/June 1984

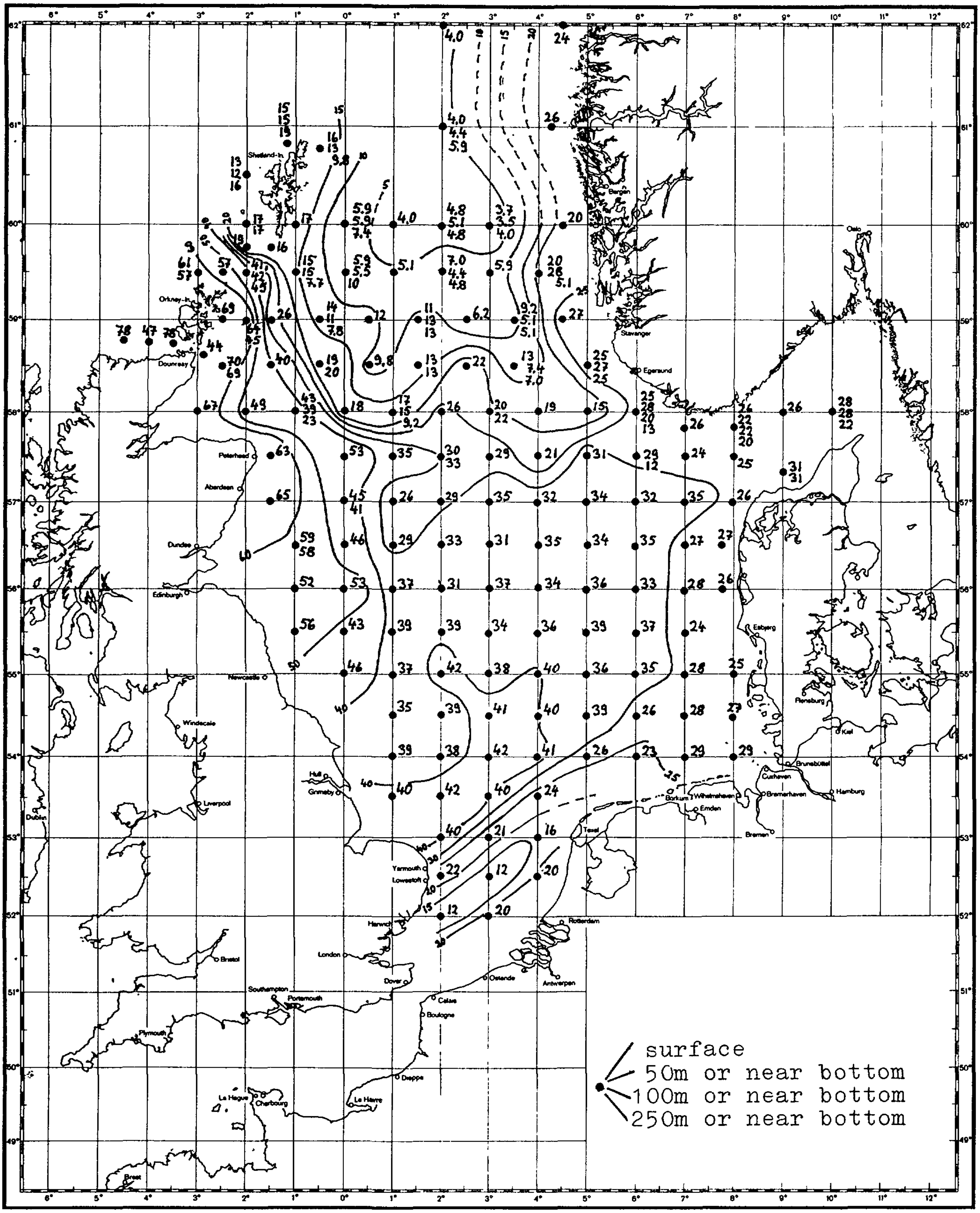


Fig. 7

^{90}Sr in different depths

September/October 1979

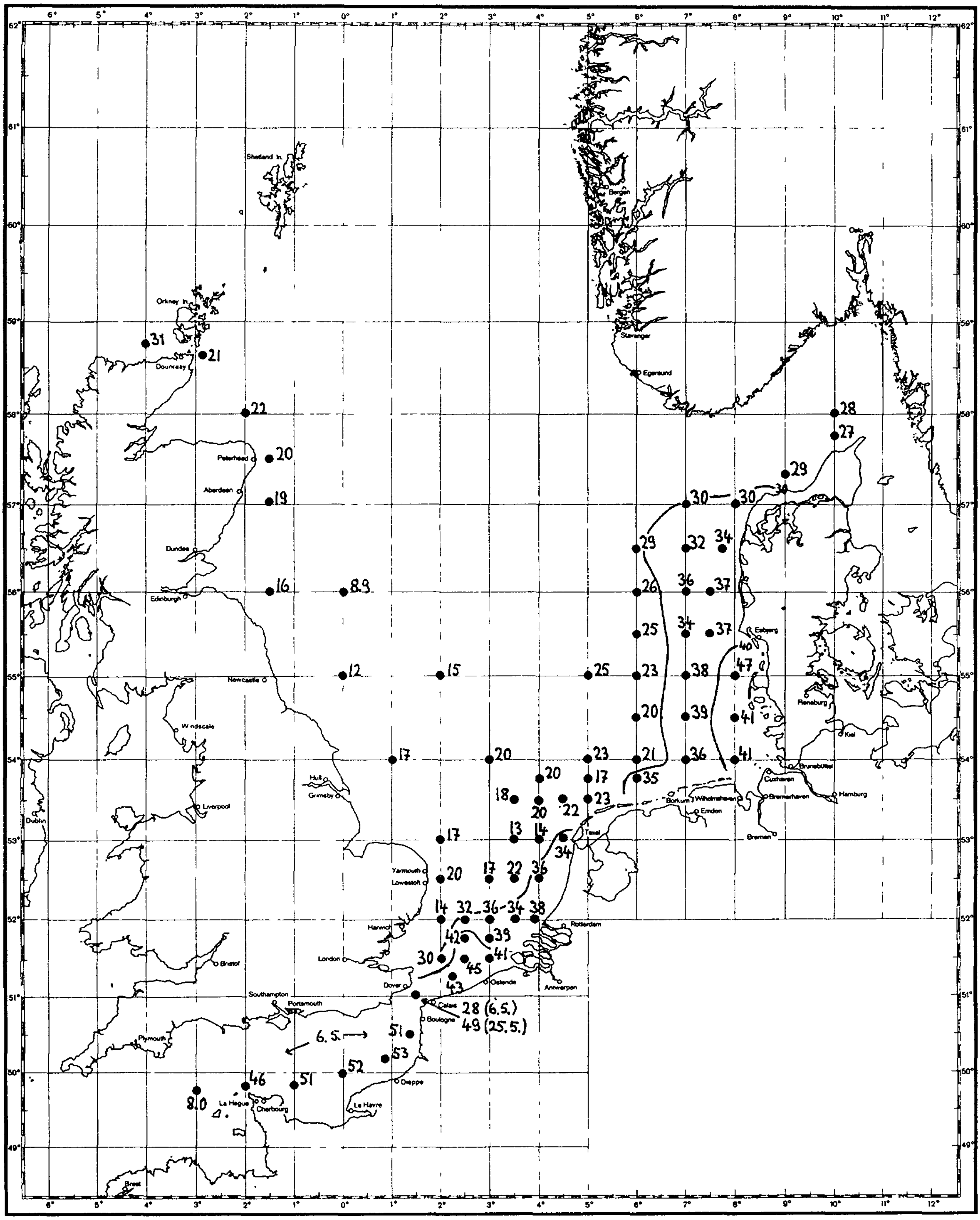


Fig. 8

^{90}Sr in surface water

May 1985

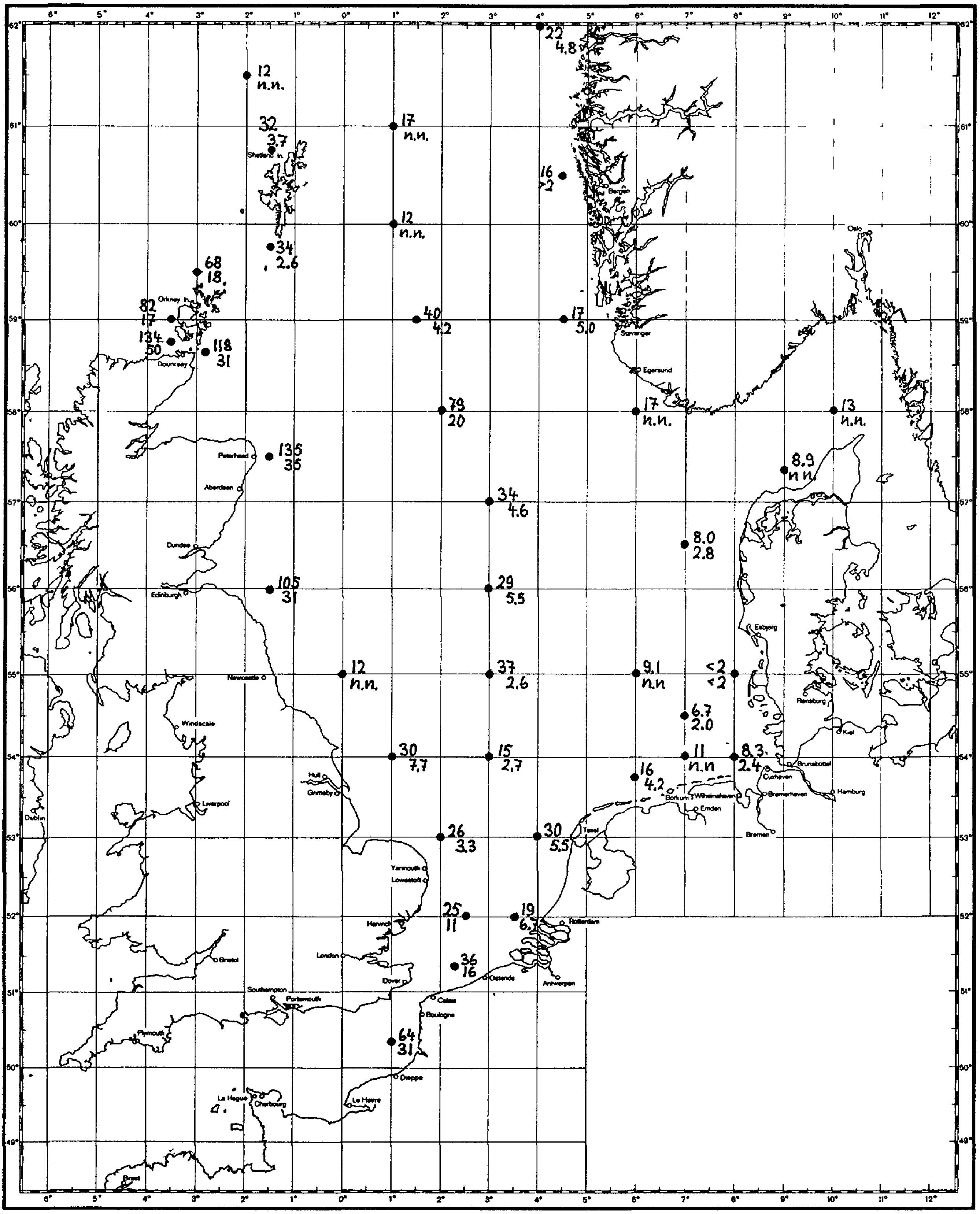


Fig. 9

$^{239+240}$Pu
$>$in surface water
^{238}Pu
August/September 1982

Fig.10

^{241}Am in surface water
August/September 1982

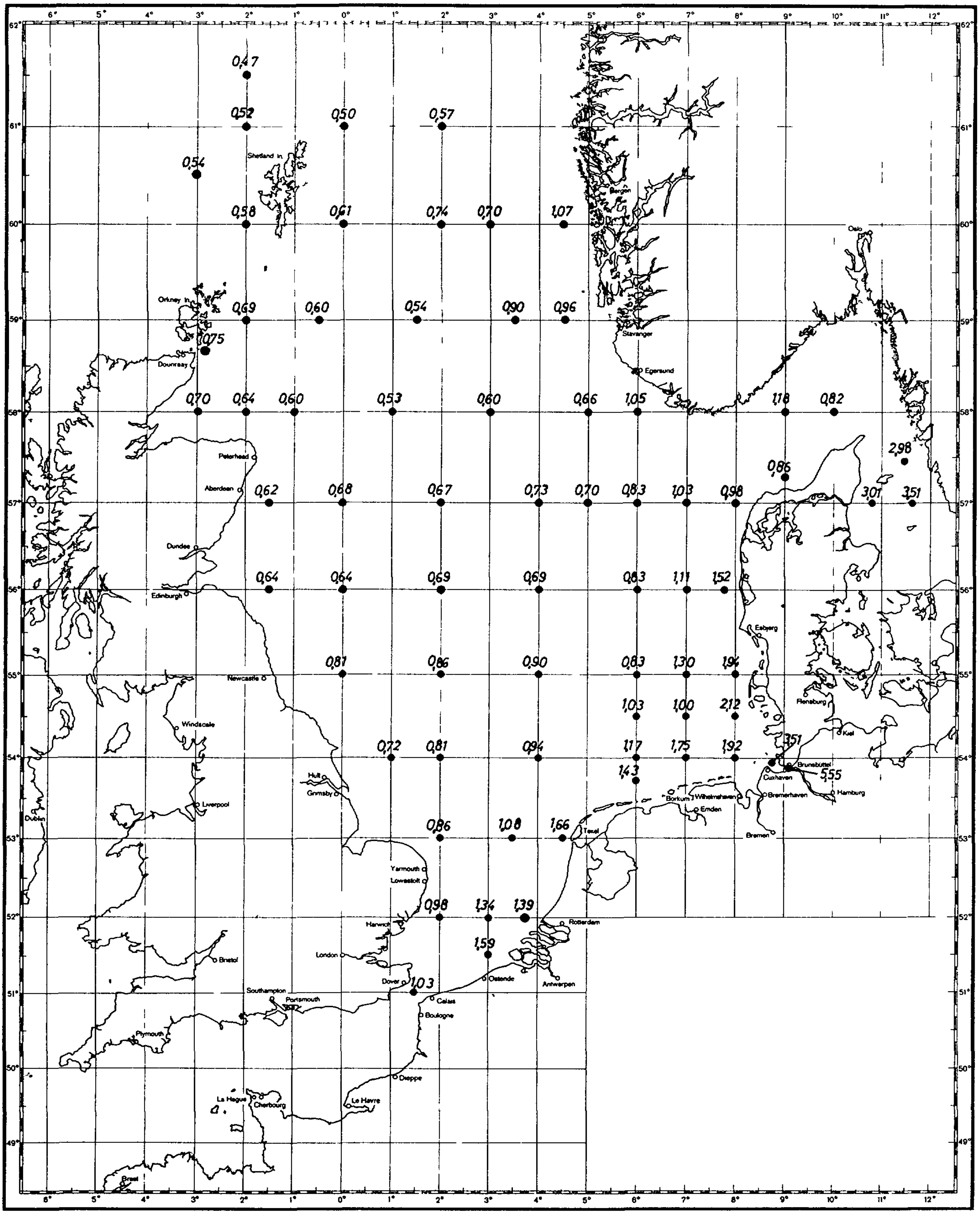


Fig.11
Tritium in surface water
November 1981

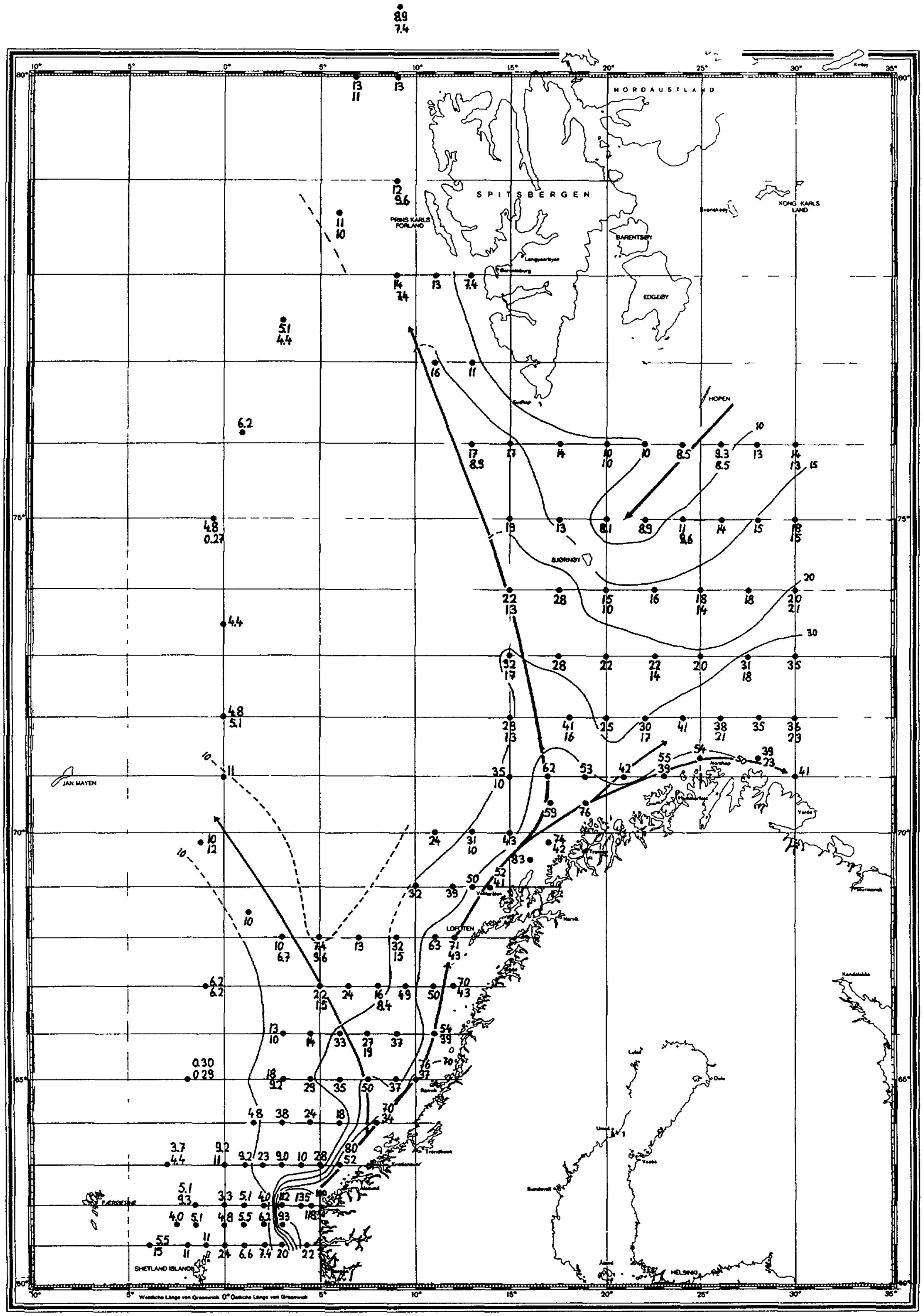


Fig. 12

$^{137+134}$Cs in surface water
and 100m depth
September 1979

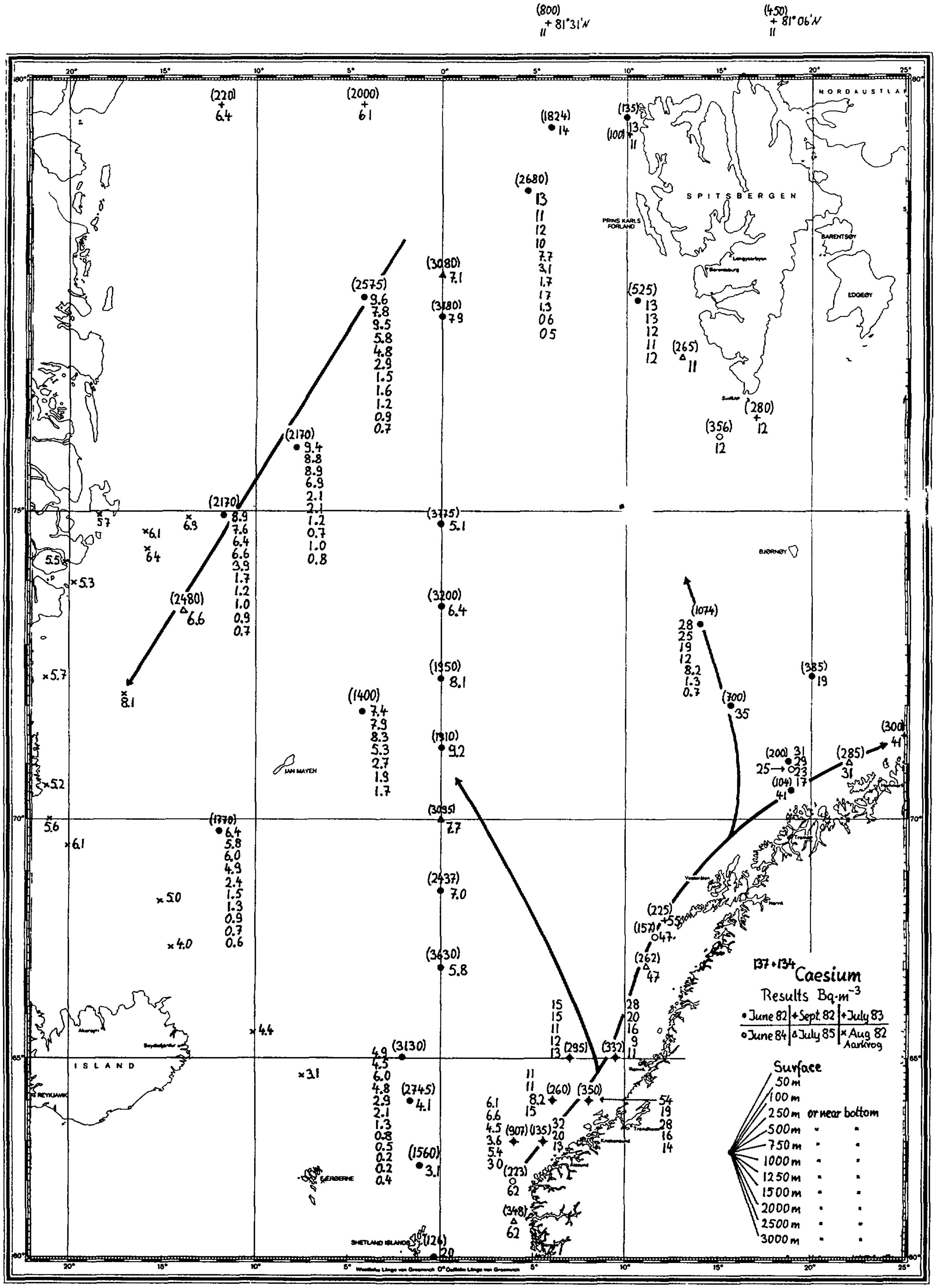


Fig.13

$^{137+134}$Cs in different depths
of the northern North Atlantic
in different years

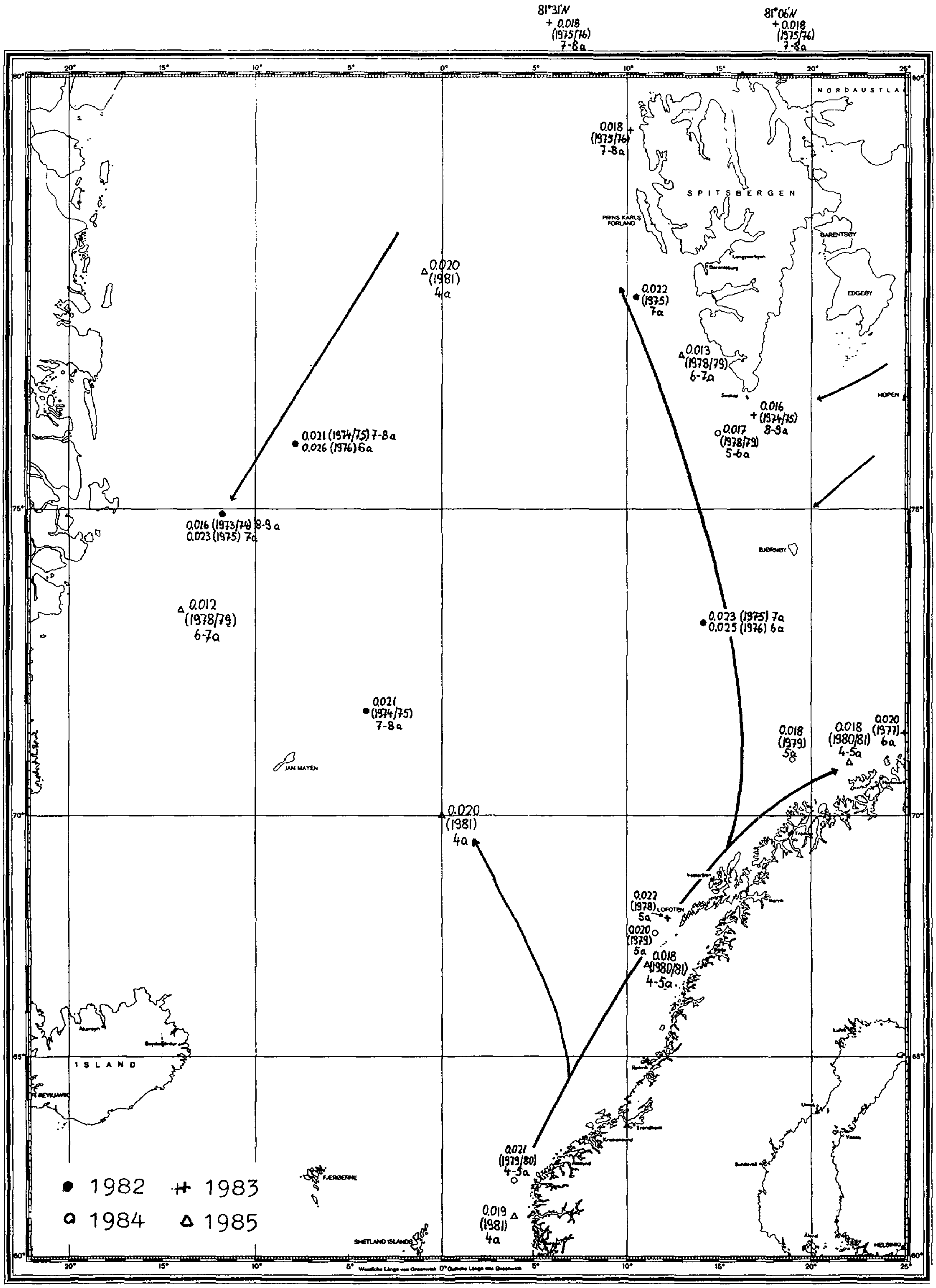


Fig.14

Ratio of ^{134}Cs : ^{137}Cs in the water (surface and 100m)

Figures in brackets indicate the relevant year of discharge of the Sellafield Works

Figures with "a" indicate the approximate transport time in years

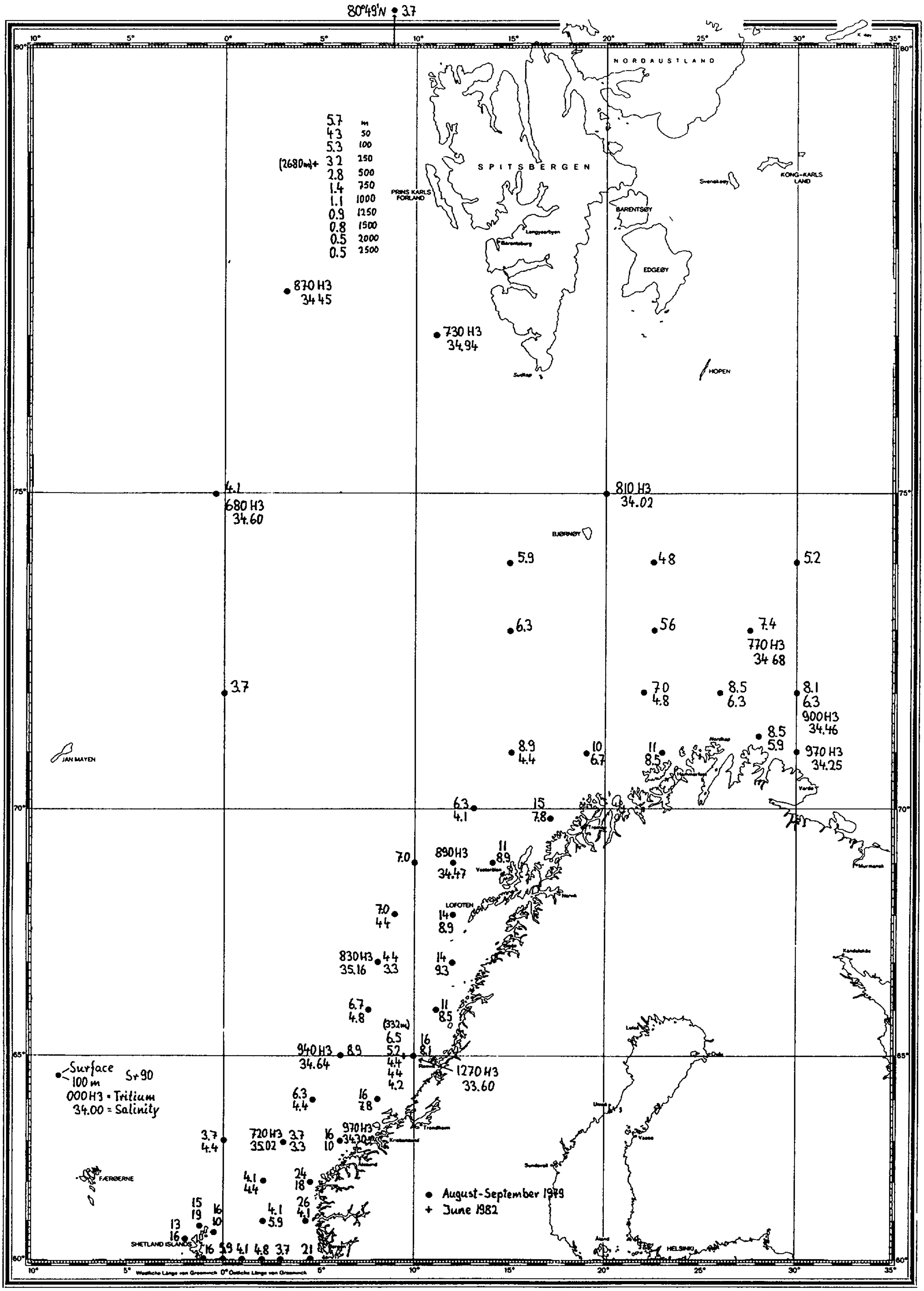


Fig. 15

Aug.-Sept.1979 /
June 1982

^{90}Sr surface and 100m depth
Tritium / Salinity surface Bq•m^{-3}

Fig.16

^{137}Cs in the water (Bq.m^{-3})

Figures in brackets = depth in m

Isolines: ratio ^{134}Cs:^{137}Cs

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WORLDWIDE DATA ON FLUXES OF $^{239,240}Pu$ AND ^{238}Pu TO THE OCEANS

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Abstract

The world's oceans contain 16 PBq of $^{239,240}Pu$, and the expected inventory is 12 PBq from weapons testing. The Irish sea contains 0.3 PBq in the sediments. The discrepancy between measurements and expectations may be due to an underestimate for fallout in rain. Data for ^{238}Pu are scarce.

1. Introduction

More than 70 per cent of the Earth's surface is ocean with a mean depth of approximately 4 kilometers. Our knowledge of plutonium inventories in the environment is based mainly upon measurements of radioactivity in soil and data from nuclear weapons testing. However, the last-mentioned source is classified information to some extent and thus not fully available.

In order to measure the content of plutonium in the oceans it is necessary to collect water from the surface to the bottom at certain intervals down through the water column. Sediments from the ocean floor should also be sampled. The plutonium concentrations in seawater are usually low. Hence rather large samples (> 50 litres) have to be collected from various depths. This takes time and ship time is limited and expensive. Thus the total number of deep ocean samples available for plutonium analyses are small considering the vast areas that need to be covered. Hence any estimate of the plutonium content in the world's oceans is encumbered with large uncertainties.

The purpose of this report is to summarize our present knowledge of plutonium inventories in the oceans and compare these measured values with those expected from global and local fallout, and from other sources.

The report will also discuss the distribution of plutonium between water and sediments. Local marine contaminations mostly in coastal waters will also be dealt with, and finally relevant references will be presented.

It is not intended in this report to examine the various processes (chemical, physical and biological) responsible for the observed distributions of plutonium in the marine environment.

2. General information on oceans and seas (Kossinna, 1921)

The total area of the Pacific Ocean including the Indian Ocean ($74.92 \cdot 10^6$ km^2) and marginal seas is $254.6 \cdot 10^6$ km^2. The volume is $1015.6 \cdot 10^6$ km^3 and average depth 3950 m.

The Atlantic Ocean including the Arctic Ocean ($14.09 \cdot 10^6$ km^2) and marginal seas cover a total area of $106.46 \cdot 10^6$ km^2. The volume is $354.7 \cdot 10^6$ km^3 and average depth 3330 m.

In Tables 2.1 and 2.2 the oceans have been divided into four latitude bands (National Academy of Science, 1971, Table 15, p. 71). In the inventory calculations I do not find it justified to use a finer division due to the lack of detailed data.

Table 2.1. Pacific Ocean Areas

Latitude	Area in 10^6 km^2	
> 30°N	41	N-Pacific: $102 \cdot 10^6$ km^2
0-30°N	61	
0-30°S	72	S-Pacific: $152 \cdot 10^6$ km^2
>30°S	80	

Table 2.2. Atlantic Ocean Areas

Latitude	Area in 10^6 km^2	
> 30^oN	35	N-Atlantic $54 \cdot 10^6$ km^2
0-30^oN	19	
0-30^oS	26	S-Atlantic $52 \cdot 10^6$ km^2
>30^oS	26	

In Table 2.3. are shown the areas of some European seas for which information on plutonium inventories (to some extent arising from local sources) are available.

Table 2.3. Areas of some European seas (marginal seas to the Atlantic Ocean)

Sea	Area in 10^6 km^2
Mediterranean Sea	2.97
North Sea	0.57
Irish Sea	0.10
Baltic Sea	0.42

3. Source terms

3.1. Nuclear weapons testing

According to UNSCEAR 1982 (Tables 21 and 22, p. 238) atmospheric nuclear tests have produced 13 PBq $^{239,240}Pu$ and 0.33 PBq ^{238}Pu. These are the source terms for global fallout.

In the Northern Hemisphere the integrated deposition density of $^{239,240}Pu$ is 39 Bq m^{-2} and in the Southern it is found to be 9.7 Bq m^{-2}. For ^{90}Sr the corresponding figures are 1804 and 565 Bq m^{-2}, respectively. The ratio $^{239,240}Pu/^{90}Sr$ in the Northern Hemisphere thus becomes 0.0216, while in the Southern it is 0.0172. UNSCEAR 1982 (Table 6, p. 230) gives detailed information on the distribution of ^{90}Sr in 10 degree latitude bands. From the above ratios we may estimate the global distribution of $^{239,240}Pu$ as shown in Table 3.1.1.

Table 3.1.1. Integrated deposition densities of $^{239,240}Pu$

Latitude band	Bq $^{239,240}Pu$ m^{-2}
> 30°N	51.1
0-30°N	26.8
0-30°S	9.1
>30°S	10.4

When deposition densities are now applied to the areas given in Tables 2.1 and 2.2. we get Tables 3.1.2. and 3.1.3., respectively.

Table 3.1.2. Expected $^{239,240}Pu$ inventories from global fallout in the Pacific Ocean

Latitude band	$^{239,240}Pu$ inventory in PBq
> 30°N	2.10
0-30°N	1.63
0-30°S	0.66
>30°S	0.83

Table 3.1.3. Expected $^{239,240}Pu$ inventories from global fallout in the Atlantic Ocean

Latitude band	$^{239,240}Pu$ inventory in PBq
> 30°N	1.79
0-30°N	0.51
0-30°S	0.24
>30°S	0.27

However besides the global fallout 4 PBq $^{239,240}Pu$ have been deposited locally around sites of nuclear explosions (Wrenn, 1974). Most of this fallout has probably been deposited in the Pacific area, where Glasstone (1962) estimated that 119 PBq ^{90}Sr at the beginning of 1956 were close-in fallout. Considering that the $^{239,240}Pu/^{90}Sr$ ratio in close-in fallout probably is higher than in global fallout it is evident that most of the 4 PBq $^{239,240}Pu$ was deposited in the Pacific. Hence the total expected inventory in the Pacific Ocean may be as high as 9.22 PBq $^{239,240}Pu$. For the Atlantic Ocean the expected inventory becomes 2.81 PBq (see Table 3.1.3.).

3.2. Debris from satellite reentry

According to UNSCEAR 1982 the ratio $^{238}Pu/^{239,240}Pu$ in global fallout is 0.0254; this ratio does not differ significantly between the two hemispheres. However, a satellite containing plutonium reentered in 1964 in the Southern Hemisphere increasing the measured ^{238}Pu levels especially in the Southern Hemisphere beyond those expected from nuclear weapons testing. The total amount of ^{238}Pu deposited from this satellite failure was 0.51 PBq or nearly twice the amount from all nuclear weapons testing. (Linsalata, 1980).

Approximately 22% (Linsalata, 1980) of the ^{238}Pu from the satellite is found in the Northern Hemisphere, while the remainder is in the Southern. This changes the global fallout $^{238}Pu/^{239,240}Pu$

ratio in the Northern Hemisphere to approximately 0.036 while in the Southern (away from close-in fallout areas) it becomes 0.18.

About 1.7 PBq ^{238}Pu and 1.2 TBq ^{239}Pu were dropped into the Pacific Ocean near Tonga Trench when an Apollo lunar probe aborted in flight (Dobry, 1980). It is thought that the plutonium has since remained intact in its containers at the bottom of the ocean.

3.3. Discharges from Nuclear reprocessing facilities

According to Cambray (1982a) and British Nuclear Fuels (1985) Sellafield in the U.K. had discharged 0.60 PBq ^{239,240}Pu to the Irish Sea by the end of 1984. The information on ^{238}Pu discharge is incomplete; however if we assume ^{238}Pu/^{239,240}Pu = 0.31 ± 0.02 (N = 7; ± 1 S.D.), which is the mean ratio observed during 1978-1984, the estimated total release of ^{238}Pu becomes 0.19 PBq. The discharges of plutonium from Sellafield to the Irish Sea are decreasing; in 1984 only 8.3 TBq ^{239,240}Pu were discharged.

Cap de la Hague in France has discharged 2.6 TBq Pu to the English Channel in the period 1966-1982 (Calmet, 1985). The ratio ^{238}Pu/ ^{239,240}Pu in environmental marine samples close to Cap de la Hague is 0.5. We may thus assume that 2/3 or 1.7 TBq have been ^{239,240}Pu and 0.9 TBq were ^{238}Pu. The annual Pu discharge in 1982 from Cap de la Hague was 0.19 TBq, or less than 2% of the discharge from Sellafield in 1984.

Other Pu releases from nuclear installations to the marine environment are insignificant compared with those from reprocessing stations.

3.4. Loss of nuclear weapons

In 1968 a B-52 bomber carrying four nuclear warheads crashed on the ice-covered Bylot Sound 11 km southwest of Thule Airbase in NW-Greenland. Approximately 1 TBq ^{239,240}Pu from this accident is located in the marine sediments in Bylot Sound. Most of the plutonium was removed from the ice and stored at the Savannah River plant. (Aarkrog, 1984). A similar accident in 1966 at Palomares in Spain did not contaminate the marine environment. (Iranco, 1980).

4. Data on plutonium in the marine environment

4.1. Pacific Ocean

The GEOSECS (Geochemical Ocean Sections) programme has produced nearly all data available for an estimate of the plutonium inventories in the Pacific Ocean. (Livingston, 1985a). Large volume seawater samples were collected from 1973 to 1974 in the Pacific and the radionuclide inventory in the water column at each station was calculated and expressed as mCi ^{239,240}Pu km^{-2}. From these measurements the authors have calculated the inventory in the North Pacific Ocean as 6.44 PBq ^{239,240}Pu (Bowen, 1980a).

The GEOSECS programme has not covered all parts of the Pacific Ocean. Let us suppose that we have the four latitude bands shown in Table 4.1, and want to have information on all longi-

Table 4.1. Estimated ^{239,240}Pu inventories in latitude bands (Table 3.1.2.) in the Pacific Ocean, based upon GEOSECS data and a few more recent observations (cf. Appendix Tables A1 and A2)

Latitude band	Mean deposit Bq m^{-2}	Inventory PBq
> 30°N	102	4.2
0-30°N	93	5.7
0-30°S	33	2.4
>30°S	?	?

tude bands between 140°E and 120°W. The missing values in such a sampling network may then be calculated (cf. the Appendix Table A3) from the observed values by an analysis of variance (anova), in which we determine the missing values assuming that there is no interaction between latitudes and longitudes (This assumption is in fact supported by the anova) (Vestergaard, 1964). It is the same principle as used in Bowens estimate above, where it was assumed that the longitudinal pattern of inventory increases from east to west, in the same manner as was found in the 30-40°N band. The present calculation is however more sophisticated as it includes not only the tendency seen in the 30-40°N band, but all latitudinal and longitudinal variations given by the data.

Nagaya (1984) found a mean deposit in 1980-1982 of 110±35 Bq ^{239,240}Pu m^{-2} (N = 5; ± 1 S.D) in the water column at five stations in the central North Pacific (betw. 25°N and 40°N and between 156°E and 170°E).

This is lower than the Geosecs data for this region (cf. Table A.1 in the appendix), but since 1973-74 some of the Pu may have entered the sediments.

Table 4.1 shows that the Pacific inventory according to the GEOSECS data may exceed 12 PBq ^{239,240}Pu. This inventory is considerably higher than that expected from global fallout alone. The estimate for the North Pacific is also higher than that of Bowen (9.9 compared with 6.4). However, if the Pu contribution from close-in fallout is 4 PBq in the Pacific Ocean (cf. 3.1), there is closer agreement between observations and predictions The estimated inventory is nevertheless at least 50% higher than that expected. We have no estimate for the > 30°S latitude band as no samples were collected in this part of the ocean. We may assume the inventory in this band to match that in the 0-30°S band (due to a possible contribution of close-in fallout in the 0-30°S latitude band) or possibly be a little lower.

In these calculations the contribution of Pu in sediments has been neglected. Four samples collected in 1980 between 30°N and

40°N and between 156°E and 170°W (cf. Appendix Fig. A4) contained 8.5 ± 3.9 Bq ^{239,240}Pu m^{-2} (± 1 S.D.) (Livingston 1986). Miamoto (1985) reported sediment values from 1981, 1982 and 1983 from 30°N and 147°E. He found 4.4, 3.3 and 3.3 Bq m^{-2}. Compared with the mean inventory in the water columns from the above areas, which was 128 Bq ^{239,240}Pu m^{-2} (cf. Table A.1 in the Appendix), the sediments contained 3-10% of the total inventory. Close to the test areas in the Pacific the sediments contain a relatively higher percentage of the total inventory. However, the inventory in sediments does not add significantly to the total inventory of ^{239,240}Pu in the Pacific Ocean.

There is a transfer of 4.7x10^{4} km^{3} yr^{-1} (Lewis, 1982) from the Pacific to the Arctic Ocean through the Bering Strait. This is only 47·10^{-6} of the total volume of the Pacific Ocean and thus of no importance for the present inventory calculations.

The vertical distribution of the Pu-activity in the Pacific water column shows a distinct subsurface concentration maximum in a depth of approximately 450 m (in 1973). This maximum had moved further down to approximately 800 m in 1982 (Appendix, Figs. A2 and A3). Furthermore, a high concentration layer close to the bottom is observed at several stations. The very few Pacific water columns collected since GEOSECS do not suggest any significant change in the Pu inventories.

4.2. Atlantic Ocean

In the Appendix, Table A4 shows the GEOSECS data used for the inventory calculations in the Atlantic Ocean. The inventories in the four latitude bands shown in Table 4.2 were calculated under the assumption that there was no variation with longitude. The contributions from each ten-degree latitude band were calculated separately before they were combined into the broader latitude bands. The areas of these 10-degree latitude bands were obtained from Kupfermann (1982).

Two deep-water samples collected close to the North Pole in 1979 (Livingston, 1984) showed 35 and 24 Bq ^{239,240}Pu m^{-2} in the water

Table 4.2 Estimated ^{239,240}Pu inventories in latitude bands (Table 3.1.2) the Atlantic Ocean based upon GEOSECS data (Appendix Table A.4)

Latitude band	Mean deposit Bq m^{-2}	Inventory PBq
> 30°N	67	2.35
0-30°N	23	0.44
0-30°S	16*	0.42
>30°S	18	0.47

*) The mean deposit at 0-30°S was assumed equal to that found at 0-10°N

columns. If we suppose these samples to be representative for the entire Arctic Ocean ($14.09 \cdot 10^{12}$ m^{-2}) the inventory in the ocean becomes 0.42 PBq ^{239,240}Pu or 18% of the total inventory in the North Atlantic Ocean north of 30°N.

The measured inventory in the North Atlantic Ocean is 2.79 PBq and the predicted value is 2.30 PBq ^{239,240}Pu. The uncertainties in the two figures exceed the difference between these values. The North Atlantic also contains some Pu from other sources than global fallout, first of all from reprocessing of nuclear fuel. However, this contribution is of minor importance as most of this activity is retained in coastal sediments close to the discharge points (Pentreath, 1985).

In the South Atlantic the difference between measured and expected Pu-inventories is greater than in the North Atlantic. However the measured values are very few, and with large errors we may again conclude that the difference is insignificant.

North Atlantic sediments have been analysed by Noshkin (1983) from the NEA dumpsite (~ 46°N ~ 17°W). He found 5.9 and 8.2 Bq

^{239,240}Pu m^{-2}. In deep sea cores on the continental rise and abyssal plain at (~ 32°N, 70-72°W) Buessler (1985) has found 3.7± 1.5 (N = 11; ± 1 S.D.) Bq ^{239,240}Pu m^{-2}. Bowen (1980b) reports inventories in open Atlantic (~ 38°N, ~ 72°W) sediments collected in 1975 and 1976 ranging from 1 to 7 Bq ^{239,240}Pu m^{-2}. Compared with neighbouring column inventories (cf. Appendix, Table A.4) the North Atlantic sediments contained approximately 5% of the total inventory. As for the Pacific we may conclude this amount to be unimportant for the total inventory estimate.

4.3. Coastal seas

4.3.1. General

Most data on Pu in the marine environment have been obtained from coastal waters. The majority of water samples have thus been surface samples and the sediments have preferentially been collected from shallow waters. The inventories of Pu found in coastal waters are however small compared to those found in the open oceans. The only exception to this is the contamination of the waters around the U.K. where a few percents of the global Pu-inventory are to be found, mainly due to authorized discharges from the Sellafield reprocessing plant.

This report will not give detailed information on measurements of Pu in surface waters and sediments. Relevant publications containing such information are included in the list of references. However the estimates of Pu inventories in some of the most important European Seas are summarized below. Furthermore, information on run-off with rivers are presented.

4.3.2. The Irish Sea

According to Pentreath (1985) 0.28 PBq ^{239,240}Pu is associated with the sea bed of the Irish Sea (see Appendix Figs. A.6 and A.7). Most of this comes from the authorized discharges from Sellafield. The water contained about 1% (1978-1979) of the total ^{239,240}Pu inventory in the Irish Sea. As the discharges of Pu are decreasing (BNF, 1985) this percentage will decrease as well.

We may conclude that around 50% (cf. 3.3.) of all ^{239,240}Pu discharged from Sellafield is found in the sediments in the Irish Sea. Probably less than 0.3 PBq ^{239,240}Pu has thus entered the North Atlantic water masses. According to Holm (1986) and Hallstadius (1986) about 80% of the plutonium released in dissolved form from reprocessing facilities in Western Europe is removed by sedimentation processes before reaching the Arctic Ocean.

4.3.3. The North Sea

Between 51°N and 61°N and between 4°W and 9°E Kautsky (1985) has found 1.5 TBq ^{239,240}Pu in the water of the North Sea. The sediment inventory has not been measured, but is assumed to be about two orders of magnitude higher (cf. 4.3.2. and 4.3.4.).

4.3.4. The Baltic Sea

Salo (1984) has calculated the ^{239,240}Pu inventories in the Baltic Sea from sediment and water measurements. In the sediments she estimates an inventory of 15 TBq, in the water 0.2 TBq, and in biota a few GBq ^{239,240}Pu.

4.3.5. Mediterranean Sea

Livingston, (1976) has measured the mean ^{239,240}Pu inventory in six sediment cores from the Mediterranean Sea to 4.3 ± 2.2 Bq m^{-2} (N = 6; ± 1 S.D). This level is similar to that found in the open oceans.

Fukai (1980b) analysed water column samples collected at four locations in the Mediterranean Sea (North-western, Tyrrhenian, Ionian and Levantine Basins) in 1976-1977. He found 52 ± 8 Bq ^{239,240}Pu m^{-2} (N = 4; ± 1 S.D). If we add a sediment inventory of 4 Bq m^{-2}, we get 56 Bq m^{-2}. The area of the Mediterranean Sea is $2.97 \cdot 10^{12}$ m^2 and the ^{239,240}Pu inventory thus becomes 0.17 PBq.

In 1975 Bowen (1979) found a total inventory of 100 Bq $^{239,240}Pu$ in the Ionian Sea (35°56'N, 17°55'E), which is higher than that reported by Fukai (49 Bq m^{-2} in the water column). Murray (1978) analysed water columns collected 1973-1974 at 4 stations in the North-western basin and found approximately 80 Bq m^{-2} compared with Fukai's 49 Bq m^{-2}. The differences between the various estimates may be due to increasing transfer with time of Pu from the water column to the sediments.

Compared with the expected inventories from fallout, the Mediteranean Sea seems to contain slightly less plutonium than expected (Fukai 1980). However the difference is hardly significant considering the uncertainties in the data.

Fukai (1979) found a $^{238}Pu/^{239,240}Pu$ ratio of 0.05 in Mediterranean seawater.

4.3.6. Run off from land with rivers

Beasley (1983a) estimated that 0.15-0.30 TBq $^{239,240}Pu$ have entered the North-East Pacific Ocean by river (Columbia) transport since the late 1950's.

The annual discharges of $^{239,240}Pu$ to the Gulf of Mexico with suspended sediments in the Mississippi River have been measured as 0.07 TBq in 1977 and as 0.02 TBq in 1983 (Scott, 1985).

The run-off $^{239,240}Pu$ from the Hudson River (Olsen, 1981) has totalled 0.015 TBq in the period 1955-1975.

The annual input of $^{239,240}Pu$ through rivers into the Mediterranean Sea has been estimated as 0.02 TBq. (Fukai, 1980).

We may thus conclude that run-off from land is not an important source of plutonium contamination of the sea.

5. Discussion

5.1. Measured versus expected ocean inventories

In the calculations of the oceanic Pu-inventories from the Geosecs data, it has been assumed that the area deposit in the marginal seas has been the same as for the nearest Geosecs stations in the open ocean. This may not be quite true. In the Mediterranean Sea the deposit thus seems a little less (see 4.3.5) than in the North Atlantic open ocean, and this is also the case in the Baltic Sea where the deposit is only 2/3 of that expected from fallout.

However the area of the marginal seas is only 9% of the total area of the Atlantic-Arctic Oceans. In case of the Pacific-Indian Oceans the marginal seas constitute 6% of the total area. It is thus of no significance if the deposit in marginal seas shows a little less Pu deposit than the open ocean.

In both the Pacific and Atlantic Oceans the measured inventories of Pu are higher than those expected from fallout delivery. In the Atlantic the discrepancy is less than in the Pacific and may be insignificant. But as a whole the oceans of the world seem to contain more plutonium than expected.

There are various possible explanations for this phenomenon:

The deposition of plutonium from atmospheric weapons testing has been underestimated, because the inventories based upon soil measurement underrate the depositions occurring over the seas. It has been shown that seaspray enriches Pu from surface seawater (Cambray, 1982b). If we suppose that seaspray over the oceans has a high efficiency for abstracting Pu from the air, the deposition of Pu over the oceans may increase. Another possibility is enhanced wet deposition of Pu over the oceans, due to higher rainfall than over land.

Another explanation could be that the available data on Pu in oceanic water columns are not representative for the total area of the oceans. Although large areas are unsampled it would be a little surprising that the locations chosen in all latitude bands in the Pacific, should be biased in the same manner. We could also imagine that the analyses of Pu in the deeper water masses were biased on the high side due to contamination problems during either the sampling or the analysis. Again this seems less likely, as the sampling and analysis have all been performed by highly experienced laboratories.

The most likely explanation thus seems to be an underestimate of the Pu deposition over ocean areas compared to land and thus an underestimate of the actual global Pu fallout from weapons testing. In order to confirm this conclusion it will be necessary to collect more water columns in the Pacific as well as in the Atlantic Ocean. The data from the Southern Hemisphere are especially scarce, but as most global fallout has been deposited in the Northern Hemisphere it is also necessary to increase the number of sampling stations here.

Most deep sea water samples have been too small for reliable ^{238}Pu determinations. It would be useful, however, if this isotope was determined as well, as this would make it possible to distinguish among various sources. Another possibility which has been used is to analyse for ^{240}Pu and ^{239}Pu separately by mass spectroscopy (Buessler, 1986). This technique has, e.g. demonstrated that local fallout from the U.S. Nevada test site has been found in the Atlantic Ocean and that this debris seems less soluble than global fallout; thus it goes to the sediments to a greater extent (cf. Appendix Fig. A.8).

5.2. Distribution between water and sediments

In the coastal waters nearly all the Pu is found in the sediments and only a few percent is seen in the water column. In the open ocean, the situation is reversed; here more than 90% is in the water column (see Appendix Fig. A.8). We would like to know how this pattern will change in the long run.

By using sediment traps Livingston (1983) has determined the average deep sea ^{239,240}Pu fluxes in the equatorial north-west Atlantic (13°05'N, 54°W) to 0.042 Bq m^{-2} yr^{-1} and in the equatorial North Pacific (15°21'N, 151°W) to 0.167 Bq m^{-2} yr^{-1}. From this and from the inventories in the water columns, 17.7 Bq m^{-2} and 77.8 Bq m^{-2}, respectively, Livingston obtained the apparent residence times (inventory/flux) of 417 yr and 462 yr, respectively. These residence times would mean that in a few thousand years essentially all Pu in the oceans would be transferred to the sediments. When Livingson however compared inventories of sediment cores with those expected from the fluxes he found that the observed inventories were 2-3 times less than those expected. He concluded that most of the particulate Pu flux dissolved at the sediment-water interface. In other words remobilization of the Pu delivered to the deep ocean seems to occur. Similar conclusions have been drawn by Noshkin (1986) who in Enewetak and Bikini lagoons observed remobilization of Pu from sediments to lagoon water. Some of the Pu remobilized may again (at another place) be resorbed by sediments. But the sorption-desorption process is not completely reversible because of the changes that occur in the relative amounts of the mixed oxidation states in solution with time.

Thus it is difficult for the time being to predict when a steady state situation for Pu in the ocean will occur and how the Pu-distribution between water and sediments then will be. Such a prediction is however necessary for the Pu dose calculations in the marine environment. It is evident that if all Pu sooner or later goes to the deep sea sediments the doses will be lower than if the Pu stays in the water column. It is also evident that if Pu remains in the water, the distribution in the column will be of great importance. The doses will be much lower if the Pu resides in the water close to the bottom than if it is in the surface layers where most of the fish are caught.

The most likely scenario for the next few thousand years probably is that the Pu will go to the sediments, and the water just over the sediments will contain enhanced Pu concentrations, whereas the remaining part of the water column will be essentially free of Pu.

6. Conclusions and recommendations

According to measurements (GEOSECS) the world's ocean contain approximately 16 PBq $^{239,240}Pu$, of which one-fourth is in the Atlantic and three-fourths in the Pacific Ocean. The expected inventory (from nuclear weapons testing) in the world's oceans is 12 PBq $^{239,240}Pu$ including local fallout at the test sites. In the Irish Sea a local contamination of 0.3 PBq $^{239,240}Pu$ from the Sellafield reprocessing plant resides in the sediments. No other sources than fallout and reprocessing add significantly to the $^{239,240}Pu$ inventories in the oceans. The discrepancy between measurements and expectations are assumed to be due to an underestimate of the rainfall and dry fallout (seaspray) and thus of the Pu-deposition over the oceans, but may also to some degree be due to inadequate sampling.

The data of ^{238}Pu are very scarce and it is not possible to conclude whether the measurements of this isotope in the oceans differ from expectations.

As it is important from a "global pollution point of view" to know the deposition of pollutants over the oceans, it is recommended that further sampling of deep-water samples in the oceans for radiochemical analysis should be pursued. Furthermore samples already collected should be analysed for plutonium as soon as possible.

Appendix: Data used in the report

Table A.1. Plutonium 239,240 inventories in Pacific water columns collected during the GEOSECS programme (Livingston, 1985a)

Cruise station	Position (cf. Fig. A.1)		Collection year	Bottom depth m	Inventory Bq m^{-2}	Inventory depth m
GX-201	34°10.5N	127°53.8W	1973	4800	57	4800
GX-202	33° 6.0N	139°34.4W	"	5129	74	5129
GX-204	31°22.8N	150° 2.1W	"	5403	77	5403
GX-212	30° 0.0N	159°50.5W	"	5877	114	5877
GX-214	32° 1.5N	176°59.9W	"	5706	143	5706
GX-217	44°36.8N	176°50.2W	"	5983	(44)	1000
GX-218	50°26.8N	176°35.0W	"	7301	121	7301
GX-222	40°10.0N	160°30.0W	"	5647	98	5647
GX-223	34°58.4N	151°50.6E	"	6140	129	6140
GX-224	34°15.6N	141°58.0E	"	9198	136	5500
GX-225	32°37.0N	161°55.0E	"	5958	150	5958
GX-226	30°34.0N	170°36.5E	"	5603	121	5603
GX-227	25° 0.0N	170° 5.0E	"	6061	126	6061
GX-229	12°53.0N	173°28.0E	"	5729	109	5729
GX-231	14° 7.0N	178°34.0W	"	5707	99	5707
GX-235	16°45.4N	161°23.7W	"	5564	77	5300
GX-239	5°53.0N	172° 0.9W	"	5868	83	5868
GX-241	4°33.8N	179° 0.2E	"	5727	(17)	600
GX-246	0°00S	178°59.0E	"	5422	(25)	1000
GX-251	4°34.0S	178°57.0E	"	5519	(10)	900
GX-263	16°41.9S	167° 3.7W	1974	5716	24	5716
GX-331	4°36.6S	125° 8.5W	"	4490	21	4490

Table A.2. Plutonium 239,240 inventories in Pacific water columns collected during various cruises (cf. Figs. A.2 and A.3)

Position	Sampling year	Inventory Bq m^{-2}	Reference
12°00'N 152°29'E	1982	134	Nakanishi, 1984
30°N 158°W	1979	81	Bowen, 1982

Table A.3. Anova estimated Bq ^{239,240}Pu m^{-2} in the Pacific Ocean (cf. the main text)

	Longitude									
	East				West					
Latitude	140° -150°	150° -160°	160° -170°	170° -180°	180° -170°	170° -160°	160° -150°	150° -140°	140° -130°	130° -120°
> 40°N	129	133	142	122	109	77	88	80	80	58
30-40°N	136	140	150	129	115	82	93	74	74	61
0-30°N	119	123	131	113	101	71	82	65	65	54
0-30°S	43	45	48	41	37	26	30	23	23	19

Table A.4. Plutonium 239,240 inventories in Atlantic water columns collected during the GEOSECS programme (Livingston, 1985a)

Cruise station	Position (cf. Fig. A.5)	Collection year	Bottom depth m	Inventory Bq m^{-2}	Inventory depth m
GX-3	51° 1.0N 43° 1.0W	1972	4321	103	4321
GX-11	63°30.0N 35°14.0W	"	2392	60	2392
GX-14	65°55.7N 27°27.0W	"	646	20	646
GX-17	74°56.2N 1° 7.2W	"	3740	54	3740
GX-19	64°10.0N 5°36.5W	"	3390	33	1000
GX-27	41°59.0N 42° 1.0W	"	4875	113	4875
GX-29	35°58.0N 47° 1.1W	"	4974	82	4974
GX-31	27° 0.0N 53°31.0W	"	6009	70	6009
GX-33	21° 0.0N 54° 2.0W	"	5195	43	5195
GX-37	12° 1.5N 51° 0.0W	"	5064	13	1000
GX-40	3° 30N 38° 0.0W	"	4273	7	1000
GX-49	7°12.6S 28° 0.0W	"	5581	10	500
GX-54	15° 0.0S 29°32.0W	"	5175	6	1000
GX-60	32°58.0S 42°31.0W	"	4401	18	4401
GX-67	44°58.0S 51°10.0W	"	5846	8	1200
GX-111	2° 0.5N 14° 1.6W	"	5163	16	5163
GX-115	28° 6.0N 25°50.0W	"	5285	52	5285

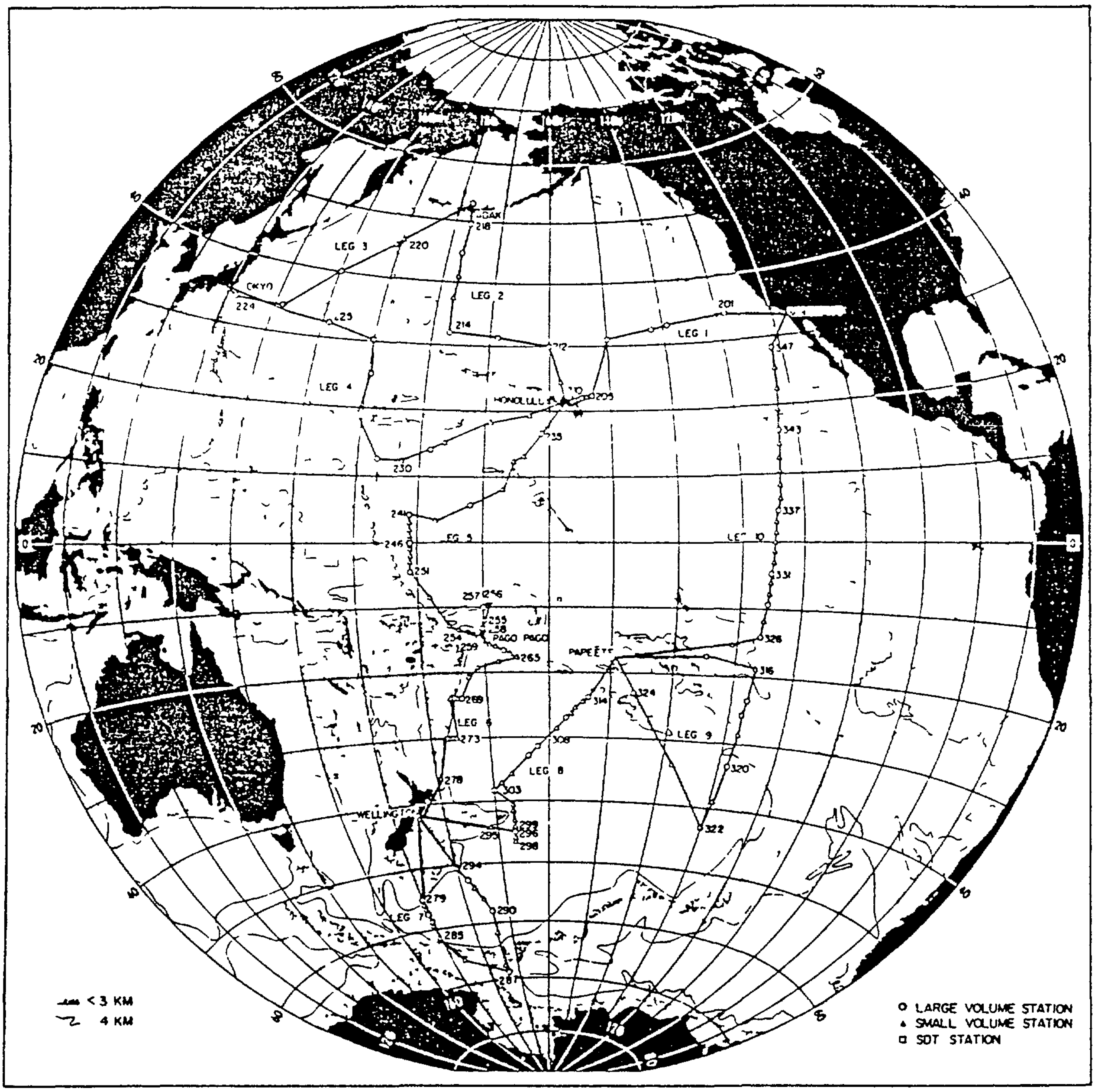


Fig. A.1. Geosecs Pacific Expedition 1973-74 (Livingston, 1985a).

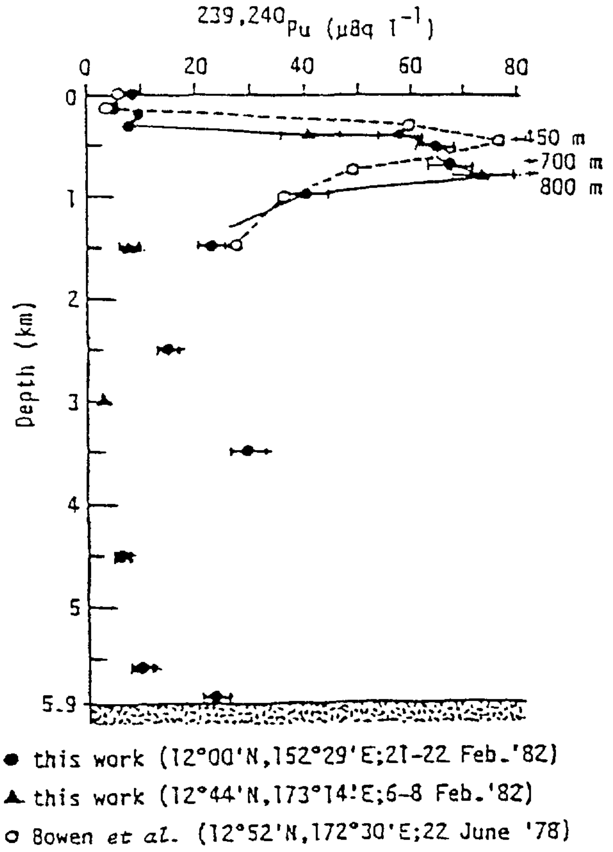


Fig. A.2. Pacific water columns (Nakanishi, 1984).

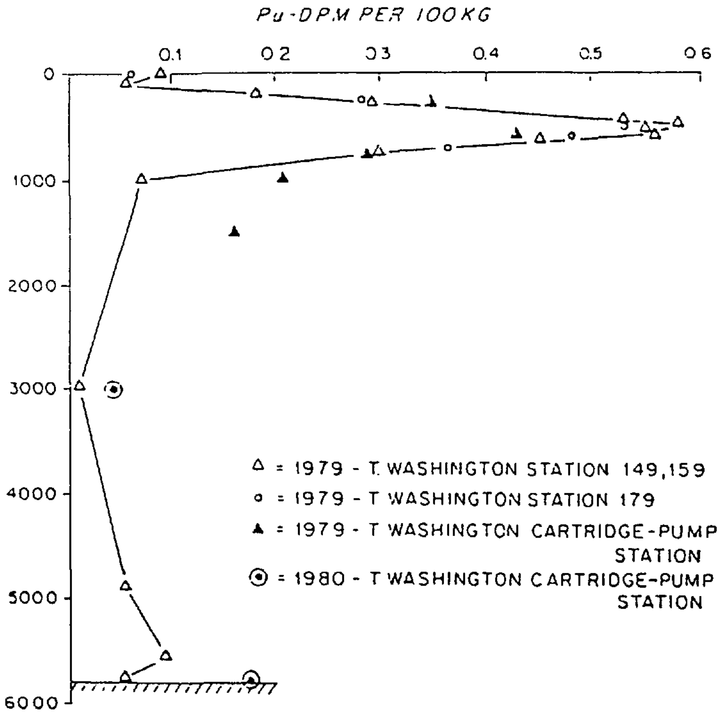


Fig. A.3. N-Pacific water profiles (Bowen, 1982).

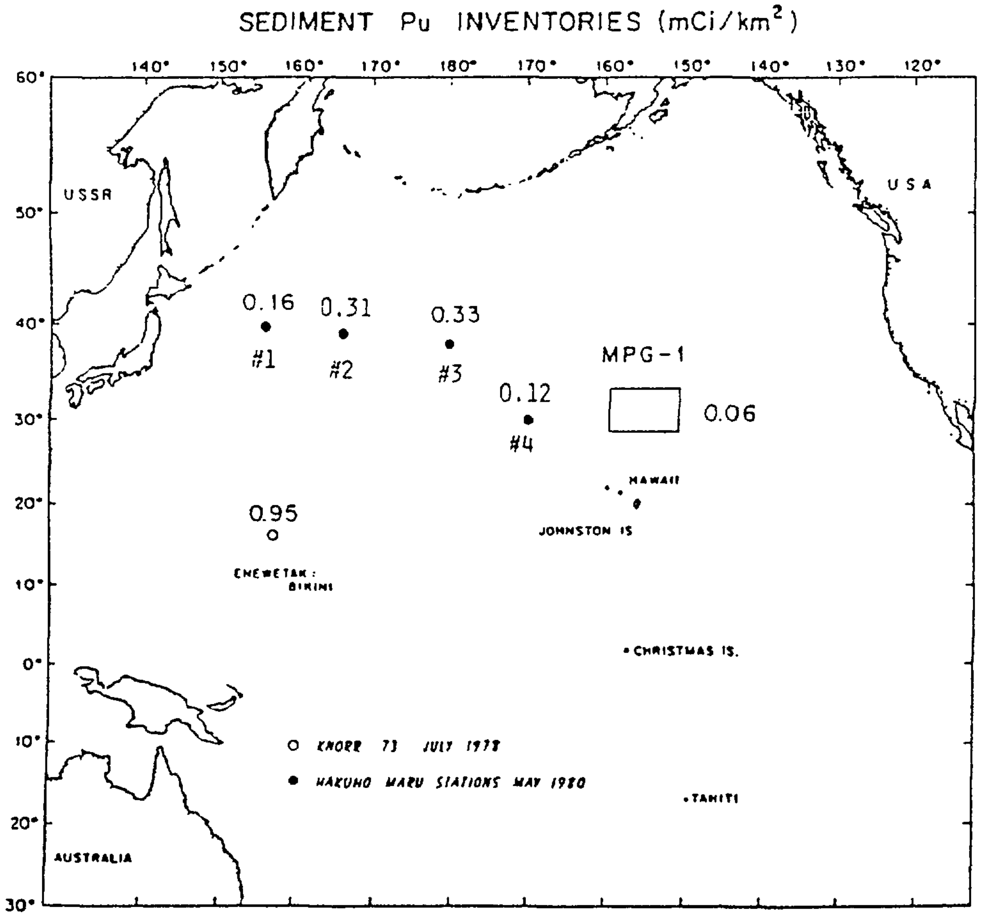


Fig. A.4. N-Pacific sediment inventories (Livingston, 1986).

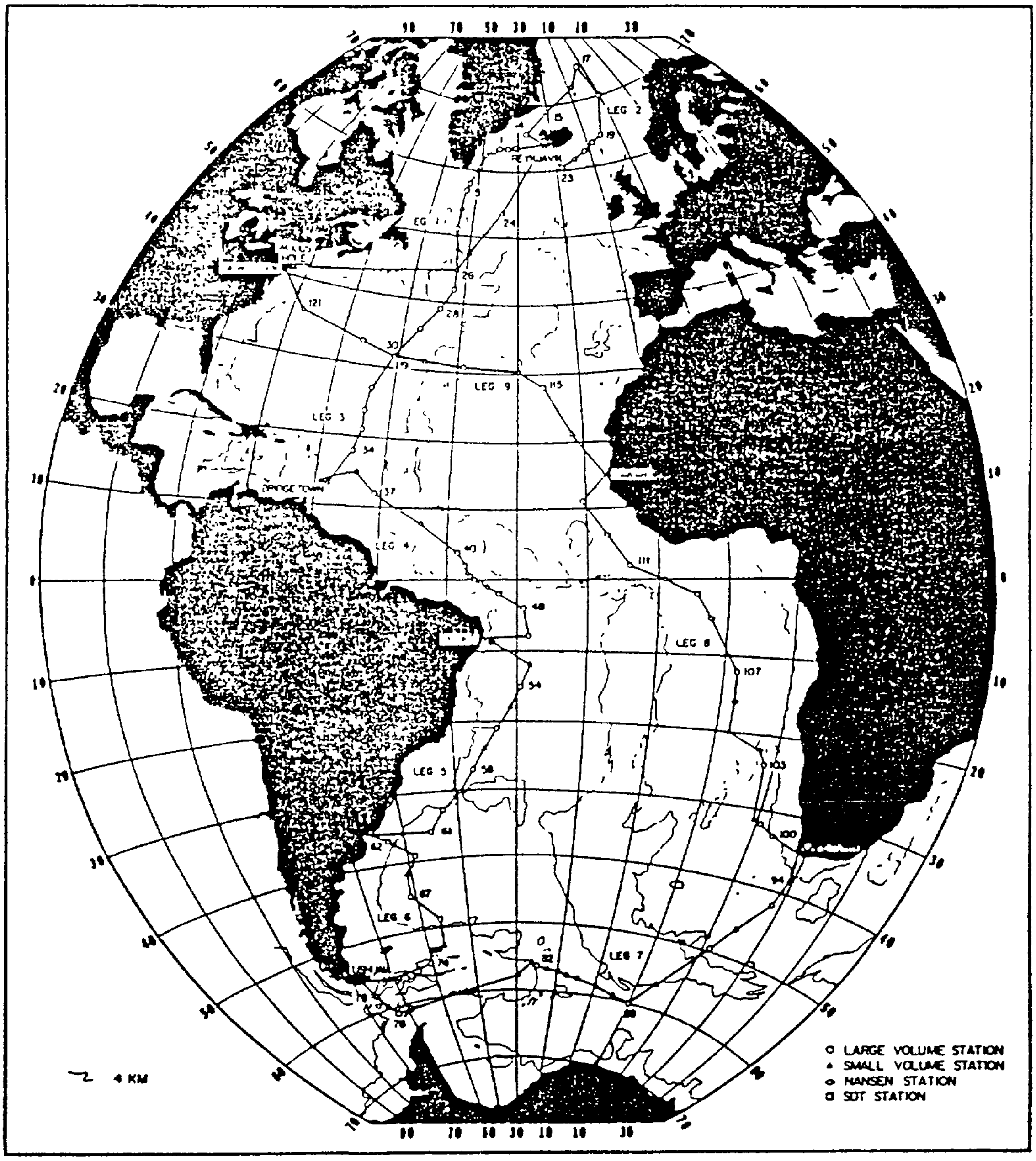


Fig. A.5. Geosecs Atlantic Expedition 1972-73 (Livingston, 1985a).

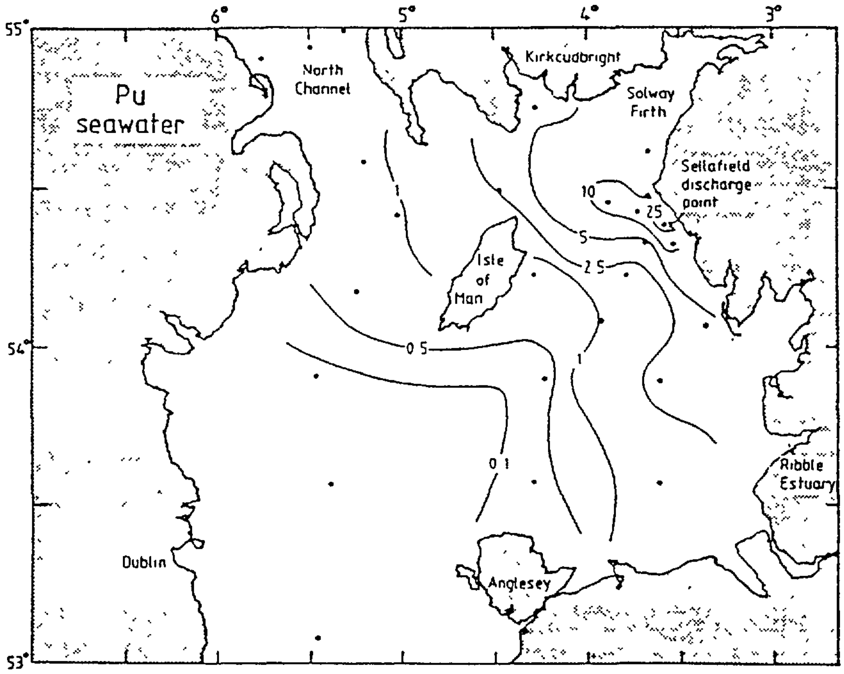


Fig. A6. Pu-concentrations in filtrate, surface waters of the Irish Sea in April 1979 (Pentreath, 1986)

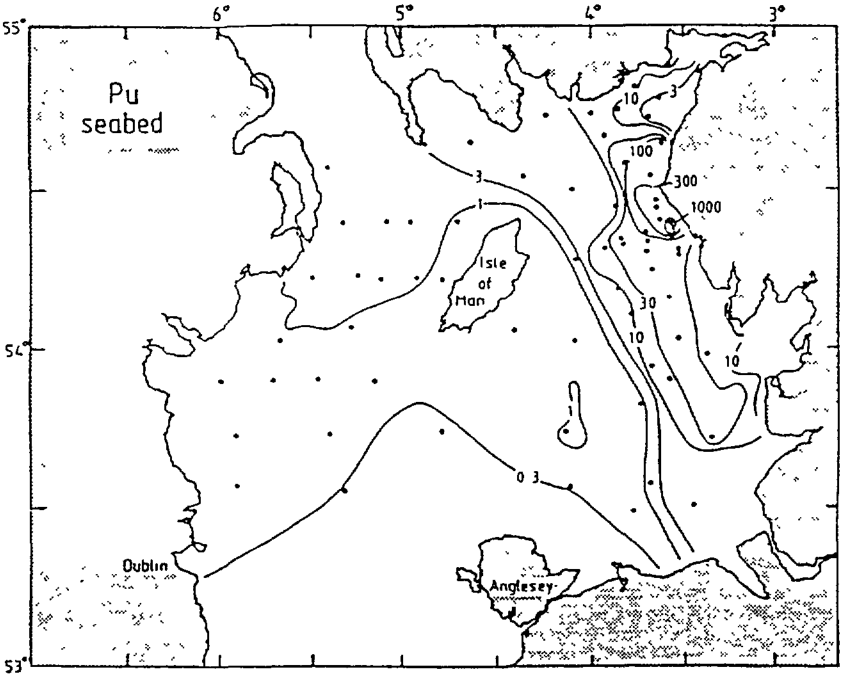
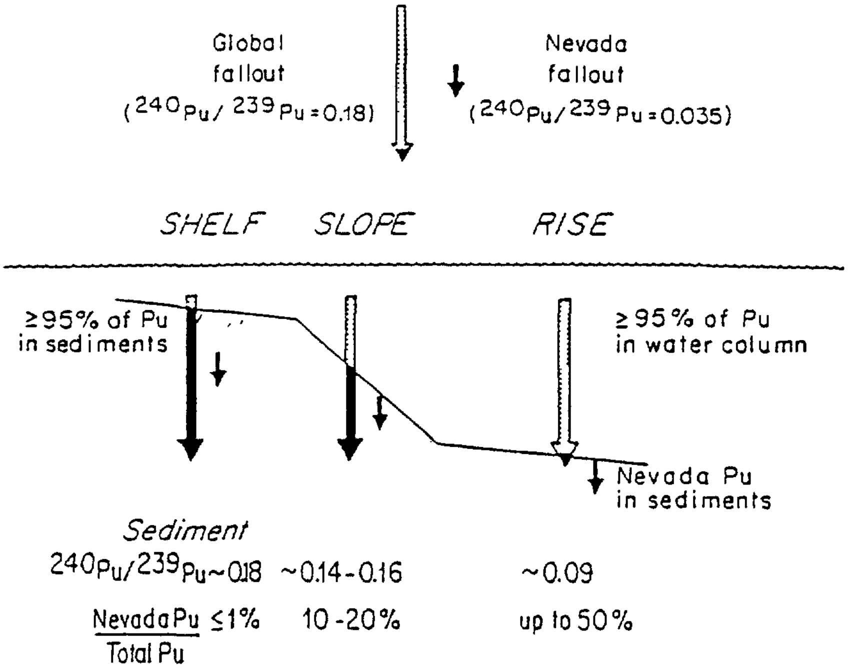


Fig. A.7. Estimated sediment inventories of Pu in the Irish Sea 1977/1978 (Pentreath, 1986).



Nevada fallout ($^{240}Pu/^{239}Pu = 0.035$) which is deposited to the N. Atlantic is rapidly removed to the sediments at all depths. Global stratospheric fallout ($^{240}Pu/^{239}Pu = 0.18$) is delivered in much larger quantities to the N. Atlantic; however, only the shallow shelf and slope sites have sufficient removal rates to deposit the bulk of this Pu to the sediments, while at the deep ocean sites, the bulk of stratospheric Pu remains in the watercolumn. The relative inventory of stratospheric vs. Nevada fallout in sediments therefore changes systematically with depth resulting in the observed decrese in the $^{240}Pu/^{239}Pu$ ratio with increasing depth (as shown below the figure).

Fig. A.8. NW Atlantic fallout Pu model (Buessler, 1986).

REFERENCES

The list of references comprise those referred to in the text. Furthermore other relevant publications containing information on Pu mostly in surface water and sediments from coastal regions are included.

In order to avoid duplications a selection of the references has been made, e.g. if authors has been dealing with the same problems for a period, only the most comprehensive publication(s) will be mentioned. Furthermore publications in refereed scientific journals will be preferred to proceedings and reports.

Aarkrog A., Dahlgaard H., Nilsson K. and Holm E. (1984). Studies of Plutonium and Americium at Thule, Greenland. Health Physics 46, 29-44.

Aarkrog A., Dahlgaard H., Hansen H., Holm E., Hallstadius L., Rioseco J. and Christensen G. (1985). Radioactive Tracer Studies in the Surface Waters of the Northern North Atlantic Including the Greenland, Norwegian and Barents Seas. Rit Fiskideildar 9, 37-42.

Beasley T.M., Ball L.A. and Blakeslay B.A. (1983a). Plutonium and Americium Export to the North-East Pacific Ocean by Columbia River Run off. Estuarine, Coastal Shelf Sci. 13, 659-669.

Beasley T.M., Carpenter R. and Jennings C.D. (1983b). Plutonium ^{241}Am and ^{137}Cs Ratios, Inventories and Vertical Profiles in Washington and Oregon Continental Shelf Sediments. Geochim. Cosmochim. Acta 46, 1931-1946.

Beasley T.M. and Jennings C.D. (1984). Inventories of ^{239,240}Pu, ^{241}Am, ^{137}Cs and ^{60}Co in Columbia River Sediments from Hanford to the Columbia River Estuary. Environ. Sci. Technol. 18, 207-212.

Bowen V.T. (1979). Element Specific Redistribution in the Marine Water Column. (Abstract from a workshop March 7-9, 1979, Gaithesburg, Maryland; USDOE, CONF 790382.

Bowen V.T., Noshkin V.E., Livingston H.D. and Volchok H.L. (1980a). Fallout Radionuclides in the Pacific Ocean: Vertical and Horizontal Distributions, largely from Geosecs Stations. Earth and Planetary Science Letters 49, 411-434.

Bowen V.T. and Livingston H.D. (1980b). Radionuclide Distributions in Sediment Cores Retrieved from Marine Radioactive Waste Dumpsites in: Impacts of Radionuclide Releases into the Marine Environment. Symp. Vienna 6-10 Oct. 1980, p. 33-63.

Bowen V.T. (1982). Transuranic Behaviour in Marine Environment IAEA-TECDOC-265, 129-140.

British Nuclear Fuels plc (1985). Annual Report on Radioactive Discharges and Monitoring of the Environment 1984 (and earlier years reports); Risley, Warrington, Cheshire, UK.

Buesseler K.O., Livingston H.D. and Sholkovitz E.R. (1985). ^{239,240}Pu and excess ^{210}Pb inventories along the shelf and slope of the norteast U.S.A. Earth and Planetary Science Letters 76, 10-22.

Buesseler K.O. (1986). Plutonium Isotopes in the North Atlantic. Ph.D. thesis. Woods Hole Oceanographic Institution and Massachusetts Institute of Technology. 219 pp.

Calmet D. and Gueguenia t P. (1985). Les Rejets d'Effluents liquides radioactifs du Centre de Traitement des Combustibles irradies da la Hague (France) et l'Evolution radiologique du Domaine marin. Annex 2 in Behaviour of Radionuclides released to Coastal Waters. IAEA-TECDOC-329. Vienna. 111-144.

Cambray R.S. (1982a). Annual Discharges of certain long-lived Radionuclides to the Sea and to the Atmosphere from the Sellafield Works, Cumbria 1957-1981. AERE-M 3269 Harwell 6 pp.

Cambray R.S. and Eakins J.D. (1982b) Pu, ^{241}Am and ^{137}Cs in Soil in West Cumbria and a Martitime Effect. Nature 300, 46-48.

Carpenter R. and Beasley T.M. (1981). Plutonium and Americium in Anoxic Marine Sediments: Evidence against Remobilization. Geochimica et Cosmochimica Acta 45, 1917-1930.

Delfanti R., Jennings C.D. and Papucci C. (1986). Plutonium Distribution in Marine and Riverine Sediments from La Specia Area (Ligurian Sea). IAEA-TECDOC-368, 35-40.

Dobry T.J. Jr. (1980). Transuranic Elements in Space Nuclear Power Systems, in Transuranic Elements in the Environment, W.C. Hanson ed., DOE/TIC-22800, 83.

Fowler S.W., Ballestra S., La Rosa J. and Fukai R. (1983). Vertical Transport of Particulate-Associated Plutonium and Americium in the Upper Water Column of the Northeast Pacific. Deep-Sea Research 30, 1221-1233.

Fukai R., Ballestra S. and Holm E. (1976). ^{241}Americium in Mediterranean Surface Waters. Nature 264, 739-740.

Fukai R., Holm E. and Ballestra S. (1979). Note on Vertical Distribution of Plutonium and Americium in the Mediterranean Sea. Oceanol. Acta 2, 129-132.

Fukai R., Ballestra S., Thein M. and Guion J. (1980a). Input of Transuranic Elements through Rivers into the Mediterranean Sea, in: Impacts of Radionuclide Releases into the Marine Environment. Symp. Vienna 6-10 Oct. 1980 p. 3-14.

Fukai R., Ballestra S. and Thein M. (1980b). Vertical Distribution of Transuranic Nuclides in the Eastern Mediterranean Sea. p. 79-87 in Proceeding of a technical committee meeting on the behaviour of transuranics in the aquatic environment and sediment-water exchanges (IAEA and CEC). Ispra, Italy 24-28 March 1980.

Germain P. and Miramand P. (1984). Distribution and Behaviour of Transuranic Elements in the Physical and Biological Compartments of the Channel French Shore. Nucl. Instrum. Methods Phys. Res. 223, 502-509.

Glasstone S. (1962), in: The Effects of Nuclear Testing, S. Glasstone, ed. (U.S. Atomic Energy Commission, 1962) 483-485.

Hallstadius L., Aarkrog A., Dahlgaard H., Holm E., Boelskifte S., Duniec S. and Persson B. (1986). Plutonium and Americium in Arctic Waters, the North Sea and Scottish and Irish Coastal Zones. Jour. of Environmental Radioactivity 4, 11-30.

Hayes D.W., Le Roy J.H. and Cross F.A. (1975). Plutonium in Atlantic Coastal Estuaries in the South-eastern United States (Savannah River Estuary, Neuse River Estuary and Newport River Estuary). in: Proc. IAEA int. symp. on transuranium nuclides in the environment. San Fransisco, CA, USA, Nov. 1975, 79-88.

Holm E., Ballestra S. and Fukai R. (1980). Particulate Plutonium and Americium in Mediterranean Surface Waters. Oceanol. Acta 3, 157-160.

Holm E., Persson B.R.R., Hallstadius L., Aarkrog A. and Dahlgaard H. (1983). Radiocesium and Transuranium Elements in the Greenland and Barents Seas. Oceanol. Acta 6, 457-462.

Holm E., Aarkrog A., Ballestra S. and Dahlgaard H. (1986). Origin and Isotopic Ratios of Plutonium in the Barents and Greenland Seas. Earth and Planetary Science Letters 79, 27-32.

Iranco E. (1980). Personal Communication. in: The Environmental and Biological Behaviour of Plutonium and some other Transuranium Elements, Report by an NEA Group of Experts (Sept. 1981), OECD, Paris. p. 28.

Kautsky H. (1985). Distribution and Content of Different Artificial Radionuclides in the Water of the North Sea during the years 1977 to 1981 (complemented with some results from 1982 to 1984). Dt. hydrogr. Z. 38, 193-224.

Kautsky H., Wedekind C. and Eicke H.-F. (1986). Radiological Investigations in the Baltic Sea, Including the Danish Straits and the Kattegat During 1982 and 1983. IAEA-TECDOC-362, 110-132.

Kossinna E. (1921). Die Tiefen des Weltmeeres. Berlin Univ., Institut f. Meeres Kunde, Veröff., N.F., A. Geogr.-naturwiss. Reihe, Heft 9, 70 pp.

Kupferman S.L., Livingston H.D. and Bowen V.T. (1979). A Mass Balance for ^{137}Cs and ^{90}Sr in the North Atlantic Ocean. Journal of Marine Reseach 37, 157-199.

Leskinen S., Jaakola T. and Miettinen J.K. (1986). Studies on the Behaviour of Long-Lived Actinides Plutonium and Americium in the Baltic Sea; Effect of Season, Distribution Coefficients in Particulate Matter and Surface Sediment. IAEA-TECDOC-368, 61-72

Lewis E.L. (1982). The Arctic Ocean: Water Masses and Energy Exchanges in: The Arctic Ocean Rey L. ed. Comité Arctique, Monaco p. 43-68.

Linsalata P., Wrenn M.E., Cohen N. and Singh N.P. (1980). Pu-239+240 and Pu-238 in sediments of Hudson River estuary. Environ. Sci. Technol., 14, 1519-1523.

Livingston H.D., Bowen V.T. and Burke J.C. (1976). Fallout Radionuclides in Mediterranean Sediments 16 pp. 25. congress and plenary assembly of CIESM Split, Yugoslavia 22 Oct. 1976.

Livingson H.D. and Bowen V.T. (1977). Windscale Effluent in the Waters and Sediments of the Minch. Nature 269, 586-588.

Livingston H.D. and Bowen V.T. (1979) Pu and ^{137}Cs in Coastal Sediments. Earth Planet. Sci. Lett. 43, 29-45.

Livingston H.D., Bowen V.T. and Kupferman S.L. (1982). Radionuclides from Windscale Discharges II: Their Dispersion in Scottish and Norwegian Coastal Circulation. Journal of Marine Research 40, 1227-1258.

Livingston H.D. and Anderson R.F. (1983). Large Particle Transport of Plutonium and other Fallout Radionuclides to the Deep Ocean. Nature 303, 228-231.

Livingston H.D., Kupferman S.L., Bowen V.T. and Moore R.M. (1984). Verticale Profile of Artificial Radionuclide Concentrations in the Central Arctic Ocean. Geochimica et Cosmochimica Acta 48, 2195-2203.

Livingston H.D., Bowen V.T., Casso S.A., Volchok H.L., Noshkin V.E., Wong K.M. and Beasley T.M. (1985a). Fallout Nuclides in Atlantic and Pacific Water Columns: GEOSECS Data. WHOI-85-19, Woods Hole, Ma 02543. 73 pp.

Livingston H.D. (1985b). Anthropogenic Radiotracer Evolution in the Central Greenland Sea. Rit Fiskideildar, 9, 43-54.

Livingston H.D. (1986). Fallout Plutonium in Western North Pacific Sediments in: Behaviour of Long-lived Radionuclides Associated with Deep-Sea Disposal of Radioactive Wastes IAEA-TECDOC-368, Vienna. p. 27-34.

Mac Kenzie A.B. and Scott R.D. (1982). Radiocaesium and Plutonium in Intertidal Sediments from Southern Scotland. Nature 299, 613-616.

Miyake Y., Sugimura Y., Saruhashi K., Katsuragi Y. and Hirose K. (1979). Plutonium and Americium in the Environment in: Proc. of the 6th Int. Cong. ofm Rad. Res. Tokyo Japan, May 1979. 940-948.

Miyamoto T., Hishida M., Shibayama N., Shiozaki M. (1986). Marine Behaviour of Long-lived Radionuclides (fallout) at the proposed Disposal Site of Radioactive Waste in Western North Pacific in: Behaviour of Long-lived Radionuclides Associated with Deep-Sea Disposal of Radioactive Wastes. IAEA-TECDOC-368, Vienna, p. 73-80.

Murray C.N. and Kautsky H. (1977). Plutonium and Americium Activities in the North Sea and German Coastal Regions. Estuarine Coastal Mar. Sci. 5, 319-328.

Murray C.N. and Fukai R. (1978). Measurement of Pu 239+240 in the North Western Mediterranean Estuarine Coastal Mar. Sci. 6, 145-151.

Murray C.N., Kautsky H. and Eicke H.F. (1979). Transfer of Actinides from the English Channel into the Southern North Sea. Nature 278, 617-620.

Nagaya Y. and Nakamura (1984). ^{239,240}Pu, ^{137}Cs and ^{90}Sr in the Central North Pacific. Journal of the Oceanographical Society of Japan 40, 416-424.

Nakanishi T., Yajima M., Senaga M., Takei, M., Ishikawa A., Sakamoto K. and Sakanoue M. (1984). Determination of ^{239,240}Pu in Seawater. Nucl. Inst. and Methods in Phys. Res. 223, 239-242.

National Academy of Sciences (1971). Radioactivity in the Marine Environment. 272 pp. Washington D.C. 20418.

Nelson D.M., Carey A.E. and Bowen V.T. (1984). Plutonium Oxidation State Distributions in the Pacific Ocean during 1980-1981. Earth and Planetary Science Letters 68, 422-430.

Noshkin V.E., Wong K.M. and Eagle R.J. (1983). Radionuclides in the Sediments from the North East Atlantic Disposal Site UCID-19924. Lawrence Livermore National Lab. CA, USA 16 pp.

Noshkin V.E., Wong K.M. and Eagle R.J. and Jokela T.A. (1986). Biogeochemical Studies of Long-lived Radionuclides in Marine Environments. IAEA-TECDOC-368, 119-127.

Olsen C.R., Simpson H.J. and Trier R.M. (1981). Plutonium, Radiocesium and Radiocobalt in Sediments of the Hudson River Estuary. Earth and Planetary Science Letters 55, 377-392.

Pentreath R.J. (1985). Radioactive Discharges from Sellafield (UK). Annex 1 in: Behaviour of Radionuclides released into Coastal Waters. IAEA_TECDOC-329 p. 67-110.

Pentreath R.J., Kershaw P.J., Harvey B.R. and Lovett M.B. (1986). The Behaviour of certain Long-lived Radionuclides in the Marine Environment. IAEA-TECDOC-368, 101-114.

Pillai K.C. and Mathew E. (1975). Plutonium in the Aquatic Environment. Its Behaviour, Distribution and Significance. in: Proc. IAEA int. symp. on transuranium nuclides in the environment. San Francisco, CA, USA, Nov. 1975. 25-44.

Pillai K.C., Mathew E., Matkar W.M., Dey N.N., Abani M.C., Chhapgar B.F. and Mullay C.D. (1982). Behaviour of Transuranic Nuclides in Coastal Environment. IAEA-TECDOC-265, 61-82.

Salo A. (1984). Strontium-90, Cesium-137, Plutonium 239 and Plutonium 240 Inventories in the Baltic Sea. 10 pp. Proc. 7. Nordic Society for Radiation Protection Ordinary Meeting Copenhagen, Denmark, 10-12 Oct. 1984.

Salo A., Tuomainen K. and Voipio A. (1986). Inventories of Certain Long-lived Radionuclides in the Baltic Sea. IAEA-TECDOC-362, 53-62.

Scott M.R., Salter P.F. and Halverson J.E. (1983). Transport and Deposition of Plutonium in the Ocean: Evidence from Gulf of Mexico Sediments. Earth and Planetary Science Letters 63, 202-222.

Scott M.R., Rotter R.J. and Salter P.F. (1985). Transport of Fallout Plutonium to the Ocean by the Mississippi River Earth and Planetary Science Letters 75, 321-326.

Triulzi C., Tassi Pelati L., Albertazzis., Delle Site A., and Marchionni V. (1982). Presence and Distribution of Plutonium Isotopes in two typical Marine Systems Taranto and Venice Gulfs. IAEA-TECDOC-265, 83-92.

UNSCEAR (1982). Ionizing Radiation: Sources and Biological Effects 773 pp. United Nations, New York.

Vestergaard J. (1964). Analysis of Variance with Unequal Numbers in Groups. Gier system library No 211. A/S Regnecentral, Copenhagen.

Wrenn M.E. (1974). Environmental Levels of Plutonium and the Transplutonium Elements, WASH-1359, 89.

Yamamoto M., Igarashi S., Yoshioka M., Kitagawa T. and Sakanoue M. (1984). Transuranic Elements in Nyu Bay sediments. Health Phys. 46, 455-460.

Yamato A., Kinoshita M. and Kurokawa Y. (1977). Plutonium Contents in the Coastal Environment of Japan. in: Proc. 4 Int. Congr. IRPA, Paris April 1977. 865-868.

Yamato A. (1984). Plutonium and Radiocerium Contents in Shallow Sea Sediment Collected at Off Shore Ibaraki Prefecture, Japan. Radioisotopes 33, 60-64.

LEAD-210 AND POLONIUM-210 IN THE WORLD'S OCEANS

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Abstract

After a brief summary of the sources of ^{210}Po and ^{210}Pb in the ocean, the published data for these two nuclides are discussed in four sections. 1. Surface sea water ($\leq$ 100 m) from the open ocean. 2. Surface sea water from coastal regions. 3. Depth profiles in the open ocean. 4. Data from atypical regions. Estimates of the global inventories of ^{210}Po and ^{210}Pb are made and compared with the ^{226}Ra inventories (Bojanowski, this volume). Over the whole water column in the three major oceans ^{210}Po and ^{210}Pb are in approximate equilibrium. The global inventory of ^{210}Pb in the water column is estimated to be 2895 ± 517 PBq or 61 ± 11% of that of ^{226}Ra.

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1. INTRODUCTION: SOURCES OF 210-Pb AND 210-Po IN THE OCEAN

There is a continuous emission of the chemically-inert rare-gas ^{222}Rn, a member of the ^{238}U series, from the surface of the continents to the atmosphere. ^{222}Rn has a half-life of 3.8 days and its daughter products in the series are shown in Fig. 1.

$$^{222}\text{Rn} \xrightarrow[\alpha]{3.8\text{d}} {}^{218}\text{Po} \xrightarrow[\alpha]{3.1\text{m}} {}^{214}\text{Pb} \xrightarrow[\beta]{27\text{m}} {}^{214}\text{Bi} \xrightarrow[\beta]{20\text{m}} {}^{214}\text{Po} \xrightarrow[\alpha]{1.6(10^{-4})\text{s}}$$

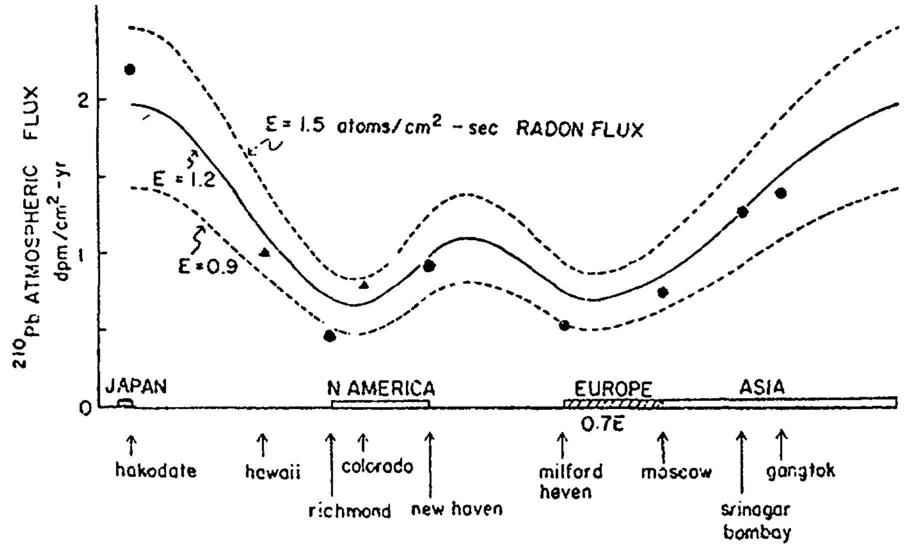
$$^{210}\text{Pb} \xrightarrow[\beta]{22\text{y}} {}^{210}\text{Bi} \xrightarrow[\beta]{5\text{d}} {}^{210}\text{Po} \xrightarrow[\alpha]{138\text{d}} {}^{206}\text{Pb (stable)}$$

Fig. 1: Decay products of ^{222}Rn with their half-lives.

The daughter products are chemically reactive and become associated rapidly with atmospheric particulates; these in turn fall to earth either as dry or wet (rain, snow) fall-out. As a result of atmospheric circulation, the fall-out occurs over the oceans as well as the continents. The residence time of the aerosols in the atmosphere can be estimated from measurements of the disequilibria between ^{222}Rn and/or those of its radioactive daughters with sufficiently long half-lives, viz. ^{210}Pb, ^{210}Bi and ^{210}Po. A recent synthesis of this problem by Lambert et al. (1982) estimates a residence time of 6.5 days for submicronic aerosols in the troposhpere. This residence time is too short for equilibrium between ^{210}Po and ^{210}Pb to be attained on the aerosols; the measured ^{210}Po/^{210}Pb activity ratio in fall-out, both wet and dry, is about 0.1 (Burton and Stewart, 1960; Lambert and Nezami, 1965).

The fall-out flux of ^{210}Pb to the earth's surface is not uniform, and the review of Turekian et al. (1977) discusses the variations which occur. There is a major latitudinal variation, the maximum fluxes being found in the medium latitudes in the Northern Hemisphere which contain the major continental land masses. Variations with longitude are also evident. The highest fluxes, about 64 atoms /cm^2/min (Fukuda and Tsunogai, 1975) are found at the eastern edge of the Asiatic land mass. In western Europe, by contrast, the flux is between 16 and 22 atoms /cm^2/min (Lambert and Nezami, 1965; Peirson et al. (1966). Turekian et al. (1977) have modelled the flux variation in terms of the land mass distribution, the air-mass circulation and the aerosol residence time. Their results are given in Figure 2, which shows the ^{210}Pb atmospheric fall-out flux in dpm/cm^2/year (1 dpm/cm^2/year = 32.1 atoms/cm^2/min) as a function of longitude in mid-latitudes (15-55°) in both the Northern and Southern Hemispheres. The emission of ^{222}Rn from the surface of the oceans is assumed to be negligible.

Northern Hemisphere (15°-55°N)



Southern Hemisphere (15°-55°S)

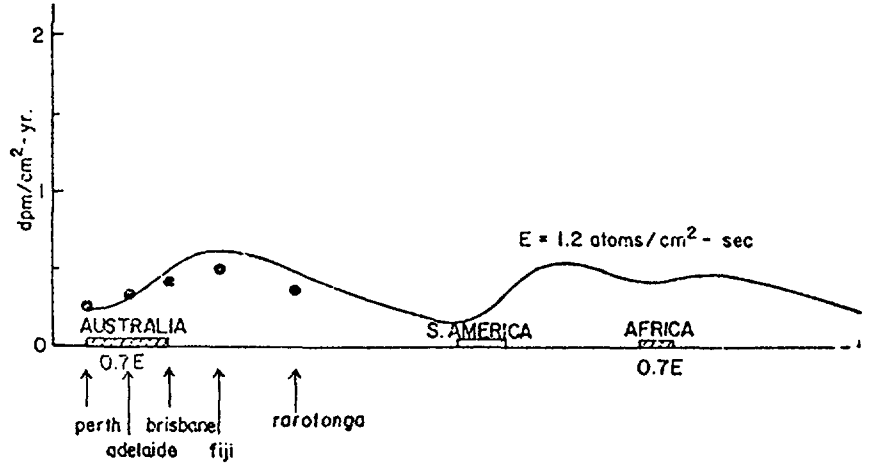


Fig. 2. The longitudinal variation of ^{210}Pb flux in mid-Northern Hemisphere (15°-55°N) and mid-Southern Hemisphere (15°-55°S). From Turekian et al. (1977)

Other possible sources of ^{210}Pb in the atmosphere are of relatively minor importance. They have been reviewed in detail by Robbins (1978); for the record, they include dust containing ^{226}Ra, the intensive use of phosphate-based fertilizers, the combustion of coal and petrol and - possibly but by no means certainly - nuclear weapons tests. In the surface layer of the ocean, the atmospheric input of ^{210}Pb which we have just discussed is the major source of this nuclide. In the water column as a whole, however, the situation is simpler: the dominant source of ^{210}Pb is in situ production resulting from the radioactive decay of its grandparent, ^{226}Ra via the short-lived intermediate ^{222}Rn. The review of Bojanowski (this volume) provides the data for this dominant source term. Input of ^{210}Pb from rivers into the ocean is probably insignificant (Robbins, 1978). Insofar as ^{210}Po is concerned, even in the surface layer the atmospheric input is negligible, and the source of ^{210}Po is in situ production from the radioactive decay of ^{210}Pb via the short-lived intermediate ^{210}Bi.

2. DATA FOR 210-Po AND 210-Pb IN THE OCEAN

The values in the literature for ^{210}Po and ^{210}Pb in sea water will be discussed in four sections:

1. Surface sea water ($\leq$ 100 m) from the open ocean
2. Surface sea water from coastal regions
3. Depth profiles in the open ocean
4. Data from atypical regions

2.1 Surface sea water concentrations in the open ocean

These are reported in 24 references in the Appendix and are summarized in Table I. Since the atmospheric input of ^{210}Pb varies with latitude, the data have been grouped latitudinally.

Table I. Concentrations of ^{210}Po and ^{210}Pb in surface sea water from the open ocean (Bq m^{-3})

	^{210}Po			^{210}Pb			$^{210}Po/^{210}Pb$		
Latitude	n	Range	Median	n	Range	Median	n	Range	Median
55-90° N	3	0.53-1.82	1.72	3	0.50-2.07	1.82	3	0.88-1.06	0.94
15-55° N	44	0.67-2.53	1.48	66	0.97-4.47	2.45	44	0.21-1.10	0.57
0-15° N	18	0.08-3.63	1.17	25	0.90-3.52	1.95	18	0.30-1.40	0.55
0-15° S	14	0.47-1.90	0.88	24	0.83-2.58	1.75	14	0.35-0.88	0.55
15-55° S	60	0.30-2.47	0.85	83	0.37-5.00	1.30	60	0.11-1.50	0.62
55-90° S	7	0.77-1.20	1.03	36	1.05-2.33	1.45	7	0.52-0.82	0.65

The 15-55° band was chosen because the most comprehensive global model available for the natural fall-out of ^{222}Rn and its daughters uses this range (Turekian et al. 1977). The fall-out of ^{210}Pb is certainly larger in the Northern Hemisphere than in the Southern, because of the larger land-masses in the North. This increase in the first component of the ^{210}Pb source term is clearly reflected in Table I in the steady decrease from north to south in the median concentrations for both ^{210}Pb and ^{210}Po (latitudes higher than 55° excluded). For the higher latitudes the data are too sparse to warrant discussion in any detail. An atypical situation in polar regions is, however, to be expected, because the natural fall-out is very low at the highest latitudes. In polar waters the atmospheric input term is thus small, and the dominant source of ^{210}Pb is in situ production from ^{226}Ra decay.

More than half the data, for both ^{210}Po and ^{210}Pb, are from the Pacific Ocean, and for this ocean it is possible to discern east-west as well as north-south variations in concentration. The maps in Nozaki et al. (1976) illustrate the general pattern, and these are reproduced in Figs. 3 and 4. The higher ^{210}Pb activities at the centres of the two hemispheric gyres, with lower values at the edges, are to be noted. A similar, but less clear, situation exists for the $^{210}Po/^{210}Pb$ activity ratios.

The disequilibrium between ^{210}Po and ^{210}Pb is important; the $^{210}Po/^{210}Pb$ activity ratio, typically about 0.6, is >1 in only 13 of the 140 samples measured. This reflects the fact that ^{210}Po is removed from surface waters more rapidly than ^{210}Pb. The removal time for ^{210}Po from

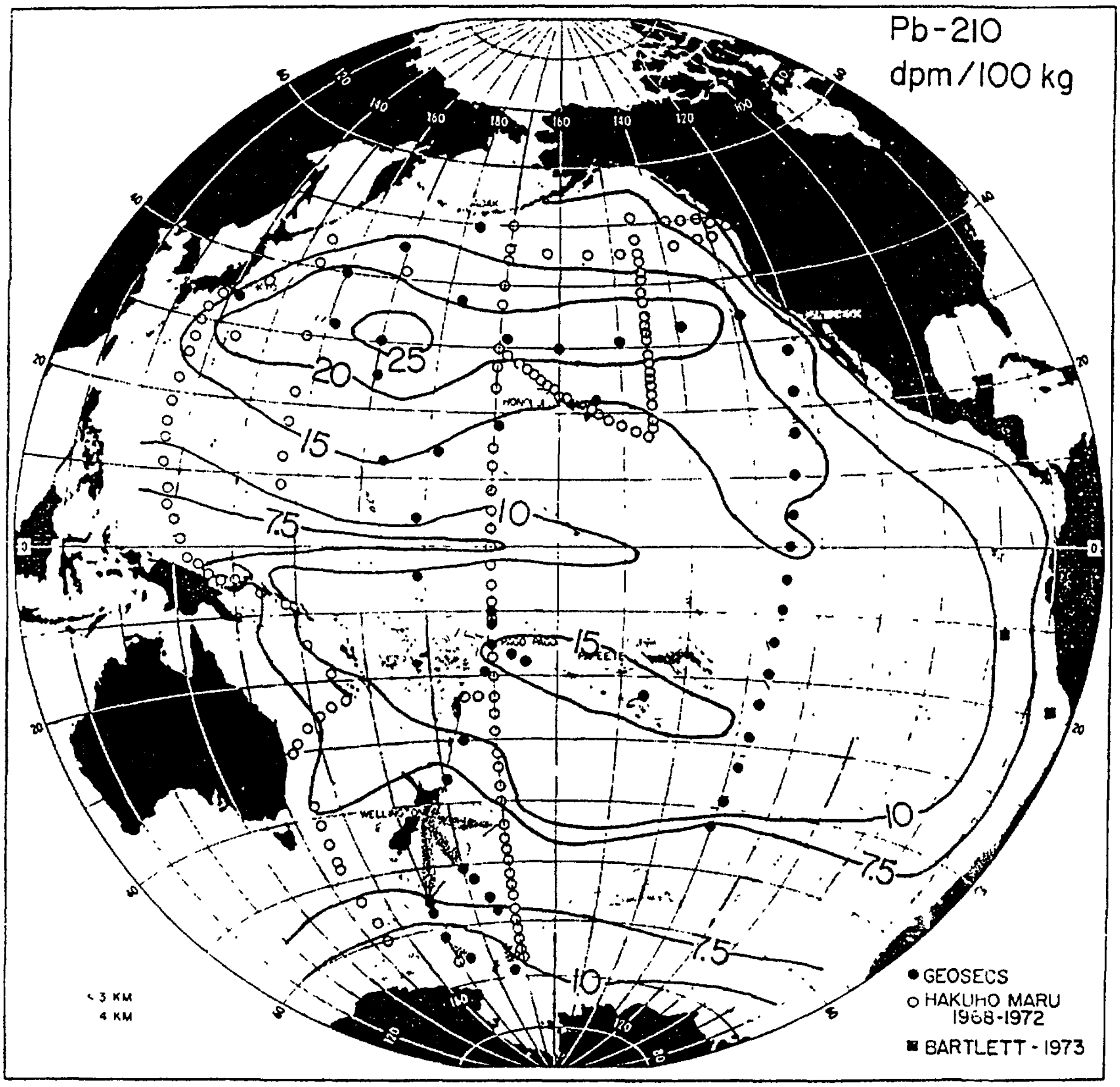


Fig. 3. ^{210}Pb distribution in surface waters of the Pacific Ocean. From Nozaki et al. (1976)

oceanic surface waters, by all processes other than radioactive decay, is typically between 0.2 and 0.7 year, whereas that for ^{210}Pb is between 0.5 and 5 years (e.g. Shannon et al., 1970; Bacon et al., 1976; Nozaki et al., 1976). The removal mechanism for both nuclides is almost certainly mediated by biological processes (e.g. sinking faecal pellets).

Those references which give particulate and dissolved concentrations usually do so via filtration through an 0.4 um filter. On this basis, both nuclides are predominantly in the dissolved form (> 90% for ^{210}Pb and > 80% for ^{210}Po), but with a larger percentage of ^{210}Po associated with particulates. The data of Bacon et al. (1976) for the north equatorial Atlantic illustrates this feature well: they show that in the 0-100 m surface layer, 4% of the ^{210}Pb and 14% of the ^{210}Po is "particulate".

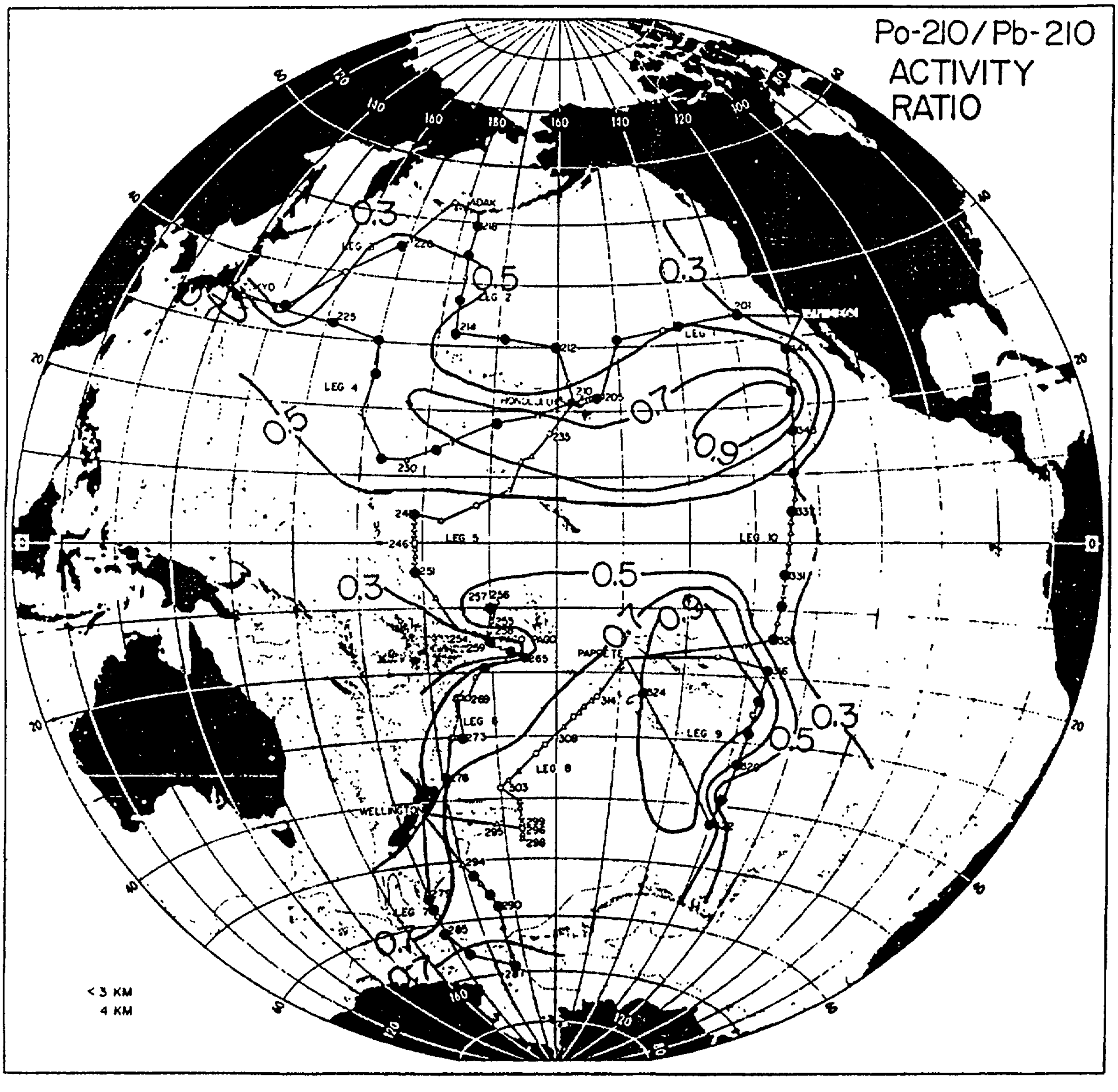


Fig. 4. The distribution of ^{210}Po/^{210}Pb activity ratios in the surface waters of the Pacific Ocean. From Nozaki et al. (1976)

2.2 Surface sea water concentrations in coastal regions

These are reported in 10 references in the Appendix. Variations observed reflect local conditions and will not be discussed in detail. The essentials can be summarized as follows:

(i) Concentrations of ^{210}Po are generally comparable with those of Table I.

(ii) Concentrations of ^{210}Pb in coastal waters are generally, but not always, lower than those in open ocean surface waters. This indicates shorter removal times for ^{210}Pb in coastal regions (e.g. Bruland et al., 1974).

(iii) The ^{210}Po/^{210}Pb activity ratios are frequently, but not always, greater than unity. It has been suggested that ^{210}Po is preferentially regenerated back to the water column as compared to ^{210}Pb in the coastal marine environment (Li et al., 1981).

(iv) Seasonal variations in both ^{210}Po and ^{210}Pb concentrations have been observed (e.g. Santschi et al., 1979; Tanaka et al., 1983).

(v) Despite the generally lower ^{210}Pb in coastal regions, it is worth noting that, of well over 100 data points for both nuclides, only one ^{210}Po and two ^{210}Pb values lie outside the Table I ranges. (In all three cases the concentrations are not far beyond the upper limits of the open ocean surface concentrations of 3.63 Bq m^{-3} for ^{210}Po and 5.0 Bq m^{-3} for ^{210}Pb).

2.3 Depth profiles in the open ocean

Depth profiles for ^{210}Po and ^{210}Pb in the open ocean have been reported in 22 of the references in the Appendix. A detailed evaluation of the data for any particular profile would require collateral discussion of the associated oceanography; here, we summarize the essentials, many of which are illustrated in Figure 5.

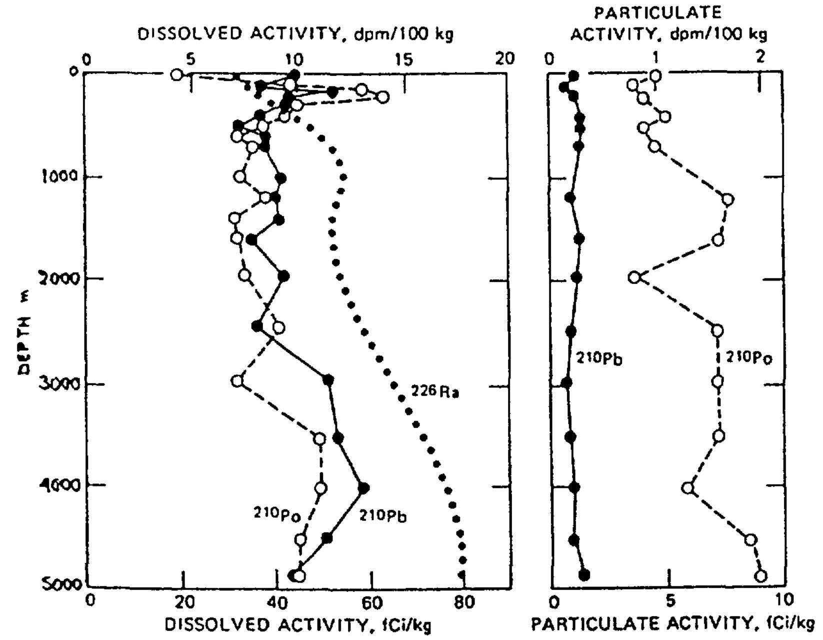


Fig. 5. Typical distributions of ^{226}Ra (●●●), ^{210}Pb (●—●) and ^{210}Po (o---o) in the tropical Atlantic Ocean. Data are from F/S METEOR, cruise 32, station 23 (Bacon, 1977). From Bacon et al. (1980a)

(i) Usually, the depth profiles show an activity excess of ^{210}Pb over ^{226}Ra in surface waters, down to about 500 m. This is the region of the water column where the atmospheric input of ^{210}Pb is significant. In some cases, the "cross-over point" between ^{210}Pb and ^{226}Ra concentrations, in activity terms, is lower (e.g. in the Labrador Sea, Bacon et al., 1980a).

(ii) Sometimes this ^{210}Pb/^{226}Ra excess in surface waters is not observed, notably in the Arctic (Moore and Smith, 1986) and Antarctic (Chung and Applequist, 1980; Chung, 1981) where the atmospheric fall-out of ^{210}Pb is small.

(iii) Below the "cross-over point" between ^{226}Ra and ^{210}Pb at about 500 m, it is the in situ decay of ^{226}Ra (via ^{222}Rn) which is the main source of ^{210}Pb. The ^{210}Pb is, however, removed from the water column preferentially with respect to ^{226}Ra by various processes (biological removal, fine particle scavenging, boundary scavenging), and the ^{210}Pb/^{226}Ra activity ratio at depth is < 1.

(iv) The ^{210}Pb depth profiles vary, but often (e.g. Bacon, 1977) show a surface or sub-surface (100-200 m) maximum with a fairly smooth decrease thereafter as the effect of the atmospheric fall-out source term falls off. Below the ^{226}Ra - ^{210}Pb "cross-over point" there is often a broad maximum at about 1000 m and a decrease in ^{210}Pb towards the bottom. The concentrations of ^{210}Pb all the way down are usually within a factor of two of those at the surface, and seldom diverge from the surface concentrations by a factor of more than three.

(v) The ^{210}Po/^{210}Pb activity ratio in open ocean surface water, is, as we have seen, almost always less than unity. Immediately below the surface the situation is complicated, with ratios > 1 sometimes being observed in the 100-300 m region. This has been attributed to recycling of ^{210}Po (Bacon et al., 1976). Below this, as a rough rule, the ^{210}Po/^{210}Pb activity ratio is about 1.0 all the way to the bottom (Bacon, 1977).

(vi) Although the ^{210}Po/^{210}Pb activity ratio, calculated from total concentrations of both nuclides, is generally about 1.0 below the surface and immediate sub-surface, there are differences between the dissolved and particulate activity ratios. Dissolved ^{210}Po/^{210}Pb averages about 0.9 whereas particulate ^{210}Po/^{210}Pb is greater than unity (e.g. Bacon, 1977).

(vii) Deep-sea removal times have been estimated in a few cases, and are about two years for ^{210}Po and from 50 to 500 years for ^{210}Pb (e.g. Thomson and Turekian, 1976).

2.4 Data from atypical regions

In eight references in the Appendix, atypical features require separate mention:

(i) The North Sea data of Spencer et al. (1980). These show unexceptional ^{210}Po concentrations but low ^{210}Pb concentrations, as in coastal regions. Almost all the ^{210}Po/^{210}Pb ratios are > 1. It is suggested that the North Sea shelf is a strong sink for ^{210}Pb, with biological removal and boundary scavenging the proposed removal mechanisms. For ^{210}Po, removed from the surface waters by biological action, it is suggested that a regeneration process from the sea floor operates.

(ii) The Cariaco Trench data of Bacon et al. (1980b). In this region the deep water, below 150 m, is apparently permanently anoxic. In the upper 300 m in this region, ^{210}Po is enriched over ^{210}Pb by some

55% over equilibrium; below that there is approximate equilibrium. A source of ^{210}Po is required; release from bottom sediments or upwelling are suggested as possibilities.

(iii) The Red Sea data of Chung et al. (1982). These are noteworthy for (a) the particularly low surface concentrations of ^{210}Po (about 0.17 to 0.25 Bq m^{-3}) and ^{210}Pb (0.30 to 0.45 Bq m^{-3}) and (b) the fact that no ^{210}Pb/^{226}Ra activity excess is observed in surface water.

(iv) The Bikini Atoll and Eniwetok Atoll data. The ^{210}Po data of Nevissi and Schell (1975) show ^{210}Po levels which tend to be on the high side, although within the 0-15° N range of Table I. They were, however, not direct measurements of ^{210}Po, and must be regarded with reserve. The data of Schell (1977) have two ^{210}Pb data points above the 0-15° N range in Table I, but are otherwise unexceptional.

(v) Enrichments of ^{210}Po and, to a lesser extent, of ^{210}Pb have been reported in the surface microlayer. The ^{210}Po enrichment has been correlated with the neuston biomass, (Bacon and Elzerman, 1980; Heyraud and Cherry, 1983).

(vi) The early Russian data for ^{210}Po cited by Parfenov (1974) show atypically high values of ^{210}Po (4 to 8 Bq m^{-3}).

3. INVENTORIES FOR 210-Po AND 210-Pb IN THE OCEAN

The data used to calculate the inventories of ^{210}Pb in the different oceans and seas are given in the Appendix.

Data for ^{210}Po, in those cases when it was measured as well as ^{210}Pb, are also listed. Depth profile data for ^{210}Po are generally sparse. As has already been noted, the ^{210}Po/^{210}Pb activity ratio, when averaged over the water column as a whole, is close to unity, so ^{210}Po total inventories are approximately the same as for ^{210}Pb. The ^{210}Po/^{210}Pb disequilibria in the top 300 m have a minimal effect on the total ^{210}Po inventories.

The inventories for ^{210}Pb for the three major oceans are given in Table II. In the Appendix, inventories for the North Sea and the Red Sea are given as well, but they will not be discussed in any detail here.

3.1 Pacific Ocean

Most of the ^{210}Pb profiles in this ocean were obtained during the GEOSECS programme, with certain additional Japanese data from the northeast Pacific. The ^{210}Pb data are much sparser than those for ^{226}Ra; the most numerous are those in the latitude bands 50°-10° N (n = 20) and 10°-50° S (n = 8). In order to facilitate comparison with the ^{226}Ra data (Bojanowski, this volume), the mean ^{210}Pb concentrations in the northern and southern Pacific have been calculated from concentrations in latitude bands of 10° irrespective of the number of data points available for each latitude band. For the whole Pacific, a ^{210}Pb inventory of 1831 ± 488 PBq is estimated; this is 63 ± 17% of the equivalent ^{226}Ra inventory of 2884 ± 197 PBq calculated by Bojanowski.

The ^{210}Pb concentrations parallel those of ^{226}Ra to a remarkable extent, showing an increase from the southwest towards the northeast. The mean concentrations of ^{210}Pb over the whole depth of the water column are 3.19 ± 0.38 Bq m^{-3} in the North Pacific and 2.07 ± 0.41 Bq m^{-3} in the South Pacific. The corresponding ^{226}Ra concentrations are 4.69 ± 0.22 Bq m^{-3} and 3.57 + 0.33 Bq m^{-3}, giving ^{210}Pb/^{226}Ra activity ratios of 0.68 ± 0.09 and 0.58 ± 0.13 respectively.

Table II. ^{210}Pb inventories in the Pacific, Atlantic and Indian Oceans. Mean concentrations are calculated from concentrations in latitude bands of 10°, except for the South Atlantic where only two data points are available.

	Volume ($10^{15}m^3$)*	Mean Bq m^{-3} (0-bottom)	Inventory (PBq)
Pacific Ocean			
North Pacific	347	3.19 ± 0.38	1107 ± 132
South Pacific	360.1	2.07 ± 0.41	745 ± 148
Whole Pacific	707.1	2.59 ± 0.69	1831 ± 488
Atlantic Ocean			
North Atlantic		1.39 ± 0.28	
South Atlantic		1.82 ± 0.69	
Whole Atlantic	346.7	1.57 ± 0.35	544 ± 121
Indian Ocean			
Northern Indian Ocean		1.30 ± 0.66**	
Southern Indian Ocean		1.99 ± 0.21**	
Whole Indian Ocean	282.6	1.84 ± 0.43**	520 ± 122

* Same reference as Bojanowski (this volume)
** Most of the data are for dissolved ^{210}Pb (see text)

The mean ^{210}Pb concentrations in the three major oceans are compared with the mean ^{226}Ra concentrations (Bojanowski, this volume) in Table III.

3.2 Indian Ocean

All the Indian Ocean data are those from the GEOSECS programme. They give an inventory for the whole Indian Ocean of 520 ± 122 PBq or 52 ± 13% of the ^{226}Ra inventory of 991 ± 61 PBq (Bojanowski, this volume). It is, however,to be noted that most (22 out of 27) of the ^{210}Pb data for this ocean refer to dissolved ^{210}Pb only. The ^{210}Pb inventory estimate is thus likely to be low, and should be raised by approximately 8% (calculated from Chung, 1987) to 562 PBq, or 57 ± 14% of the ^{226}Ra inventory.

Table III. ^{226}Ra and ^{210}Pb in the Pacific, Atlantic and Indian Oceans. Mean ^{210}Pb concentrations are calculated from concentrations in latitude bands of 10°, except for the South Atlantic where only two data points are available.

	Mean Bq m^{-3} (0-bottom) ^{226}Ra*	 ^{210}Pb	$^{210}Pb/^{226}Ra$
Pacific Ocean			
North Pacific	4.69 ± 0.22	3.19 ± 0.38	0.68 ± 0.09
South Pacific	3.57 ± 0.33	2.07 ± 0.41	0.58 ± 0.13
Whole Pacific	4.13 ± 0.65	2.59 ± 0.69	0.63 ± 0.19
Atlantic Ocean			
North Atlantic	1.94 ± 0.33	1.39 ± 0.28	0.72 ± 0.19
South Atlantic	2.93 ± 0.47	1.82 ± 0.69	0.62 ± 0.26
Whole Atlantic	2.41 ± 0.65	1.57 ± 0.35	0.65 ± 0.23
Indian Ocean			
Northern Indian Ocean	3.73 ± 0.41	1.30 ± 0.66**	0.35 ± 0.18
Southern Indian Ocean	3.48 ± 0.39	1.99 ± 0.21**	0.57 ± 0.09
Whole Indian Ocean	3.54 ± 0.37	1.84 ± 0.43**	0.52 ± 0.13

* Bojanowski, this volume
** Most of the data are for dissolved ^{210}Pb (see text)

3.3 Atlantic Ocean

Very few GEOSECS data are available for ^{210}Pb in the Atlantic Ocean. Almost all the Atlantic data are those of Bacon _et al_. (1980a) in the Labrador Sea and of Bacon (1977) in the North Atlantic. Only two profiles are available for the South Atlantic (Somayajulu and Craig, 1976).

The ^{210}Pb inventory for the whole Atlantic is estimated to be 544 ± 121 PBq, or 64 ± 16% of the corresponding ^{226}Ra inventory of 851 ± 95 PBq (Bojanowski, this volume).

The remarkable constancy of the $^{210}Pb/^{226}Ra$ activity ratio throughout the three oceans, with the exception of the North Indian Ocean, is worth noting. In the North Pacific it is 0.68 ± 0.09, in the South Pacific 0.58 ±

0.13, in the North Atlantic 0.72 ± 0.19, in the South Atlantic 0.62 ± 0.26 and in the South Indian it is, after allowing for the particulate contribution, about 0.63 ± 0.10. These ratios are based on the mean ^{210}Pb and ^{226}Ra concentrations in each latitude band. If in fact one recalculates the ^{210}Pb/^{226}Ra ratios directly for each latitude band and then takes the mean, the constancy in the ^{210}Pb/^{226}Ra ratio becomes even more striking: in the North Pacific it is 0.68 ± 0.05, in the South Pacific 0.59 ± 0.09, in the North Atlantic 0.66 ± 0.13, in the South Atlantic 0.62 ± 0.08 and in the South Indian 0.62 ± 0.04. Only for the North Indian Ocean is it noticeably lower at 0.37 ± 0.16. In view of this constancy it seems likely that the inventory estimate for the South Atlantic Ocean is reliable even though it is based on only two profiles.

4. CONCLUSION

The global inventories of ^{210}Pb in the Pacific, Atlantic and Indian Oceans are 1831 ± 488, 544 ± 121 and 520 ± 122 PBq respectively, giving a grand total of 2895 ± 517 PBq. The grand total is 61 ± 11% of the total ^{226}Ra inventory (Bojanowski, this volume). Although ^{210}Po data is much sparser, except in surface waters, the ^{210}Po inventories are approximately the same as those for ^{210}Pb. In terms of concentration, the ^{210}Pb (and presumably ^{210}Po) concentrations follow the ^{226}Ra concentrations (Bojanowski, this volume) and increase consistently from North Atlantic to South Atlantic to South Indian to South Pacific to North Pacific.

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REFERENCES

O, P ! Bacon, M.P.: ^{210}Pb and ^{210}Po results from F.S. "Meteor" cruise 32 in the North Atlantic. "Meteor" Forsch.-Ergeb., Reihe A 19, 24-36 (1977)

O, C, U ! Bacon, M.P. and A.W. Elzerman: Enrichment of ^{210}Pb and ^{210}Po in the sea-surface microlayer. Nature 284, 332-334 (1980)

O, P ! Bacon, M.P., D.W. Spencer and P.G. Brewer: ^{210}Pb/^{226}Ra and ^{210}Po/^{210}Pb disequilibria in seawater and suspended particulate matter. Earth Planet. Sci. Lett. 32, 277-296 (1976)

O, P ! Bacon, M.P., D.W. Spencer and P.G. Brewer: Lead-210 and polonium-210 as marine geochemical tracers: review and discussion of results from the Labrador Sea. In "Natural Radiation Environment III", Houston, vol. I, 473-501, U.S. Department of Energy. CONF 780422 (1980a)

P, U ! Bacon, M.P., P.G. Brewer, D.W. Spencer, J.W. Murray and J. Goddard: Lead-210, polonium-210, manganese and iron in the Cariaco Trench. Deep-Sea Res. 27A, 119-135 (1980b)

C ! Bruland, K.W., M. Koide and E.D. Goldberg: The comparative marine geochemistries of lead 210 and radium 226. J. Geophys. Res. 79, 3083-3086 (1974)

A ! Burton, W.M. and N.G. Stewart: Use of long-lived natural radioactivity as an atmospheric tracer. Nature 186, 584-589 (1960)

O, P ! Chung, Y.: ^{210}Pb and ^{226}Ra distributions in the circumpolar waters. Earth Planet. Sci. Lett. 55, 205-216 (1981)

P ! Chung, Y.: ^{210}Pb in the western Indian Ocean: distribution, disequilibrium, and partitioning between dissolved and particulate phases. Earth Planet. Sci. Lett. 85; 28-40 (1987)

O, P ! Chung, Y. and M.D. Applequist: ^{226}Ra and ^{210}Pb in the Weddell Sea. Earth Planet. Sci. Lett. 49, 401-410 (1980)

O, P ! Chung, Y. and H. Craig: ^{210}Pb in the Pacific: the GEOSECS measurements of particulate and dissolved concentrations. Earth Planet. Sci. Lett. 65, 406-432 (1983)

P, U ! Chung, Y., R.C. Finkel and K. Kim: ^{226}Ra, ^{210}Pb and ^{210}Po in the Red Sea. Earth Planet. Sci. Lett. 58, 213-224 (1982)

P ! Chung, Y., R. Finkel, M.P. Bacon, J.K. Cochran and S. Krishnaswami: Intercomparison of ^{210}Pb measurements at GEOSECS station 500 in the northeast Pacific. Earth Planet. Sci. Lett. 65, 393-405 (1983)

O, P ! Cochran, J.K., M.P. Bacon, S. Krishnaswami and K.K. Turekian: ^{210}Po and ^{210}Pb distributions in the central and eastern Indian Ocean. Earth Planet. Sci. Lett. 65, 433-452 (1983)

O, P ! Craig, H., S. Krishnaswami and B.L.K. Somayajulu: ^{210}Pb - ^{226}Ra: radioactive disequilibrium in the deep sea. Earth Planet. Sci. Lett. 17, 295-305 (1973)

C ! Folsom, T.R. and T.M. Beasley: Contribution from the alpha emitter, polonium-210, to the natural radiation environment of the marine organisms. In "Radioactive contamination of the marine environment". IAEA, Vienna, 625-632 (1973)

A ! Fukuda, K. and S. Tsunogai: Pb-210 in precipitation in Japan and its implication to the transport of continental aerosols across the ocean. Tellus, 27, 514-521 (1975)

O, P ! Goldberg, E.D.: Geochronology with Pb-210. In "Radioactive dating". IAEA, Vienna, 121- (1963)

U, C ! Heyraud, M. and R.D. Cherry: Correlation of ^{210}Po and ^{210}Pb enrichments in the sea-surface microlayer with neuston biomass. Contin. Shelf Res. 1, 283-293 (1983)

C ! Iyengar, M.A.R., V. Kannan, S. Ganapathy and P.R. Kamath: Po-210 in the coastal waters at Kalpakkam. In "Natural Radiation Environment" pp. 227-233 (K.G. Vohra, K.C. Pillai, U.C. Mishra and S. Sadavisan, eds.) J. Wiley and Sons (1982)

O, P ! Krishnaswami, S., B.L.K. Somayajulu and Y. Chung: ^{210}Pb/^{226}Ra disequilibrium in the Santa Barbara Basin. Earth Planet. Sci. Lett. 27, 388-392 (1975)

A ! Lambert, G. and M. Nezami: Importance des retombées sèches dans le bilan du plomb-210. Ann. Geophys. 21, 245-251 (1965)

A ! Lambert, G., G. Polian, J. Sanak, B. Ardouin, A. Buisson, A. Jegou et J.C. Le Roulley: Cycle du radon et de ses descendants: application à l'étude des échanges troposphère-stratosphère. Ann. Geophys. 38, 497-531 (1982)

C ! Li, Y.H., P.H. Santschi, A. Kaufman, L.K. Benninger and H.W. Feely: Natural radionuclides in waters of the New York Bight. Earth Planet. Sci. Lett. 55, 217-228 (1981)

O, P ! Moore, R.M. and J.N. Smith: disequilibria between ^{226}Ra, ^{210}Pb and ^{210}Po in the Arctic Ocean and the implications for chemical modification of the Pacific water inflow. Earth Planet. Sci. Lett. 77, 285-292 (1986)

U ! Nevissi, A. and W.R. Schell: ^{210}Po and ^{239}Pu, ^{240}Pu in biological and water samples from the Bikini and Eniwetok atolls. Nature 255, 321-323 (1975)

O, P ! Nozaki, Y. and S. Tsunogai: ^{226}Ra, ^{210}Pb and ^{210}Po disequilibria in the western North Pacific. Earth Planet. Sci. Lett. 32, 313-321 (1976)

O	Nozaki, Y., J. Thomson and K.K. Turekian: The distribution of ^{210}Pb and ^{210}Po in the surface waters of the Pacific Ocean. Earth Planet. Sci. Lett. 32, 304-312 (1976)
O, P	Nozaki, Y., K.K. Turekian and K. von Damm: ^{210}Pb in GEOSECS water profiles from the North Pacific. Earth Planet. Sci. Lett. 49, 393-400 (1980)
O, U	Parfenov, Y.D.: Polonium-210 in the environment and in the human organism. Atomic Energy Rev. 12, 75-143 (1974)
A	Peirson, D.H., R.S. Cambray and G.S. Spicer: Lead-210 and polonium-210 in the atmosphere. Tellus, 18, 427-433 (1966)
O	Rama, M. Koide and E.D. Goldberg: Lead-210 in natural waters. Science 134, 98-99 (1961)
A	Robbins, J.A.: Geochemical and geophysical applications of radioactive lead. In: The biogeochemistry of lead in the environment. Part A. 285-393, J.O. Nriagu ed. Elsevier/North Holland Biomedical Press (1978)
C	Santschi, P.H., Y.H. Li and J. Bell: Natural radionuclides in the water of Narragansett Bay. Earth Planet. Sci. Lett. 45, 201-213 (1979)
U, C	Schell, W.R.: Concentrations, physio-chemical states and mean residence times of ^{210}Pb and ^{210}Po in marine and estuarine waters. Geochim. Cosmochim. Acta 41, 1019-1032 (1977)
O	Shannon, L.V., R.D. Cherry and M.J. Orren: Polonium-210 and lead-210 in the marine environment. Geochim. Cosmochim. Acta 34, 701-711 (1970)
O, P	Somayajulu, B.L.K. and H. Craig: Particulate and soluble ^{210}Pb activities in the deep sea. Earth Planet. Sci. Lett. 32, 268-276 (1976)
O, P U	Spencer, D.W., M.P. Bacon and P.G. Brewer: The distribution of ^{210}Pb and ^{210}Po in the North Sea. Thalassia jugosl. 16, 125-154 (1980)
C	Tanaka, N., Y. Takeda and S. Tsunogai: Biological effect on removal of Th-234, Po-210 and Pb-210 from surface waters in Funka Bay, Japan. Geochim. Cosmochim. Acta 47, 1783-1790 (1983)
O, P	Thomson, J. and K.K. Turekian: ^{210}Po and ^{210}Pb distributions in ocean water profiles from the eastern South Pacific. Earth Planet. Sci. Lett. 32, 297-303 (1976)
P	Tsunogai, S. and K. Harada: ^{226}Ra and ^{210}Pb in the western North Pacific. In "Isotope marine chemistry", 157-164, Uchida Rokakuho, Tokyo 1980.
O	Tsunogai, S. and Y. Nozaki: Lead-210 and polonium-210 in the surface water of the Pacific. Geochem. J. 5, 165-173 (1971)
O	Turekian, K.K., D.P. Kharkar and J. Thomson: The fates of ^{210}Pb and ^{210}Po in the ocean surface. J. Rech. atmos. 8, 639-646 (1974)

O, P ! Turekian, K.K. and Y. Nozaki: ^{210}Po and ^{210}Pb in the eastern South Pacific. The role of upwelling on their distributions in the water column. In "Isotope marine chemistry", 157-164, Uchida Kokakuho, Tokyo 1980.

A ! Turekian, K.K., Y. Nozaki and L.K. Benninger: Geochemistry of atmospheric radon and radon products. Ann. Rev. Earth Planet. Sci. 5, 227-255 (1977)

C ! Wong, K.M., V.F. Hodge and T.R. Folsom: Plutonium and polonium inside giant brown algae. Nature 237, 460-462 (1972)

O = Open ocean (surface)
P = Profiles
U = Unusual
C = Coastal (surface)
A = Atmosphere

Data for Lead-210, Polonium-210 Inventories: Pacific Ocean

Coordinates	^{210}Pb		^{210}Po		Reference
	Bq cm^{-2}	Bq m^{-3}	Bq cm^{-2}	Bq m^{-3}	
57° 01 N, 170° 03 E	1.05	2.76			Tsunogai & Harada (1980)
44° 06 N, 154° 03 E	1.97	3.85			"
44° 40 N, 177° 30 W	2.28	3.81			Nozaki *et al.* (1980)
" "	1.67	2.79			Chung & Craig (1983)
40° 10 N, 160° 30 E	1.47	2.61			"
34° 10 N, 127° 54 W	1.39	2.95			"
34° 58 N, 151° 50 E	2.19	3.58			Nozaki *et al.* (1980)
33° 06 N, 139° 34 W	2.11	4.21			"
32° 01 N, 176° 59 W	1.97	3.72			"
30° 58 N, 168° 28 W	1.77	3.09			Chung & Craig (1983)
30° 36 N, 170° 36 E	2.03	3.64	2.11	3.78	Bacon *et al.* (1976)
" "	2.06	3.68			Nozaki *et al.* (1980)
30° 00 N, 159° 50 W	2.18	3.74			"
28° 30 N, 121° 30 W	1.44	3.36			Craig *et al.* (1973)
28° 28 N, 144° 58 E	1.87	3.18			Nozaki & Tsunogai (1976)
28° 28 N, 145° 00 E	1.35	2.30			"
28° 27 N, 144° 59 E	2.31	3.95			Tsunogai & Harada (1980)
28° 24 N, 145° 00 E	2.19	3.73			Nozaki & Tsunogai (1976)
28° 27 N, 122° 11 W	1.36	3.17			Chung & Craig (1983)
" "	1.40	3.27			Chung *et al.* (1983)
" "	1.33	3.10			"
" "	1.52	3.55			"
25° 00 N, 170° 00 E	2.25	3.82			Nozaki *et al.* (1980)
16° 31 N, 122° 59 W	1.28	3.04			Chung & Craig (1983)
" "	1.54	3.66			Nozaki *et al.* (1980)
14° 06 N, 178° 38 W	1.61	2.82			Chung & Craig (1983)
" "	2.31	4.07			Nozaki *et al.* (1980)
12° 54 N, 173° 28 W	2.14	3.73			"
04° 33 N, 178° 59 E	1.54	2.69			Chung & Craig (1983)
04° 38 S, 125° 08 W	1.09	2.44			"
15° 28 S, 75° 52 W	0.96	2.39	0.93	2.32	Turekian & Nozaki (1980)
16° 41 S, 167° 03 W	1.48	2.59			Chung & Craig (1983)
18° 59 S, 74° 58 W	1.10	2.63	0.94	2.23	Thomson & Turekian (1976)
23° 44 S, 153° 37 W	1.14	2.48			Chung & Craig (1983)
23° 57 S, 174° 31 W	0.99	1.65			"
32° 49 S, 163° 35 W	1.20	2.13			"
33° 20 S, 128° 24 W	1.06	2.54 D	0.97	2.32 D	Bacon *et al.* (1976)
44° 56 S, 166° 39 W	0.74	1.39			Chung & Craig (1983)
57° 31 S, 169° 47 E	0.93	1.78			"
69° 18 S, 173° 30 E	0.80	1.92			"

D = dissolved

Data for Lead-210, Polonium-210 Inventories: Indian Ocean

Coordinates	^{210}Pb		^{210}Po		Reference
	Bq cm^{-2}	Bq m^{-3}	Bq cm^{-2}	Bq m^{-3}	
13° 21 N, 53° 37 E	0.23	0.83 D			Chung (1987)
12° 30 N, 84° 29 E	0.28	0.83 D	0.24	0.72 D	Cochran *et al.* (1983)
8° 31 N, 86° 03 E	0.49	1.33 D	0.52	1.43 D	"
6° 12 N, 64° 24 E	0.91	1.94 D			Chung (1987)
5° 00 N, 79° 57 E	0.64	1.53 D	0.42	1.00 D	Cochran *et al.* (1983)
3° 56 N, 56° 50 E	0.86	1.86			Chung (1987)
0° 01 N, 80° 03 E	1.00	2.15 D	0.87	1.87 D	Cochran *et al.* (1983)
0° 04 S, 50° 53 E	0.99	1.94			Chung (1987)
1° 12 S, 90° 45 E	0.96	2.08 D	1.02	2.20 D	Cochran *et al.* (1983)
5° 00 S, 80° 00 E	1.30	2.53 D	1.17	2.28 D	"
5° 02 S, 91° 47 E	1.23	2.50 D			"
6° 09 S, 50° 55 E	0.93	1.92 D			Chung (1987)
9° 22 S, 95° 02 E	1.18	2.25 D			Cochran *et al.* (1983)
10° 01 S, 79° 59 E	1.22	2.28 D	0.68	1.28 D	"
12° 17 S, 53° 39 E	0.96	2.03			Chung (1987)
14° 59 S, 79° 58 E	1.26	2.52 D	1.23	2.45 D	Cochran *et al.* (1983)
17° 18 S, 55° 51 E	0.93	2.05 D			Chung (1987)
19° 29 S, 101° 18 E	1.39	2.38 D	1.41	2.42 D	Cochran *et al.* (1983)
20° 06 S, 79° 59 E	1.11	2.30 D	1.16	2.40 D	"
23° 00 S, 74° 01 E	0.78	1.87 D	0.74	1.78 D	"
27° 00 S, 67° 06 E	0.73	1.47 D	0.59	1.18 D	"
27° 04 S, 56° 58 E	0.96	1.88 D			Chung (1987)
29° 15 S, 109° 58 E	1.24	2.22 D	1.15	2.07 D	Cochran *et al.* (1983)
37° 48 S, 57° 35 E	0.91	1.71			Chung (1987)
47° 40 S, 37° 52 E	0.79	1.72 D			"
59° 59 S, 60° 59 E	0.97	2.04			"
64° 11 S, 83° 59 E	0.75	2.07 D			"

D = dissolved

Data for Lead-210, Polonium-210 Inventories: Atlantic Ocean

Coordinates	^{210}Pb		^{210}Po		Reference
	Bq cm^{-2}	Bq m^{-3}	Bq cm^{-2}	Bq m^{-3}	
53° 40.0 N, 40° 00.8 W	0.42	1.21	0.38	1.10	Bacon *et al.* (1980)
52° 41.7 N, 35° 28.3 W	0.41	1.02	0.37	0.92	"
51° 10.5 N, 43° 40.0 W	0.51	1.28	0.45	1.13	"
47° 45.5 N, 35° 47.0 W	0.49	1.10	0.40	0.90	"
33.8° N, 13.4° W	0.72	1.63	0.63	1.42	Bacon (1977)
21.7° N, 24.0° W	0.86	1.73	0.95	1.93	"
16.5° N, 31.9° W	0.85	1.72	0.85	1.73	"
15.7° N, 48.5° W	0.55	1.45	0.55	1.43	"
14.7° N, 57.5° W	0.52	0.97	0.63	1.18	"
14.5° N, 66.0° W	0.51	1.05	0.50	1.02	"
4° S, 29° W	0.67	1.33			Somayajulu & Craig (1976)
60° S, 0°	1.21	2.30			"

Data for Lead-210, Polonium-210 Inventories: North Sea

Coordinates	^{210}Pb		^{210}Po		Reference
	Bq m^{-2}	Bq m^{-3}	Bq m^{-2}	Bq m^{-3}	
58° 26.5 N, 1° 46.6 E	41	0.37	66	0.60	Spencer *et al.* (1980)
59° 19.9 N, 1° 45.2 E	148	1.35	107	0.97	"
59° 19.6 N, 0° 0.5 E	87	0.62	98	0.70	"
58° 25.8 N, 0° 0.5 E	67	0.50	94	0.70	"
58° 25.5 N, 1° 14.3 E	35	0.32	60	0.55	"
58° 54.6 N, 2° 49.9 E	117	1.02	109	0.95	"
58° 50.3 N, 0° 41.4 E	73	0.52	98	0.70	"
60° 20.5 N, 1° 57.0 E	122	1.52	106	1.33	"
61° 29.5 N, 3° 26.6 E*	402	1.15	413	1.18	"
58° 50.0 N, 0° 40.0 E	67	0.48	112	0.80	"
61° 49.5 N, 0° 49.1 E*	433	1.97	425	1.93	"
63° 50.0 N, 0° 51.8 E*	1914	0.87	2090	0.95	"
58° 50.5 N, 0° 38.8 E	74	0.53	98	0.70	"
* Excluding samples from the Norwegian Sea Mean value	83±36	0.72±0.42	95±18	0.80±0.23	

Volume = 55 x 10^{12} m^3**

	^{210}Pb	^{210}Po
Total inventory (TBq)	40±23	44±13

** The Water Encyclopedia. A compendium of useful information on water resources. D.K. Todd, ed. Water Information Center, Port Washington, N.Y. (1970)

Data for Lead-210, Polonium-210 Inventories: Red Sea

Coordinates	^{210}Pb		^{210}Po		Reference
	Bq cm^{-2}	Bq m^{-3}	Bq cm^{-2}	Bq m^{-3}	
27° 14 N, 34° 28 E	0.057	0.48 D			Chung *et al.* (1982)
19° 57 N, 38° 29 E	0.072	0.37	0.039	0.20	"
14° 42 N, 42° 10 E	0.015	0.25 D			"
12° 07 N, 43° 55 E	0.040	0.58 D			"
Mean value	0.046±0.024	0.42±0.14			

Volume = 251 x 10^{12} m^3*

Total inventory (TBq) = 105±35

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* The Water Encyclopedia. A compendium of useful information on water resources. D.K. Todd, ed. Water Information Center, Port Washington, N.Y. (1970)

INVENTORY OF RADIUM ISOTOPES IN THE OCEANS

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Abstract

Inventories are derived for ^{226}Ra in the oceans, using profile data from the literature, especially the GEOSECS cruises. Several methods yielded identical results within statistical error. Recommended values are: Atlantic Ocean (including the Arctic) 0.84 ± 0.10 E 18 Bq; Pacific 2.92 ± 0.16 E 18 Bq; Indian 1.02 ± 0.09 E 18 Bq; giving a grand total of 4.78 ± 0.27 E 18 Bq. Losses from radioactive decay of 2.046 E 15 Bq y^{-1} are approximately balanced by the flux from the sea floor. Losses from the sea surface are about E 10 Bq y^{-1}. River input is about 0.42 E 15 Bq y^{-1} and ground water influx provides 3-22 E 13 Bq y^{-1}. The errors are sufficiently large, that no departures from equilibrium caused by anthropogenic activities can be demonstrated.

1. INTRODUCTION

There are four radium isotopes occuring in nature, all being radioactive. Their nuclear characteristics are as follows:

	^{223}Ra	^{224}Ra	^{226}Ra	^{228}Ra
Name of series	actinium	thorium	uranium	thorium
Type	4n+3	4n	4n+2	4n
First member	^{235}U	^{232}Th	^{238}U	^{232}Th
Immediate parent	^{227}Th	^{228}Th	^{230}Th	^{232}Th
Descendant	^{219}Rn	^{220}Rn	^{222}Rn	^{228}Ac
Mode of decay	alpha	alpha	alpha	beta
Half-life	11.436d	3.665d	1617y	5.7y
Specific activity /Bq g^{-1}/	1.90E15	5.89E15	362E10	1.01E13

These radium isotopes have a common chemical predecessor thorium which, due to its high chemical reactivity, is almost absent from the open ocean water /7,29,37,40,51,58,61,63/. It means that virtually all the radium present in seawater must have been derived from external sources. It has long been recognized that the principal source of oceanic radium is the seafloor which acts as a sink for the thorium isotopes continuously produced from uranium in the water column /73,91/. The decaying thorium atoms recoil radium nuclei to interstitial waters from which they diffuse to the overlying bottom water and spread over the whole ocean by diffusion and transport processes /23,24,36/. The time scale for oceanic mixing and circulation processes is such that the shortest-lived ^{223}Ra and ^{224}Ra are unable to reach the open ocean. They are thus confined to a narrow near-shore water zone, close to their place of origin, and are hardly detectable beyond the shelf break /49/. In the open ocean they are intimately associated with their longer-lived grandparents and parents /^{231}Pa - ^{227}Th and ^{228}Th/ from which they form. As the concentrations of protactinium and thorium isotopes in sea water are extremely low, rarely exceeding 10^{-2} Bq m^{-3} /61,68/ the ^{223}Ra and ^{224}Ra isotopes present no radiological interest and deserve no further attention.

Of the remaining radium isotopes the ^{226}Ra is more important and the best studied one. The half-life is long enough for this nuclide to sustain an independent existence in the oceans and the relatively stable oceanic circulation system has resulted in a quasi steady-state ^{226}Ra distribution in all parts of it, including even the larger inland seas with a restricted water exchange. The concentrations of ^{226}Ra in seawater vary within a rather narrow range of 0.8-8 Bq m^{-3}. Specific analytical methods based on radon extraction were devised to measure these activities with high accuracy. As a result, a large number of good quality data have became available for inventory purposes.

As far as ^{228}Ra is concerned the situation is less satisfactory. Apart from coastal areas, where the concentrations are comparable to those of ^{226}Ra, activity levels of ^{228}Ra decrease progressively in the seaward direction attaining sometimes values as low as 10^{-2}Bq m^{-3}. Surface concentrations display a rather large scatter whereas in deep waters the levels are so low that precise measurements pose serious problems. Due to the patchy distribution of ^{228}Ra, both horizontally and vertically, and its transient character, an estimation of its inventories can be made-at present-only very approximately.

2. DIMENSIONS AND LIMITS OF THE OCEANS

The basic data on volumes, surfaces and depths were drawn from the Marine Atlas /32/. The World Ocean has been considered to consist of three majorbasins /the North Polar Ocean being merged in the Atlantic Ocean/ each including the adjacent seas that connect with them. The boundaries between the oceans were set as recommended by the International Hydrographic Bureau./Spec.Publ.No.23, 3nd ed., Monte Carlo, 1953/. While the areas of oceanic surfaces differ only very slightly from one source to another /2,32,95/, the differences in volumes and the mean depths are somewhat larger but do not exceed 2.5% compared to Kossina's data /95/.

In inventory calculations a more detailed division of water volumes and surfaces is desirable as it helps improve the precision of estimates by reducing the overall calculation error. The areas of oceans divided into latitude strips were taken from /2/. The respective volumes were not available except for the Pacific Ocean /Whitehead's unpublished data/. The volumes of oceans between selected isobaths were determined from bathygraphic curves reproduced in /32/. They are presented in Table 1.

TABLE 1. VOLUMES OF OCEANS IN DIFFERENT WATER LAYERS AS DERIVED FROM BATHYGRAPHIC CURVES [32]

LAYER	PACIFIC 707.1 x $10^{15}m^3$		ATLANTIC 329.7 x $10^{15}m^3$		ARCTIC 17.0 x $10^{15}m^3$		ATL.+ARCTIC 346.7 x $10^{15}m^3$		INDIAN 282.6 x $10^{15}m^3$	
m	Δ%	Σ%	Δ%	Σ%	Δ%	Σ%	Δ%	Σ%	Δ%	Σ%
0-500	11.75	11.75	12.62	12.62	26.16	27.16	13.33	13.33	12.53	12.53
500-1000	10.94	22.69	11.75	24.37	17.25	44.41	12.02	25.35	11.88	24.41
1000-2000	22.06	44.75	23.28	47.65	29.52	73.93	23.59	48.94	23.36	47.77
2000-3000	20.13	64.88	21.00	68.65	19.67	93.60	20.93	69.87	22.11	69.88
3000-4000	18.09	82.97	17.56	86.21	6.11	99.71	17.00	86.87	17.65	87.53
4000-5000	11.13	93.10	10.74	96.95	0.29	100.00	10.23	97.10	10.04	97.57
5000-6000	3.48	97.58	2.56	99.51			2.43	99.53	2.32	99.89
6000-7000	0.69	98.27	0.33	99.84			0.31	99.84	0.11	100.00
7000-8000	0.64	98.91	0.16	100.00			0.16	100.00		
8000-9000	0.57	99.48								
>9000	0.52	100.00								

3. SURVEY OF DATA FOR ^{226}Ra INVENTORY

Although ^{226}Ra was the first radioactive element ever measured in seawater /83/ it was not until 1938 that correct levels of this radionuclide began to appear in the literature /26/. The earlier works were reviewed and critically evaluated in /28/ and /89/. In 1967 Szabo /84/ made a compilation of more recent data which were about 400 in number. Using these data he assigned representative concentration ranges to the oceans and several seas, but a more detailed picture of radium distribution in oceans remained obscure until the works of Broecker et al. /5,6/, which brought the first detailed depth profiles for this radionuclide.

The proncipal and the most abundant source of radium data suitable for inventory purposes were GEOSECS profiles. The results concerning ^{226}Ra were available in numerous publications /all listed in the references/ and are included in the recently issued GEOSECS atlas of shorebased analyses /29/. These were complemented with some more recent results for the Pacific from Japanese cruises /35, 86/ and for the Atlantic from "Meteor" cruise, FRG /75/ and the TTO Programme /67/. In total, 166 profiles were considered: 57 for the Atlantic, 69 for the Pacific and 40 for the Indian Ocean. Each profile consited of 6 to 26 samples which makes a total of nearly 3000 individual radium samples.

4. QUALITY OF DATA

The quality of GEOSECS data were checked during numerous intercalibration exercises which were attended by all groups involved in radium measurements. The error on the individual result expresed as the standard deviation of a single measurement divided by the square root of the number of consecutive measurements was found to be close to ± 4% in all groups. The first intercalibration revealed, however, a systematic 20% range between the mean profiles obtained by the participating groups /9/. Subsequent intercalibrations in which a common ^{226}Ra standard was used substantially improved data comparability, with the maximum discrepancy of only 10% /22/. In the final exercise the mean systematic difference between profiles measured by two independent groups was reduced to 2±3% /20/. The Japanese workers made a quality test measuring radium profiles at stations occupied earlier by a GEOSECS group. The results were identical within ± 3% /86/. Some measurements following the GEOCECS Programme are characterized by even higher precision attaining ±1% in some cases /75,79/. It can thus be concluded that all the data utilised in this inventory are of proved quality.

5. INVENTORY CALCULATION

The calculation of radium standing crop under surface unit area for a given water column was approached by two ways: graphically and arithmetically. In the first method the radium values were plotted against depth on a square grid paper and the best line was drawn trough the points. The figure representing a vertical distribution of ^{226}Ra in a water column /several examples of such profiles are given in Fig.1/ was then dissected and weighted on an analytical balance. To obtain subinventories for different water layers it was

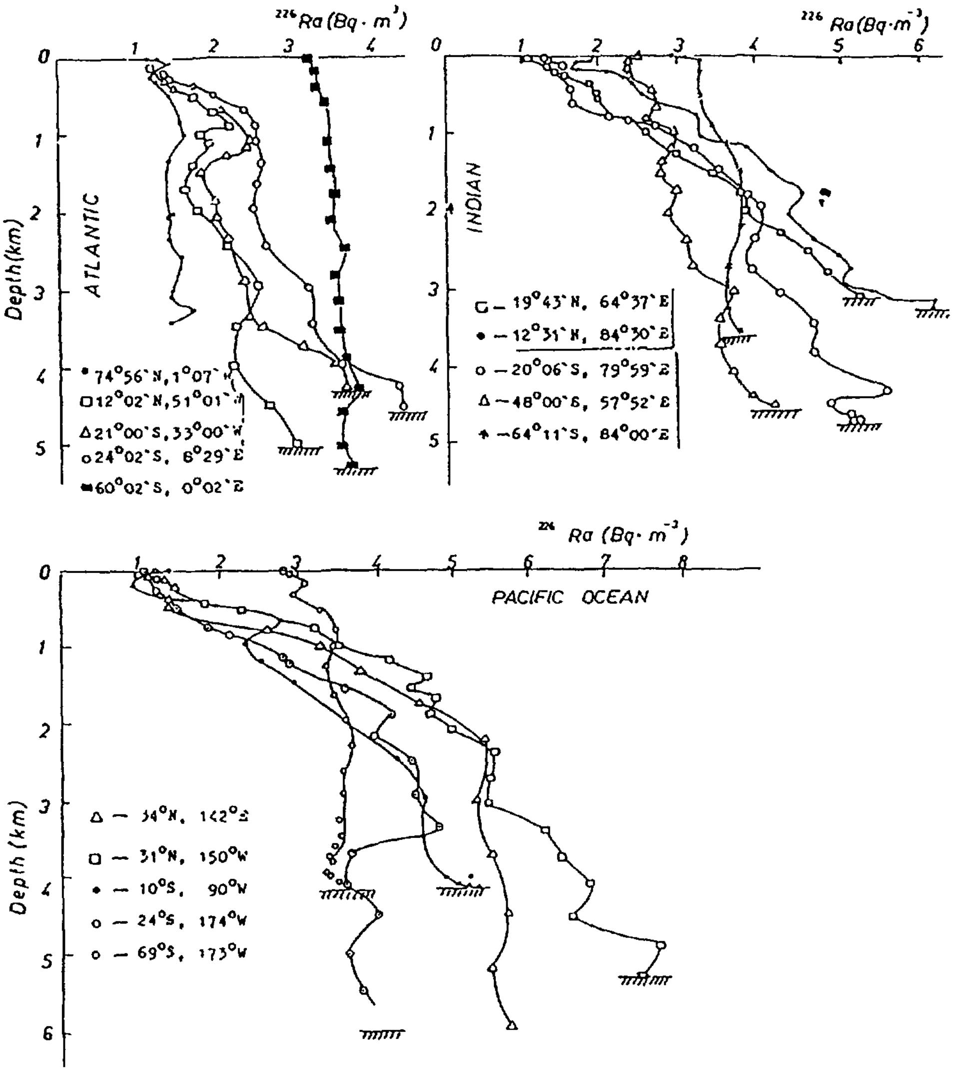

FIG.1. Typical ^{226}Ra profiles in oceans.

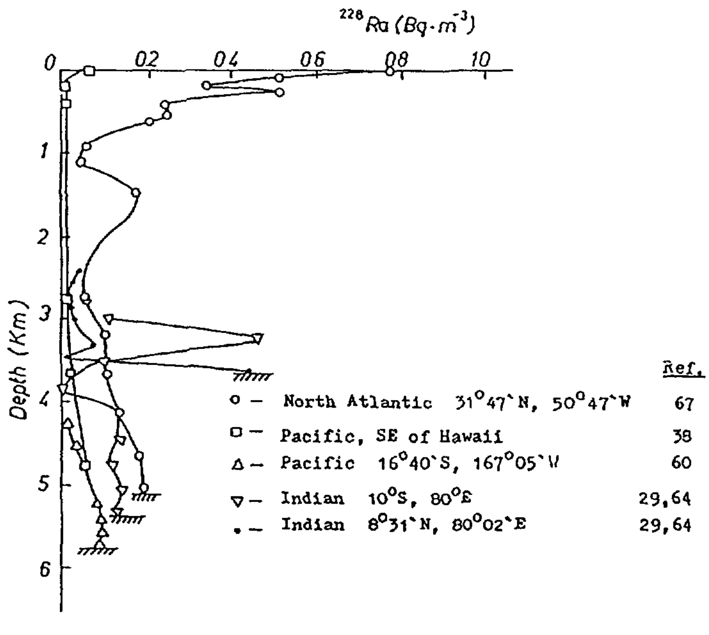

FIG.2. Vertical distribution of ^{228}Ra in oceans.

further cut in strips along the isobaths of 0-0.5km and then by 1km up to 6km isobath, and each strip was weighted separately. Mean radium concentrations for different water layers and for the entire profile were then calculated by dividing the standing crops by the respective depths and thincknesses of water layers. To convert different activity concentration units into Bq m^{-3} a mean seawater density of 1.025 kg m^{-3} and ^{226}Ra specific activity of 3.62×10^{10} Bq g^{-1} were used throughout. The reproducibility of this method was found to be better than 2% except for the upper 0-0,5km layer where it was 5-10% depending on the shape of the profile. All Pacific profiles were processed using this method and the results are shown in Table 3.

For the Atlantic and the Indian Oceans the lengthy graphical procedure was replaced by a simpler arithmetic method after it had been found that both methods yielded the same results within 1-4% for the entire profiles. A linear interpolation was made between the data points and the inventory was calculated by summing up the products of the mean radium concentrations for each water layer by the thickness of these layers. The results are compiled in Tables 2 and 4.

From the profile and sub-profile inventories presented in Tables 2-4 one can derive total inventories for each ocean using different methods of calculation.

The simplest one is to assume that the weighted ^{226}Ra concentrations in profiles are distributed randomly over the whole ocean and that the distribution is normal. The inventories obtained by multiplying the arithmetic mean ^{226}Ra concentrations of all profiles in a given ocean by the volumn of this ocean are shown in Table 6 line c/. The values are accompanied with rather large errors which arised from the adopted assumptions.

In order to arrive at more accurate estimates one must recognize that ^{226}Ra concentrations in oceanic water vary both horizontally and vertically /the latter is clearly seen in Fig.1/. Indeed, vertical profiles show up to sevenfold differences between surface and near-bottom concentrations, whereas meridionally there are threefold differences in surface ^{226}Ra concentrations. To account for this variability subinventories were calculated for the horizontal water layers /the volumes had been determined as described in par.2/ by multiplying the average ^{226}Ra concentrations in the layers by their volumes. Since the coverage of oceans with ^{226}Ra profiles was uneven, the oceans were divided in strips by 10° latitude and the mean ^{226}Ra concentrations were calculated for each strip, together

Text continued on p.169.

TABLE 2. ^{226}Ra PROFILES IN THE ATLANTIC OCEAN

Latitude band	Coordinates		Depth m	Mean ^{226}Ra concentration in layer /km/ Bq m^{-3}*								$\sum^{226}$Ra Bq cm^{-2}	Reference
				0-0.5	0.5-1	1-2	2-3	3-4	4-5	>5	0-bottom		
1	2	3	4	5	6	7	8	9	10	11	12	13	14
90-80°N	85°50'	108°50'	1800	1.77	1.34	1.66					1.61	0.289	56
80-70°N	74°56'	1°07'	3703	1.29	1.55	1.42	1.46	1.46			1,44	0.533	6
70-60°N	63°32'	35°14'	2406	1.41	1.36	1.58	1.79				1.54	0.370	6
60-50°N													
50-40°N	46°02'	11°02'	4775	/1.40/	/1.60/	/1.80/	2.32	2.85	3.16		2.29	1.092	75
	40°18'	18°34'	5400	/1.40/	/1.60/	/1.80/	2.08	2.75	3.11	3.20	2.32	1.252	75
40-30°N	39°00'	44°02'	3969	1.34	1.61	1.69	1.88	2.00			1.76	0.697	67
	38°17'	69°10'		1.60									67
	35°59'	47°01'	4995	1.26	1.69	1.55	1.71	1.99	2.30		1.81	0.905	56
	35°57'	47°00'	4583	1.43	1.66	1.86	2.08	2.19	2.28		1.96	0.901	67
	35°47'	68°00'	5000	1.27	1.52	1.65	1.83	1.91	2.13		1.78	0.890	22
	34°39'	67°21'		1.22									67
	33°	13°	4440	1.37							2.09	0.930	1
	32°47'	23°16'	5350	/1.40/	/1.58/	/1.76/	2.33	2.82	3.10	3.22	2.36	1.263	75
	31°47'	50°47'	5049	1.43	1.55	1.72	2.02	2.23	2.53		2.00	1.012	67
	31°40'	68°28'		1.44									7
	30°41'	38°58'	3644	1.33	1.50	1.72	2.16	2.76			1.94	0.706	29
30-20°N	28°44'	25°10'	5260	/1.40/	/1.58/	/1.76/	/2.09/	2.96	3.10	3.26	2,33	1.224	75
	28°02'	26°00'	5308	1.40	1.58	1.76	2.09	2.63	3.04	3.15	2:26	1.198	6
	27°00'	53°34'	6048	1.29	1.55	1.80	2.22	2.38	2.76	3.02	2.27	1.375	29
	22-30°	55-62°	5600	1.47	1.57	2:07	2.37	2.72	2.91	3.16	2.41	1.349	8
	24°46'	26°59'	5450	/1.39/	/1.63/	/1.89/	2.41	2.82	/2.84/	/3.16/	2.36	1.289	75
	23°50'	54°02'	4295	1.50	1.70	1.93	2.28	2.40	2.54		2.09	0.896	67
	21°01'	53°59'		1.38									
	21°	24°	4950	1.38	1.69	2.01	2.26	2.74	2.87		2.24	1.107	1
	21°00'	54°00'	5234	1.34	1.65	1.82	2.15	2.38	2.59	2.74	2.12	1.108	29
20-10°N	16°28'	27°01'	4565	/1.47/	/1.85/	/1.99/	2.28	2.89	3.36		2.35	1.072	75
	16°	32°	4940	1.57	2.01	1.98	2.21	2.79	3.04		2.20	1.090	1
	15°	49°	3820	1.48	1.95	1.77	2.10	2.22			1.90	0.726	1
	15°	57°	5003	1.37							1.88	0.939	1
	15°	66°	4880	1.37	1.89	1.99	2.02				1.90	0.926	1
	15°02'	53°57'		1.56									67
	15°	46°	4000	1.54	2.29	1.93	2.22	2.64			2.18	0.871	5
	12°02'	51°01'	5069	1.43	1.97	1.76	2.18	2.33	2.64	2.97	2.13	1.080	29
10-0°N	9°29'	26°00'	5300	/1.47/	/2.00/	/1.85/	2.23	2.59	2.97	3.21	2.33	1.234	75
	3°57'	38°31'	4400	1.49	2.00	1.83	1.98	2.34	2.59		2.03	0.893	6
	2°31'	19°29'	5020	/1.53/	/2.12/	/1.89/	2.26	2.67	2.89	2.96	2.31	1.159	75
	2°00'	14°01'	5158	1.53	2.12	1.89	2.23	2.51	2.73	2.82	2.26	1.164	29
	0°00'	19°29'	5200	/1.53/	/2.12/	/1.89/	2.35	2.47	3.51	3.75	2.46	1.280	75
0-10°S	4°00'	29°00'	5034	1.55	1.64	1.78	2.05	2.33	3.57	3.75	2.27	1.145	29
	5°02'	24°02'	5300	/1.55/	/1.64/	1.99	2.27	2.54	3.72	4.14	2.52	1.335	75
10-20°S	12°00'	2°00'E	5571	1.63	2.32	2.32	2.62	3.09	3.21	3.17	2.70	1.502	6
	15°03'	29°32'	5085	1.54	2.29	2.03	2.31	2.35	3.48	3.62	2.44	1.240	6
20-30°S	21°00'	33°00'	4072	1.46	2.15	2.06	2.21	2.78	3.59		2.32	1.000	29
	24°00'	8°29'E	4637	1.61	2.42	2.51	2.71	3.25	4.52		2.88	1.336	45
	27°00'	37°01'	4589	1.24	2.07	2.32	2.25	2.76	3.51		2.41	1.105	29
30-40°S	32°58'	42°31'	4415	1.37	1.93	2.58	2.31	2.91	3.98		2.51	1.110	6
40-50°S	41°46'	18°27'E	4947	1.61	2.71	2.94	2.93	3.23	3.87		3.02	1.493	45
	48°39'	46°01'	5970	2.26	2.78	3.03	3.23	3.52	3.71	3.98	3.33	1.986	45
	49°34'	11°28'E	4190	2.74	3.09	3.13	3.55	3.63			3.33	1.394	45
50-60°S	55°01'	50°02'	4143	2.63	3.20	3.34	3.56	3.52	3.66		3.34	1.359	45
	56°16'	24°55'	7808	3.00	3.42	3.59	3.52	3.55	3.64	3.61	3.54	2.764	45
	57°44'	66°08'	4605	2.59	2.83	3.38	3.63	3.95	3.93		3.48	1.605	45
60-70°S	60°02'	0°02'E	5364	3.27	3.43	3.52	3.58	3.59	3.60	3.66	3.54	1.897	45
	62°21'	28°16'	4200	3.38	/3.38/	/3.50/	/3.58/	/3.58/	3.64		3.52	1.477	18
70-80°S	70°22'	14°44'	4000	3.46	/3.50/	/3.54/	/3.58/	/3.60/			3.55	1.420	18
	73°27'	38°19'	2500	3.05	3.24	/3.45/	3.55				3.35	0.837	18
	75°52'	39°43'	500	2.88							2.88	0.144	18

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* The values are weighted means of the data available for a given depth interval in the profile /usually 2 8/; in brackets are values interpolated from the neighboring profiles.

TABLE 3. ^{226}Ra PROFILES IN THE PACIFIC OCEAN

Latitude band	Coordinates		Depth	^{226}Ra concentration in layer /km/ Bq m^{-3}								Average for profile	$\sum{}^{226}$Ra	Ref.
	φ	λ	m	0-0.5	0.5-1	1-2	2-3	3-4	4-5	5-6	>6		Bq cm^{-2}	
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
60°-50° N	57°05'N	177°05'W	3650	1.57	2.79	3.80	5.64	7.15				4.46	1.63	86
	57°01'	179°03'E	3835	1.31	2.41	4.11	5.43	6.94				4.48	1.72	86
	57°00'	174°54'E	3820	1.51	2.79	3.90	5.89	6.90				4.61	1.76	86
	57°00'	171°00'E	1900	1.64	3.15	3.80						3.06	0.58	86
	53°07'	177°18'W	3721	2.77	3.42	4.63	5.59	6.07				4.75	1.77	20
	50°27'	176°35'W	7300	2.31	3.80	4.56	5.03	5.43	5.39	5.60	5.80	5.01	3.66	20
50°-40° N	44°40'	177°03'W	5900	2.09	3.44	4.52	5.08	5.28	6.15	5.49		4.87	2.87	20
	44°04'	154°02'E	5130	1.48	2.53	3.75	5.23	6.47	6.03	6.05		4.73	2.43	86
	43°04'	138°33'E	3515 J	1.97	2.55	2.59	2.61	2.58				2.50	0.88	35
	41°20'	137°20'E	3578 J	1.90	2.43	2.55	2.62	2.66				2.45	0.88	35
	40°10'	160°30'E	5571	2.15	3.53	4.43	5.42	5.11	5.32	5.68		4.73	2.64	35
40°-30° N	38°35'	134°45'E	2953 J	2.00	2.54	2.56	2.58					2.47	0.73	35
	37°44'	135°12'E J	2925 J	1.71	2.27	2.51	2.58					2.35	0.69	35
	34°53'	151°51'E	6127	1.26	2.48	4.10	5.31	5.52	5.55	5.57	5.57	4.67	2.86	20
	34°18'	120°04'W SB	570 SB	1.75	2.53							1.85	0.11	44
	34°16'	141°38'E	9200	1.27	2.27	4.01	5.32	5.35	5.81	5.78	5.71	5.03	4.63	20
	34°08'	127°57'W	4705	1.89	3.12	4.51	5.29	6.02	7.05			4.95	2.33	20
	33°08'	139°05'W	4994	1.51	3.53	4.50	5.41	6.06	6.62			5.02	2.51	20
	32°37'	161°55'E	5944	1.36	2.57	4.26	5.44	5.40	5.65	6.00		4.82	2.86	20
	32°04'	177°07'W	5294	1.39	2.80	4.22	4.89	5.26	5.75	5.70		4.52	2.40	20
	31°22'	150°01'W	5403	1.53	3.06	4.54	5.67	6.29	6.81	7.17		5.27	2.85	20
	31°02'	168°27'W	5669	1.45	2.82	4.05	4.89	5.40	6.05	6.46		4.74	2.68	20
	30°34'	170°37'E	5590	1.26	2.65	4.17	5.18	5.94	5.90	5.53		4.73	2.64	20
30°-20° N	29°39'	159°53'W	5820	1.36	2.57	4.21	5.09	5.75	6.28	6.72		4.92	2.82	20
	28°30'	121°30'W	4280	1.91	3.11	4.28	5.75	6.64	6.97			4.94	2.12	20
	28°27'	144°59'E	5850	1.65	2.73	4.61	5.92	6.09	6.42	6.56		5.26	3.08	86
	23-29°	162° E	5000	1.35	2.33	4.15	4.82	5.49	5.81			4.42	2.21	8
	25°00'	170°05'E	5876	1.22	2.97	4.23	5.50	5.80	5.51	5.37		4.74	2.79	20
20°-10° N	19°57'	139°42'E	4825	1.83	3.08	4.67	5.48	6.47	6.60			5.08	2.45	86
	17°	161° W	5535	1.55	2.97	4.17	5.50	6.37	6.20	5.82		4.99	2.76	20
	16°31'	123°00'W	4262	1.82	2.95	4.18	5.55	6.90	7.32			4.91	2.09	46
	14°07'	178°34'W	5657	1.35	2.92	4.12	5.42	5.82	5.10	4.92		4.56	2.58	20
	12°53'	173°28'E	5719	1.55	2.84	4.27	5.71	5.67	5.28	5.38		4.67	2.70	20
10°-0° N	8°07'	113°55'W	4035	1.53	2.50	3.62	4.94	5.82	5.95			4.11	1.66	19
	6°	173° W	5606	1.96	2.98	4.14	5.27	5.28	5.28	5.20		4.56	2.56	20
	4°51'	124°05'W	4336	1.77	2.90	4.22	5.36	6.43	6.78			4.74	2.06	46
	4°34'	179°00'E	5721	1.78	2.87	4.30	5.14	5.55	5.17	4.83		4.54	2.60	20
	2°	101° W	4270	1.62	2.33	3.97	4.63	5.35	5.80			4.10	1.75	8
0°-10° S	0°00'	178°59'E	5406	1.61	2.76	4.49	5.53	5.64	5.00	4.62		4.57	2.47	20
	4°34'	178°57'E	5500	1.59	2.44	3.95	5.02	5.30	4.67	4.02		4.17	2.30	20
	4°38'	125°08'W	4511	1.51	2.61	3.95	4.88	5.65	5.85			4.33	1.95	46
	5°01'	114°04'W	4162	1.72	2.09	3.51	4.59	5.21	5.71			3.88	1.62	19
	6°30'	107°24'W	3105	1.67	2.56	3.43	4.89	5.15				3.52	1.09	19
10°-20° S	10°00'	90°09'W	4128	1.27	2.42	3.01	4.18	4.65	5.12			3.48	1.44	19
	10°	170° W	5180	1.53	2.62	3.94	4.53	5.05	4.14	3.65		3.94	2.04	20
	12°00'	119°43'W	3660	1.60	2.62	3.67	4.87	5.09				3.83	1.40	15
	13°36'	112°19'W	3041	1.60	2.51	3.42	4.69	5.04				3.41	1.04	15
	14°00'	103°01'W	4189	1.29	2.49	3.68	4.76	5.26	5.42			3.97	1.66	15
	14°00'	134°04'W	4430	1.44	2.47	3.52	4.56	5.01	5.11			3.86	1.71	15
	15°59'	89°57'W	4496	1.36	2.39	3.65	4.46	4.96	5.36			3.92	1.76	15
	16°40'	167°05'W	5714	1.33	2.52	3.72	4.66	4.30	3.87	3.87		3.72	2.12	20
	17°58'	172°01'W	5850	1.17	1.88	3.42	4.09	3.85	3.26	3.11		3.21	1.88	20
20°-30° S	23°01'	146°05'W	5050	1.41	2.12	3.51	4.71	4.78	4.75	4.75		3.91	1.97	20
	23°39'	127°09'W	3677	1.40	2.05	3.65	4.32	4.93				3.55	1.30	46
	23°44'	153°37'W	4602	1.37	1.96	3.34	4.78	4.53	4.12			3.65	1.68	20
	23°59'	174°26'W	5999	1.29	2.02	3.43	4.41	4.17	3.82	3.86		3.56	2.13	20
	27°	157° W	5364	1.36	2.02	3.40	4.68	4.45	4.09	4.67		3.73	2.00	20
30°-40° S	32°50'	163°38'W	5618	1.16	1.94	3.31	4.61	4.16	3.74	3.64		3.49	1.96	20
	33°21'	128°24'W	4166	1.60	1.90	3.35	4.48	4.62	4.72			3.60	1.50	46
	38°23'	170°04'W	4839	1.40	1.87	3.21	4.13	3.42	3.16			3.11	1.50	20
40°-50° S	43°01'	129°57'W	4863	1.86	2.00	3.05	4.08	4.38	4.46			3.56	1.73	46
	44°59'	166°42'W	5340	1.53	2.00	2.98	3.43	3.59	3.60	3.69		3.11	1.66	45
50°-60° S	52°40'	178°05'W	5347	1.72	2.38	2.95	3.43	3.68	3.75	3.49		3.19	1.71	20
	57°35'	169°36'E	5300	2.45	2.83	2.97	3.41	3.63	3.56	3.40		3.25	1.72	20
	58°00'	174°00'W	5177	2.53	2.86	3.12	3.44	3.54	3.73	3.86		3.32	1.72	45
	59°21'	166°20'E	4707	2.10	2.60	2.66	3.02	3.45	3.41			2.95	1.39	12
60°-70° S	63°28'	151°02'E	3724	2.44	2.74	3.10	3.33	3.34				3.07	1.14	12
	64°52'	163°47'E	2974	2.88	3.26	3.42	3.33					3.27	0.97	12
	69°05'	173°30'W	4149	2.94	3.41	3.45	3.61	3.56	3.64			3.45	1.43	45

TABLE 4. ^{226}Ra PROFILES IN THE INDIAN OCEAN

Latitude band	Coordinates		Depth m	Mean ^{226}Ra concentration in layer /km/ Bq m^{-3}								$\sum{}^{226}$Ra Bq cm^{-2}	Reference
				0-0.5	0.5-1	1-2	2-3	3-4	4-5	>5	0-bottom		
1	2	3	4	5	6	7	8	9	10	11	12	13	14
20-10°N	19°43'N	64°37'E	3200	1.55	2.21	3.29	4.44	5.04			3.32	1.06	17,29
	13°21'	53°16'	2800	1.64	2.25	3.56	4.40				3.22	0.90	17,29
	12°59'	64°28'	4100	1.69	2.04	3.28	4.29	4.38	4.60		3.48	1.43	17,29
	12°30'	84°29'	3321	2.13	3.05	4.01	4.76	5.68			3.99	1.33	25,29
10-0°N	8°31'	86°03'	3659	2.06	2.92	3.79	4.67	5.62			4.00	1.47	25,29
	6°12'	64°24'	4700	1.45	2.51	3.42	4.06	4.20	4.43		3.57	1.68	17,29
	5°00'	79°57'	4196	2.01	2.90	3.96	4.84	5.59	5.62		4.28	1.80	25,29
	3°56'	56°50'	4650	1.65	2.48	3.44	4.37	4.39	4.56		3.71	1.72	17,29
	0°01'	80°03'	4651	1.86	2.91	3.87	4.65	5.18	5.57		4.24	1.97	25,29
0-10°S	0°04'S	50°53'E	5050	1.39	2.37	3.48	4.38	4.78	4.45	4.46	3.80	1.92	17,29
	1°12'	90°45'	4619	1.97	2.782	3.77	4.71	5.07	5.31		4.16	1.92	25,29
	5°00'	80°00'	5122	1.91	2.85	3.96	4.73	5.27	5.40	5.46	4.37	2.24	25,29
	5°02'	91°47'	4942	1.87	2.90	3.86	4.31	4.80	4.96		4.05	2.00	25,29
	6°09'	50°55'	4800	1.60	2.60	3.54	4.25	4.44	4.28		3.70	1.78	17,29
	9°22'	95°02'	5255	1.92	3.15	3.75	4.39	4.83	5.03	4.99	4.15	2.18	25,29
10-20°S	10°01'	79°59'	5347	2.07	3.16	3.96	4.63	5.04	5.24	5.36	4.37	2.34	25,29
	12°17'	54°39'	4650	1.57	2.47	3.39	3.98	4.12	3.87		3.45	1.60	17,29
	13°02'	97°08'		1.87	3.31	3.98							29
	14°59'	79°58'	5016	1.72	2.72	3.98	4.25	4.81	5.36	5.38	4.13	2.07	25,29
	17°18'	55°51'	4550	1:40	2.42	3.56	4.33	4.35	4.25		3.62	1.65	17,29
	19°29'	101°18'	5842	1.66	2.82	3.65	4.13	4.47	4.83	4.72	3.99	2.33	25,29
20-30°S	20°06'	79°59'	4800	1.52	2.14	3.49	4.00	4.36	4.93		3.72	1.79	17,25,29
	23°00'	74°01'	4150	1.24	1.93	3.35	3.71	4.67	4.73		3.38	1.40	17,25,29
	24°28'	104°55'		1.50	2.46	3.27							29
	27°00'	67°06'	5000	1.22	1.59	2.84	4.05	4.05	4.03		3.27	1.64	17,25,29
	27°04'	56°58'	5100	1.38	1.62	2.87	3.74	3.94	3.76	3.90	3.18	1.62	17,29
	29°15'	109°58'	5572	1.73	2.40	3.40	3.94	4.48	4.71	4.42	3.79	2.11	25,29
30-40°S	37°48'	57°35'	5400	1.44	1.91	2.76	3.23	3.62	3.69	3.60	3.04	1.64	17,29
	38°10'	128°04'	5774	1.66	1.82	2.77	3.54	3.67	3.89	3.98	3.24	1.87	48
	39°57'	109°58'	4650	1.76	2.04	2.89	3.25	3.81	3.88		3.09	1.44	29
40-50°S	47°38'	128°04'	3970	1.68	1.98	2.72	3.27	3.48			2.82	1.12	48
	47°40'	57°52'	4600	2.44	2.75	2.83	3.07	3.55	3.77		3.11	1.43	17,29
50-60°S	53°00'	103°01'	3780	3.04	3.28	3.22	3.62	3.62			3.39	1.28	29
	56°02'	128°09'	4586	2.90	3.05	3.23	3.52	3.59	3.62		3.36	1.54	48
	57°59'	125°35'	4700	2.97	3.32	3.25	3.68	3.67			3.38	1.59	48
	59°59'	60°59'	4730	3.29	3.26	3.47	3.64	3.44	3.70		3.50	1.65	16
	59°19'	92°38'	4450	3.09	3.19	3.46	3.62	3.64	3.71		3.49	1.55	29
60-70°S	64°04'	138°12'	3430	2.91	3.04	3.21	3.42	3.52			3.24	1.11	48
	64°11'	83°59'	3620	3.29	3.42	3.77	3.76	3.59			3.62	1.31	16
	66°14'	142°36'	340	2.93							2.93	0.09	48

TABLE 5. MEAN CONCENTRATIONS OF ^{226}Ra IN DIFFERENT WATER LAYERS FOR SINGLE 10° LATITUDE BANDS

Latitude band	n*	^{226}Ra concentration in layer /km/ Bq m^{-3}								$\sum {}^{226}$Ra Bq cm^{-2}
		0-0.5	0.5-1	1-2	2-3	3-4	4-5	>5	0-bottom	
					ATLANTIC OCEAN					
90-80°N	1	1.77	1.34	1.66					1.61	0.289
80-70	1	1.29	1.55	1.42	1.46	1.46			1.44	0.533
70-60	1	1.41	1.36	1.58	1.79				1.54	0.370
60-50	0	1.40	1.43	1.69	1.99				1.92	0.77
50-40	2	1.40	1.60	1.80	2.20±0.21	2.80±0.09	3.14±0.05		2.30±0.03	1.17±0.14
40-30	7	1.37±0.11	1.59±0.07	1.71±0.10	2.00±0.21	2.27±0.37	2.47±0.38	3.21±0.02	2.04±0.24	0.91±0.18
30-20	8	1.39±0.06	1.62±0.06	1.88±0.12	2.23±0.12	2.63±0.22	2.83±0.20	3.08±0.18	2.26±0.11	1.19±0.16
20-10	6	1.47±0.08	1.99±0.16	1.90±0.11	2.17±0.09	2.57±0.29	3.01±0.36	2.97	2.08±0.18	0.96±0.13
10-0	5	1.51±0.03	2.07±0.07	1.87±0.03	2.21±0.14	2.52±0.12	2.94±0.35	3.19±0.41	2.28±0.16	1.15±0.15
Average N	9	1.45±0.14	1.62±0.26	1.72±0.16	2.01±0.27	2.38±0.48	2.88±0.25	3.11±0.11	1.94±0.33	0.82±0.35
0-10°S	2	1.55	1.64	1.89±0.19	2.16±0.19	2.44±0.19	3.64±0.13	3.95±0.37	2.39±0.22	1.24±0.17
10-20	2	1.58±0.08	2.30±0.03	2.18±0.26	2.46±0.27	2.72±0.66	3.35±0.24	3.40±0.40	2.57±0.23	1.37±0.42
20-30	3	1.44±0.19	2.21±0.18	2.30±0.23	2.39±0.28	2.93±0.28	3.87±0.56		2.54±0.30	1.15±0.17
30-40	1	1.37	1.93	2.58	2.31	2.91	3.98		2.51	1.11
40-50	3	2.20±0.57	2.86±0.20	3.03±0.10	3.24±0.31	3.46±0.21	3.79±0.14	3.98	3.23±0.18	1.62±0.32
50-60	3	2.74±0.23	3.15±0.30	3.44±0.13	3.57±0.06	3.67±0.24	3.74±0.16	3.61	3.45±0.10	1.91±0.75
60-70	2	3.32±0.10	3.40±0.05	3.51±0.01	3.58	3.59	3.62±0.04	3.66	3.53±0.02	1.69±0.37
70-80	2	3.13±0.30	3.37±0.23	3.50±0.09	3.57	3.60			3.26±0.34	0.80±0.64
Average S	8	2.17±0.80	2.61±0.68	2.80±0.65	2.91±0.64	3.16±0.47	3.71±0.20	3.72±0.24	2.93±0.47	1.36±0.36
Overall	17	1.78±0.65	2.08±0.70	2.23±0.71	2.46±0.66	2.83±0.61	3.36±0.48	3.45±0.37	2.41±0.65	1.07±0.44
					PACIFIC OCEAN					
50-50°N	6	1.85±0.56	3.06±0.50	4.13±0.38	5.52±0.32	6.50±0.73	5.39	5.60	4.40±0.68	1.85±1.00
50-40	3	1.91±0.37	3.17±0.55	4.23±0.42	5.24±0.17	5.62±0.74	5.83±0.45	5.74±0.28	4.78±0.08	2.65±0.22
40-30	10	1.47±0.21	2.78±0.37	4.26±0.21	5.27±0.25	5.69±0.38	6.13±0.55	6.03±0.59	4.86±0.23	2.86±0.69
30-20	5	1.50±0.28	2.74±0.31	4.30±0.18	5.42±0.46	5.95±0.44	6.20±0.56	6.22±0.74	4.86±0.31	2.60±0.42
20-10	5	1.62±0.20	2.95±0.09	4.28±0.22	5.53±0.11	6.25±0.50	6.10±0.92	5.37±0.45	4.84±0.22	2.52±0.27
10-0	5	1.73±0.17	2.72±0.28	4.05±0.27	5.07±0.29	5.69±0.47	5.80±0.64	5.02	4.41±0.29	2.13±0.44
Average N	6	1.68±0.18	2.90±0.19	4.21±0.10	5.34±0.18	5.92±0.40	5.91±0.30	5.66±0.44	4.69±0.22	2.44±0.37
0-10°S	5	1.62±0.08	2.49±0.25	3.87±0.42	4.97±0.35	5.39±0.24	5.31±0.56	4.32	4.09±0.41	1.89±0.55
10-20	9	1.38±0.18	2.44±0.22	3.56±0.26	4.53±0.26	4.80±0.46	4.61±0.85	3.54±0.39	3.70±0.27	1.67±0.34
20-30	5	1.37±0.05	2.03±0.06	3.47±0.12	4.58±0.20	4.57±0.30	4.20±0.39	4.43±0.49	3.68±0.15	1.82±0.33
30-40	3	1.39±0.22	1.90±0.04	3.29±0.07	4.41±0.25	4.07±0.61	3.87±0.79	3.64	3.40±0.26	1.65±0.27
40-50	2	1.70±0.29	2.00±0.00	3.02±0.06	3.76±0.58	3.99±0.70	4.03±0.76	3.69	3.34±0.40	1.70±0.08
50-60	4	2.20±0.37	2.67±0.22	2.93±0.19	3.33±0.20	3.58±0.10	3.61±0.16	3.58	3.18±0.16	1.64±0.16
60-70	3**	2.75±0.27	3.14±0.35	3.32±0.19	3.42±0.16	3.45	3.64		3.26±0.19	1.18±0.23
Average S	6	1.61±0.32	2.26±0.32	3.36±0.35	4.26±0.60	4.40±0.65	4.27±0.61	3.87±0.40	3.57±0.33	1.73±0.10
**not included in the average										
Whole Pacific	12	1.64±0.25	2.58±0.42	3.78±0.51	4.80±0.70	5.16±0.94	5.09±0.97	4.77±1.02	4.13±0.65	2.08±0.45
					INDIAN OCEAN					
20-10°N	4	1.75±0.26	2.39±0.45	3.54±0.34	4.47±0.20	5.03±0.65	4.60		3.50±0.34	1.18±0.24
10-0	5	1.21±0.26	2.74±0.23	3.70±0.25	4.52±0.31	5.00±0.67	5.05±0.64		3.96±0.32	1.72±0.18
Average N	2	1.78±0.05	2.57±0.31	3.62±0.14	4.50±0.05	5.02±0.03	4.83±0.40		3.73±0.41	1.45±0.48
0-10°S	6	1.78±0.23	2.78±0.27	3.73±0.18	4.46±0.21	4.87±0.28	4.91±0.45	4.97±0.50	4.04±0.25	2.01±0.17
10-20	6	1.72±0.23	2.82±0.36	3.75±0.26	4.26±0.24	4.50±0.37	4.71±0.64	5.15±0.38	3.91±0.37	2.00±0.36
20-30	6	1.43±0.19	2.02±0.38	3.20±0.28	3.89±0.15	4.30±0.30	4.43±0.51	4.16±0.46	3.47±0.27	1.71±0.26
30-40	3	1.62±0.16	1.92±0.11	2.81±0.07	3.34±0.17	3.70±0.10	3.82±0.11	3.79±0.34	3.12±0.10	1.65±0.22
40-50	2	2.06±0.67	2.36±0.68	2.78±0.10	3.17±0.18	3.52±0.06	3.77		2.97±0.26	1.28±0.28
50-60	5	3.06±0.15	3.22±0.11	3.33±0.13	3.62±0.08	3.59±0.09	3.68±0.05		3.42±0.07	1.60±0.06
60-70	3	3.04±0.21	3.23±0.34	3.49±0.50	3.59±0.30	3.55±0.06	3.66±0.07		3.43±0.34	1.21±0.18
Average S	7	2.10±0.67	2.62±0.53	3.30±0.40	3.76±0.47	4.01±0.56	4.14±0.53	4.52±0.65	3.48±0.39	1.64±0.31
Whole ocean	9	2.03±0.60	2.61±0.47	3.37±0.37	3.92±0.52	4.24±0.65	4.29±0.56	4.52±0.65	3.54±0.37	1.60±0.31

* – number of integrated profiles

with their respective standard deviations. When no value existed for a given strip a mean of the neighboring values was used. The sum of subinventories in each water layer yields a total inventory which is very similar to the one determined by the first method /Table 6 line a// but is loaded with a smaller error /the error is expressed as a square root of the sum of variances for each subinventory/.

TABLE 6. ^{226}Ra INVENTORIES IN THE WORLD OCEANS

Water layer km	ATLANTIC			PACIFIC			INDIAN		
	Volume $10^{15}m^3$	Mean ^{226}Ra Bq m^{-3}±16	Inventory 10^{15}Bq±16	Volume $10^{15}m^3$	Mean ^{226}Ra Bq m^{-3}±16	Inventory 10^{15}Bq±16	Volume $10^{15}m^3$	Mean ^{226}Ra Bq m^{-3}±16	Inventory 10^{15}Bq±16
0-0.5	46.2	1.78±0.65	82±30	83.1	1.64±0.25	137±21	35.4	2.03±0.60	72±21
0.5-1	41.7	2.08±0.70	87±29	77.4	2.58±0.42	200±33	33.6	2.61±0.47	88±16
1-2	81.8	2.23±0.71	182±58	156.0	3.78±0.51	590±80	66.0	3.37±0.37	222±24
2-3	72.6	2.46±0.66	179±48	142.3	4.80±0.70	683±100	62.5	3.92±0.52	245±33
3-4	58.9	2.83±0.61	167±36	127.9	5.16±0.94	660±120	49.9	4.24±0.65	212±32
4-5	35.5	3.36±0.48	119±17	78.7	5.09±0.97	401±76	28.4	4.29±0.56	122±16
5-6	10.0	3.45±0.37	35±4	24.6	4.77±1.02	117±25	6.8	4.52±0.65	31±4
6				17.1	5.69±0.12	97±2			
Total a/	346.7		851±95	707.1		2884±197	282.6		991±61
b/			841±70			2934±145			1016±53
c/			839±196			2941±476			1020±120
d/						2923±56			

Method of calculation

a/ Summation of subinventories for the horizontal water layers shown in the table

b/ As above, except that the upper 0-2 km layer was divided in 10° strips by latitude and the radium content of each strip was calculated separately and then summed up

c/ Arithmetic mean of the weighted mean radium concentrations in profiles multiplied by the total volume of the ocean /$\bar{C}_{Tot.} \cdot V_{Tot.}$/

d/ Summation of subinventories for each 10° latitude strips /$\Sigma\, V_i \cdot \bar{C}_i$/ - Table 7

Further refinement can be made by using the information on volumes of the water in each latitude strip and the respective weighted mean ^{226}Ra concentrations. Such an approach has been possible for the Pacific Ocean for which the partial volumes by latitude were known /cf.par.2/. The results of calculations are shown in Table 7 and the total inventory /Table 6 line d/ is in a good agreement with the other estimates.

For the remaining two oceans a simplified method was applied in which only the upper 0-2km water layer was divided in latitude bands and the volumes of each band was determined by multiplying the area of each strip by the tickness of the layer. The areas were taken from Ref.2 and were normalized to a 1km isobath by consulting the bathymetric charts near the continents for each strip.

Because of the distinct features of the North Polar Ocean it was taken apart from the Atlantic Ocean and the inventory was calculated separately for that basin. The detailed calculations are not shown here but the final results are included in Table 6 line b/.

TABLE 7. ^{226}Ra INVENTORIES IN THE PACIFIC OCEAN

Latitude band	Volume $10^{15} m^3$	^{226}Ra Bq m^{-3}	n	Inventory 10^{15} Bq
50°N	25.2	4.40±0.68	6	111±17
50-40	36.9	4.78±0.08	3	176±3
40-30	58.9	4.86±0.23	9	286±13
30-20	70.4	4.86±0.31	5	342±22
20-10	76.7	4.84±0.22	5	371±17
10-0	78.9	4.41±0.29	5	348±23
0-10°S	64.9	4.09±0.41	5	265±26
10-20	61.9	3.70±0.27	9	229±17
20-30	59.2	3.68±0.15	5	218±9
30-40	54.8	3.40±0.26	3	186±14
40-50	46.2	3.34±0.18	2	154±10
50-60	40.2	3.18±0.16	4	128±7
60-70	32.9	3.26±0.19	3	107±6
TOTAL	707.1		64	2923±56

n - number of profiles

It can be concluded from the above data that all methods of calculation applied here yield results which can be regarded as identical within the stated uncertainties.

The errors associated with analytical procedures and with the methods used for integrating ^{226}Ra profiles are largely of random character and as such should not contribute significantly to the overall error because of the mutual cancellation effect. Contributing are systematic errors due to the uncertainty on ^{226}Ra standard /~ 1%/ and on ocean volumes /~ 2.5%/ which are here treated additively. The recommended inventories are thus as follows:

Ocean	^{226}Ra inventory x 10^{18} Bq $\pm 1\sigma$
- Atlantic	0.84+0.10
- Pacific	2.92±0.16
- Indian	1.02±0.09
Total	4.78±0.27

6. RADIUM OUTPUT

Radium is lost from sea water by radioactive decay at a rate of $4.29 \times 10^{-4} yr^{-1}$. The losses from individual oceans are as follows:

	Unit	Atlantic	Pacific	Indian
Total loss	10^{12}Bq	360	1250	436
Per area	Bq $m^{-2}yr^{-1}$	3.38	7.02	5.72
Per volume	10^{3}Bq m^{-3}	1.04	1.77	1.54

Other losses are negligible or difficult to estimate quantitatively. Removal to bottom sediments with sinking particles may be important in certain regions /14,52,80,98/. Fluxes of particulate radium were reported to be as high as 4.3 Bq $m^{-2}yr^{-1}$ on a global scale /92/ but most of the ^{226}Ra redissolves in deep waters. Biological processes thus appear to modify the distribution pattern of radium in a water column without changing significantly its inventory.

Radium loss from the sea surface due to the action of wind can be assessed using the figures for total salt losses quoted in /97/ and the average concentration of ^{226}Ra in surface waters /1.4 Bq m^{-3}/. The estimated loss is of 10^{10}Bq yr^{-1} and is trivial compared to the radioactive decay.

There are some losses to the sediments by adsorption and ion-exchange but these processes are largely obscured by a desorptive ^{226}Ra flux from the decaying ^{230}Th atoms. Manganese-rich slowly accumulating sediments of the North Pacific were demonstrated to uptake radium at a rate of 1 to 2 Bq $m^{-2}yr^{-1}$ /100/.

7. RADIUM INPUT

7.1. Flux from the bottom

The principal source of oceanic radium are bottom sediments, and more precisely - the ^{230}Th which is constantly generated in seawater from the decaying ^{234}U and is rapidly transferred to the bottom.

An average oceanic water /S=34.7‰/ contains 41.0 Bq m^{-3} of ^{238}U and 46.8 Bq m^{-3} of ^{234}U /47/. This is equivalent to the 1.48, 3.02 and 1.21×10^{14}Bq of ^{230}Th produced annually in the Atlantic, Pacific and Indian Oceans, respectively. The corresponding fluxes to the bottom are 3.38, 7.02 and 5.72 Bq $m^{-2}yr^{-1}$. The areal distribution of ^{230}Th in bottom sediments is uneven depending on the water depth and sedimentation rate /23,24,91/. About 70±20 percent of the ^{226}Ra produced in sediments returns to the bottom water. More slowly accumulating sediments have higher ^{230}Th content and hence higher production rates of ^{226}Ra at the sediment-wator interface and higher radium fluxes.

There are relatively few data available on ^{226}Ra fluxes from the seafloor. They were calculated either from ^{222}Ra profiles in pore waters /23/ or from excess ^{230}Th inventory in sediment columns. They are presented below:

Ocean	Coordinates		Depth km	n	^{226}Ra flux Bq m^{-2} yr^{-1}	Ref.
Pacific	40°-38°N	150°E-170°W	5.4-5.7	4	1.5-13.5	101
Pacific	10°-11°N	178°E-180°W	5.4-5.9	3	42-68	99
Pacific	27°-35°N	138°-149°E	3.8-6.1	2	3.7-6.2	100
Pacific	9°-15°N	125°-139°W	4.6-5.1	3	14-35	23
Atlantic	17°-37°N	23°-33°W	2.7-3.4	2	0.3-0.4	23
Indian	14°S	78°E	5.2	1	5.8	23
Indian	26°S	74°E	4.1	1	4.3	23
Indian	39°S	120°E	4.9	1	4.3	23
Indian	53°S	36°E	4.3	1	1.8	23
Indian	67°S	30°E	4.1	1	3.2	23

It can be noticed that the calculated fluxes range over two orders of magnitude. This large scatter demonstrates clearly the difficulty in deriving representative averages for any ocean. What can be only concluded from the available data is that the observed fluxes are large enough to balance the ^{226}Ra losses occuring in seawater by radioactive decay.

7.2. Discharge with water

Rivers have long be considered to be of no importance as a source of ^{226}Ra to the oceans /28,54,76/. However, the information on ^{226}Ra concentrations in major World rivers is too scanty to prove it. The reported concentrations of dissolved radium range from 0.2 to 87 Bq m^{-3}, i.e. over two orders of magnitude /30/, Seasonal and long-term changes have not been studied systematically and choosing an average ^{226}Ra concentration is somewhat arbitrary and is fraught with a large uncertainty. In order to assess approximatively the magnitude of riverine radium discharges a review was made of the available literature data /28,30,39,41,54,55,76/ to choose probable values. Desorptive fluxes from suspended mater were taken into consideration /39,50,53,55/. Values on water discharges and suspended load carried to the oceans was taken from /31/.

The results are as follows:

	Atlantic	Pacific	Indian
Water discharge $10^{12}m^3yr^{-1}$	25.0	14.1	5.64
Suspended load $10^{12}kg\ yr^{-1}$	3.19	7.15	8.19
Suspended load kg m^{-3}	0.128	0.507	1.452
^{226}Ra dissolved Bq m^{-3}	1±0.5	2±1	3±2
^{226}Ra desorbed Bq kg^{-1} susp.	15±5	20±10	20±10
<u>Total riverine input:</u>			
/dissolved + desorbed/ $10^{15}Bq\ yr^{-1}$	0.07±0.02	0.17±0.08	0.18±0.08
/dissolved + desorbed/ Bq $m^{-2}yr^{-1}$	0.7±0.2	0.9±0.5	2.3±1.1

Apart from surface water there is an appreciable discharge of ground waters into the oceans. They were estimated at 0.98, 0.7 and 0.51×10^{12} m^3yr^{-1} for the Atlantic, Pacific and Indian Oceans, respectively /93/. In choosing an average ^{226}Ra concentration for these waters we meet with the same difficulties as for the riverine ^{226}Ra concentrations. The reported data range from 10^{-3} to 10^2 Bq m^{-3}, five orders of magnitude /94/. If a narrower range is selected, e.g. 10–100 Bq m^{-3} we arrive at the ground water ^{226}Ra supply of 1 to 10, 1 to 7 and 1 to 5×10^{13} Bq yr^{-1} with the corresponding fluxes of 0.1 to 0.9, 0.04 to 0.4 and 0.07 to 0.6 Bq $m^{-2}yr^{-1}$ for the Atlantic, Pacific and Indian Oceans, respectively. Circulation of seawater through hydrothermal systems associated with ocean spreading centers can bring large quantities of ^{226}Ra to oceans. Concentrations as high as 700 Bq m^{-3} of ^{226}Ra were found in seawater which had percolated through hydrothermal vents in the Galapagos Rise area /87/. Additional quantities of radium can be supplied with volcanic effusives /2/. The significance of these sources in the oceanic ^{226}Ra budget is hard to evaluate because they are not sufficiently documented.

A general conclusion can be made that while the total inventory of ^{226}Ra in the oceans rests on a solid data base the sources and sinks cannot,at present,be estimated with a comparable accuracy from available data . The uncertainties are too large to unveal any tendency of departure from a steady state due to man's activity on a global scale.

REFERENCES

1. BACON,M.P., SPENCER,D.W., BREWER,P.G., "^{210}Pb/^{226}Ra and ^{210}Po/^{210}Pb disequilibria in seawater and suspended particulatematter", Earth Planet.Sci.Lett., 32/1976/ 277-296.
2. BAUMGARTNER,A., REICHEL,E., The World Water Balance, Elsevier, Amsterdam-Oxford-New York 1975, 195 p.
3. BENNETT,J.T., KRISHNASWAMI,S., TUREKIAN,K.K., MELSON,W.G., HOPSON,C.A., "The uranium and thorium decay radionuclides in Mt.St.Helens effusives", Earth Planet.Sci.Lett., 60 /1982/ 61-69.
4. BLOCH,S., "Some factors controlling the concentration of uranium in the world ocean", Geochim.Cosmochim.Acta, 44 /1980/ 373-377.
5. BROECKER,W.S., PENG,T.H., "The distribution of radon in the BOMEX area", Earth Planet.Sci.Lett. 11/1971/ 99-108.
6. BROECKER,W.S., GODDARD,J., SARMIENTO,J.L., "The distribution of ^{226}Ra in the Atlantic Ocean", Earth Planet.Sci.Lett., 32/1976/ 220-235.
7. BROECKER,W.S., KAUFMAN,A., TRIER,R.M., "The residence time of thorium in surface sea water and its implications regarding the rate of reactive pollutants", Earth Planet. Sci.Lett. 20/1973/ 35-44.
8. BROECKER,W.S., LI,Y.H., CROMWELL,J., "Radium-226 and radon-222: concentration in Atlantic and Pacific Oceans", Science 158/1967/ 1307-1310.
9. BROECKER,W.S., KAUFMAN,A., KU,T.L., CHUNG,Y.C., CRAIG,H., "Radium-226 measurements from the 1969 North Pacific GEOSECS station", J.Geophys.Res., 75/1970/ 7682-7685.
10. CHAN,L.H., CHUNG,Y., "Barium and radium in the Dead Sea", Earth Planet.Sci.Lett., 85/1987/ 41-53.
11. CHAN,L.M., EDMOND,J.M., STALLARD,R.F., BROECKER,W.S., CHUNG,Y.C., WEISS,R.F., KU,T.L., "Radium and barium at GEOSECS stations in the Atlantic and Pacific", Earth Planet. Sci.Lett., 32/1976/ 258-267.

12. CHUNG,Y.C., "Radium-226 and Ra-Ba relationships in Antarctic and Pacific waters", Earth Planet.Sci.Lett., 23/1974/ 125-135.

13. CHUNG,Y., "^{226}Ra in the Pacific Ocean", Earth Planet.Sci. Lett., 49/1980a/ 267-292.

14. CHUNG,Y., "Radium-barium-silica correlations and a two-dimensional radium model for the world ocean", Earth Planet.Sci.Lett., 49/1980b/ 309-318.

15. CHUNG,Y., "A ^{226}Ra section across the East Pacific Rise", Earth Planet.Sci.Lett., 49/1980c/ 319-328.

16. CHUNG,Y., "^{210}Pb and ^{226}Ra distribution in the Circumpolar Waters", Earth Planet.Sci.Lett., 55/1981/ 205-216.

17. CHUNG,Y., "^{226}Ra in the western Indian Ocean", Earth Planet. Sci.Lett., 85 /1987/ 11-27.

18. CHUNG,Y., APPELQUIST,M.D., "^{226}Ra and ^{210}Pb in the Weddell Sea", Earth Planet.Sci.Lett., 49/1980/ 401-410.

19. CHUNG,Y.C., CRAIG,H., "Radium-226 in the eastern Equatorial Pacific", Earth Planet.Sci.Lett., 17/1973/ 306-318.

20. CHUNG,Y., CRAIG,H., "^{226}Ra in the Pacific Ocean", Earth Planet.Sci.Lett., 49/1980/ 267-292.

21. CHUNG,Y., FINKEL,R.C., KIM,K., "^{226}Ra, ^{210}Pb and ^{210}Po in the Red Sea", Earth Planet.Sci.Lett., 58/1982/ 213-224.

22. CHUNG,Y., CRAIG,H., KU,T.L., GODDARD,J., BROECKER,W.S., "Radium-226 measurements from three GEOSECS intercalibration stations", Earth Planet.Sci.Lett., 23/1974/ 116-124.

23. COCHRAN,J.K., "The flux of ^{226}Ra from deep-sea sediments", Earth Planet.Sci.Lett., 49/1980/ 381-392.

24. COCHRAN,J.K., KRISHNASWAMI,S., "Radium, thorium, uranium and ^{210}Pb in deep-sea sediments and sediment pore waters from the North Equatorial Pacific", Amer.J.Sci., 280/1980/ 849-889.

25. COCHRAN,J.K., BACON,M.P., KRISHNASWAMI,S., TUREKIAN,K.K., "^{210}Po and ^{210}Pb distributions in the central and eastern Indian Ocean", Earth Planet.Sci.Lett. 65/1983/ 433-452.

26. EVANS,R.D., KIP,A.F., MOBERG,E.G., "The radium and radon content of Pacific ocean water, life and sediments", Amer. J.Sci., 36/1938/ 241-259.

27. FEELY,H.W., KIPPHUT,G.W., TRIER,R.M., KENT,C., "^{228}Ra and ^{228}Th in coastal waters", Estuarine coast.Mar.Sci., 2/1980/ 179-205.

28. FØYN,E., KARLIK,B., PETTERSSON,H., RONA,E., "The radioactivity of seawater", Göteborgs Kungl.Vetenskaps.och Vitterhets.Samhälles Handlingar Ser.B /6/ No 12 /1939/ 3-44.

29. GEOSECS Atlas, International Decade of Ocean Exploration, NSF, vol.7 /1987/, U.S.Govt.Printing Office, Washington, D.C.

30. GMELIN Handbuch der Anorganischen Chemie, 31. RADIUM, Main Series, 8th Ed., Suppl. vol.1, Springer Verlag, Berlin, Heidelberg, New York, 1977.

31. GORDEEV,V.V., Retshnoyi Stok v Okean i Tsherty yiegho Geokhimiyi, Izd.Nauka, Moskva 1983, 160 p.

32. GORSHKOV,S.G./ed./, The Atlas of Oceans . I.Pacific ocean /1974/, II.Atlantic and Indian Oceans /1977/, Ministry of Defence, Dept. of Navigation and Oceanography, USSR.

33. GRASHCHENKO,S.M., NIKOLAYEV,D.S., KOLYADIN,L.B., KUZNETSOV, YU.V., LAZAREV,K.F., "Radium concentration in the waters of the Black Sea", Dokl.Akad.Nauk SSSR, 123/1960/ 1171-1172.

34. HANOR,J.S., CHAN,L.H., "Non conservative behavior of barium during mixing of Mississippi River and Gulf of Mexico waters", Earth Planet.Sci.Lett., 37/1977/ 242-250.

35. HARADA,K.,TSUNOGAI,S., "^{226}Ra in the Japan Sea and the residence time of the Japan Sea water", Earth Planet.Sci. Lett., 77/1986/ 236-244.

36. KADKO,D., "A detailed study of some uranium series nuclides at an abyssal hill area near the East Pacific Rise at 8°45'N", Earth Planet.Sci.Lett., 51/1980/ 115-131.

37. KAUFMAN,A., LI,Y.H., TUREKIAN,K.K.,"The removal rates of ^{234}Th and ^{228}Th from waters of the New York Bight", Earth Planet Sci.Lett., 54/1981/ 385-392.

38. KAUFMAN,A., TRIER,R.M., BROECKER,W.S., FEELY,H.W., "Distribution of ^{228}Ra in the World Ocean", J.Geophys.Res., 78/1973/ 8827-8848.

39. KEY,R.M., STALLARD,R.F., MOORE,W.S., SARMIENTO,J.L., "Distribution and flux of ^{226}Ra and ^{228}Ra in the Amazon River Estuary", J.Geophys.Res., 90/C4/ /1986/ 6995-7004.

40. KNAUSS,K.G., KU,T.L., MOORE,W.S., "Radium and thorium isotopes in the surface waters of the East Pacific and coastal Southern California", Earth Planet.Sci.Lett., 39/1978/235-249.

41. KOCZY,F.F., TOMIC,E., HECHT,F., "Zur Geochemie des Urans im Ostseebecken", Geochim.Cosmochim.Acta, 11 /1957/ 86-102.

42. KORT,V.G., "Vodoobmen mezdhu okeanami", Okeanologiya 11/1962/ 593-598.

43. KRISHNASWAMI,S., LAL,D., SOMAYAJULU,B.L.K., "Investigations of gram quantities of Atlantic and Pacific surface particulates", Earth Planet.Sci.Lett., 32/1976/ 403-419.

44. KRISHNASWAMI,S., SOMAYAJULU,B.L.K., CHUNG,Y., "^{210}Pb/^{226}Ra disequilibrium in the Santa Barbara Basin", Earth Planet. Sci.Lett.,27/1975/ 388-392.

45. KU,T.L., LIN,M.C.,"^{226}Ra distribution in the Antarctic Ocean", Earth Planet.Sci.Lett., 32/1976/ 236-248.

46. KU,T.L., HUH,C.A., CHEN,P.S., "Meridional distribution of ^{226}Ra in the Eastern Pacific along GEOSECS cruise tracks", Earth Planet.Sci.Lett., 49/1980/ 293-308.

47. KU,T.L., KNAUSS,K.G., MATHIEU,G.G., "Uranium in open ocean: concentration and isotopic composition", Deep-Sea Res., 24 /1977/ 1005-1017.

48. KU,T.L., LI,Y.H., MATHIEU,G.G., WONG,H.K., "Radium in the Indian-Antarctic Ocean south of Australia", J.Geophys.Res., 75/1970/ 5286-5292.

49. LEVY,D., MOORE,W.S., "^{224}Ra in continental shelf waters", Earth Planet.Sci.Lett., 73/1985/ 226-230.

50. LI,Y.H., CHAN,L.H., "Desorption of Ba and ^{226}Ra from riverborne sediments in the Hudson estuary", Earth Planet.Sci. Lett., 43/1979/ 343-350.

51. LI,Y.H., FEELY,H.W., SANTSCHI,P.H., "^{228}Th-^{228}Ra radioactive disequilibrium in the New York Bight and its implications for coastal pollution", Earth Planet.Sci. Lett., 42/1979/ 13-26.

52. LI,Y.H., KU,T.L., MATHIEU,G.G., WOLGEMUTH,K., "Barium in the Antrctic Ocean and implications regarding the marine geochemistry of Ba and ^{226}Ra", Earth Planet.Sci.Lett., 19/1973/ 352-358.

53. LI,Y.H., MATHIEU,G., BISCAYE,P., SIMPSON,H.J., "The flux of ^{226}Ra from estuarine and continental shelf sediments", Earth Planet.Sci.Lett., 37/1977/ 237-241.

54. MIYAKE,Y., SUGIMURA,Y., TSUBOTA,H., "Content of uranium, radium and thorium in river waters in Japan", in: The Natural Radiation Environment /Adams,J.A. ed./, Univ. Chicago Press, Chicago /1964/ 219-225.

55. MOORE,D.G., SCOTT,M.R., "Behavior of ^{226}Ra in the Mississippi River mixing zone", J.Geophys.Res., 91/C12/ /1986/ 14317-14329.

56. MOORE,R.M., SMITH,J.N., "Disequilibria between ^{226}Ra, ^{210}Pb and ^{210}Po in the Arctic Ocean and the implications for chemical modification of the Pacific water inflow", Earth Planet.Sci.Lett., 77/1986/ 285-292.

57. MOORE,W.S., "Oceanic concentrations of radium-228", Earth Planet.Sci.Lett., 6/1969/ 437-426.

58. MOORE,W.S., "The measurement of ^{228}Ra and ^{228}Th in seawater", J.Geophys.Res., 74/1969/ 694-704.

59. MOORE,W.S., "Radium-228: application to thermocline mixing studies", Earth Planet.Sci.Lett., 16/1972/ 421-422.

60. MOORE,W.S., "Sampling ^{228}Ra in the deep ocean", Deep-Sea Res., 23/1976/ 647-651.

61. MOORE,W.S., "The thorium isotope content of ocean water", Earth Planet.Sci.Lett., 53/1981/ 419-426.

62. MOORE,W.S., "Radium-228 in the South Atlantic Bight", J.Geophys.Res., 92/C5/ /1987/ 5177-5190.

63. MOORE,W.S., SACKETT,W.M., "Uranium and thorium series inequilibrium in sea water", J.Geophys.Res., 69/1964/ 5401-5405.

64. MOORE,W.S., SANTSCHI,P.H., "Ra-228 in the deep Indian Ocean", Deep-Sea Res., 33/1986/ 107-120.

65. MOORE,W.S., BRULAND,K.W., MICHEL,J., "Fluxes of uranium and thorium series isotopes in the Santa Barbara Basin", Earth Planet.Sci.Lett., 53/1981/ 391-399.

66. MOORE,W.S., FEELY,H.W., LI,Y.H., "Radium isotopes in sub-arctic waters", Earth Planet.Sci.Lett., 49/1980/ 329-340.

67. MOORE,W.S., KEY,R.M., SARMIENTO,J.L., "Techniques for precise mapping of ^{226}Ra and ^{228}Ra in the ocean", J.Geophys.Res., 90/1985/ 6983-6994.

68. NOZAKI,Y., NAKANISHI,T., "^{231}Pa and ^{230}Th profiles in the open ocean water column", Deep-Sea Res., 32/1985/ 1209-1220.

69. OKUBO,T., "Radium-228 in the Japan Sea", J.Oceanogr.Soc. Japan, 36/1980/ 279-286.

70. OKUBO,T., "^{228}Ra in surface water of the Seto Inland Sea", J.Oceanogr.Soc.Japan, 37/1981/ 279-286.

71. OKUBO,T., "Radioactive disequilibrium of thorium series nuclides in surface waters of the Seto Inland Sea", J. Oceanogr.Soc.Japan, 38/1982/ 1-7.

72. OKUBO,T., FURUYAMA,K., SAKANOUE,M., "Distribution of ^{228}Ra in surface sea water of the East Indian Ocean", Geochem.J., 13/1979/ 201-206.

73. PETTERSSON,H., "Manganese nodules and oceanic radium",in: Papers in Marine Biology and Oceanography . Supplement to Deep-Sea Res.,3/1955/ 335-345.

74. REID,D.F., MOORE,W.S., SACKETT,W.M., "Temporal variation of Ra-228 in the near-surface Gulf of Mexico", Earth Planet.Sci. Lett., 43/1979/ 227-236.

75. RHEIN,M., CHAN,L.H., ROETHER,W., SCHLOSSER,P., "^{226}Ra and Ba in North-east Atlantic Deep Water", Deep-Sea Res., 34/1987/ 1541-1564.

76. RONA,E., URRY,W.D., "Radioactivity of ocean sediments,VII. Radium and uranium content of ocean and river waters", Am.J.Sci.,250/1952/ 241-262.

77. SAKANOUE,M., OKUBO,T., FURUYAMA,K., "^{228}Ra in sea water",in: Isotope Marine Chemistry /E.D.Goldberg, Y.Horibe and K. Saruhashi, eds./. Uchida Rokakuho Publ. Co.Ltd. Tokyo, Japan 1980, 247-258.

78. SANTSCHI,P.H., LI,Y.H., BELL,J., "Natural radionuclides in the water of Narragansett Bay", Earth Planet.Sci.Lett., 45 /1979/ 201-213.

79. SCHLOSSER,P., RHEIN,M., ROETHER,W., KROMER,B., "High-precision measurement of oceanic ^{226}Ra", Mar.Chem., 15/1984/ 203-216.

80. SHANNON,L.V., CHERRY,R.D., "Radium-226 in marine phytoplankton", Earth Planet.Sci.Lett., 11/1971/ 339-343.

81. SOMAYAJULU,B.L.K., RENGARIAN,R., "^{228}Ra in the Dead Sea", Earth Planet.Sci.Lett., 85/1987/ 54-58.

82. STILLER,M., CHUNG,Y.C., "Radium in the Dead Sea: A possible tracer for the duration of meromixis", Limnol.Oceanogr., 29 /1984/ 574-586.

83. STRUTT,R.J., "On the distribution of radium in the earth's crust", Proc.Roy.Soc., A78 /1906/ 150.

84. SZABO,B.J., "Radium content in plankton and seawater in the Bahamas", Geochim.Cosmochim.Acta, 31/1967/ 1321-1331.

85. TRIER,R.M., BROECKER,W.S., FEELY,H.W., "Radium-228 profile at the second GEOSECS intercalibration station, 1970, in the North Atlantic", Earth Planet.Sci.Lett., 16/1972/ 141-145.

86. TSUNOGAI,S., HARADA,K., "^{226}Ra and ^{210}Pb in the Western North Pacific" In: Isotope Marine Chemistry, E.D.Goldberg, Y.Horibe and K.Saruhashi, eds., pp. 165-191. Uchida Rokakuho, Tokyo, 1980.

87. TUREKIAN,K.K., COCHRAN,J.K., "Flow rates and reaction rates in the Galapagos Rise spreading center hydrotermal system as inferred from ^{228}Ra/^{226}Ra in vesicomyid clam shells", Proc.Natl.Acad.Sci.USA,83/1986/ 6241-6244.

88. UNESCO, World register of rivers discharging into the oceans, summary of main results. WORRI Project, October 1978.

89. VINOGRADOV,A.P., Vvedeniye v Geokhimiyu Okeana, Izd.Nauka, Moskva 1967, 213 p.

90. YAMADA,M., NOZAKI,Y., "Radium isotopes in coastal and open ocean surface waters of the Western North Pacific", Mar. Chem., 19 /1986/ 379-389.

91. YANG,H.S., NOZAKI,Y., SAKAI,H., MASUDA,A., "The distribution of ^{230}Th and ^{231}Pa in the deep-sea surface sediments of the Pacific Ocean", Geochim.Cosmochim.Acta, 50/1986/ 81-89.

92. BACON,M.P., EDMOND,J.M., "Barium at GEOSECS III in the South west Pacific", Earth Planet.Sci.Lett., 16/1972/ 66-74.

93. DJAMALOV,R.G., ZEKTSER,I.S., MASKNETELI,A.W., Ground Water Discharge into Seas and Oceans, M: Nauka, Moskva, 1977, 96p. /in russian/.

94. IYENGAR,M.A.R., "Distribution /of ^{226}Ra/ in nature", in: The Behaviour of Radium in Waterways and Aquifers, IAEA-TECDOC-301, Vienna, 1984, 57-86.

95. KOSSINA,E., Der Tiefen des Weltmeeres, Veröffentl.Inst.Meereskunde, N.F., No.9, E.S.Mittler und Sohn, Berlin, 1921, 70p.

96. MOORE,W.S., "Radium-228 in the South Atlantic Bight", J. Geophys.Res., 92/C5/ /1987/ 5177-5190.

97. STEPANOV,V.N., The World Ocean, Izd. Znaniye, Moskva, 1974, 225p.

98. TSUNOGAI,S., YAMADA,M., "^{226}Ra in Bering Sea sediments and its implication as a geochronometer", Geochem.J., 13/1979/ 231-238.

99. TSUNOGAI,S., YAMADA,M., MORITANI,T., "Radiochemical studies on the pelagic sediments collected in the east of Marshal Islands /GH78-1 area/, Cruise Rept.17, Geol.Surv.Jap., 1981, 246-250.

100. YAMADA,M., TSUNOGAI,S., Behaviour of radium in deep-sea sediments, 地球化学 15, 53-59 (1981)

101. YANG,H.S., NOZAKI,Y., SAKAI,H., NAGAYA,Y., NAKAMURA,K., "Material and man-made radionuclide distributions in North-west Pacific deep-sea sediments: rates of sedimentation, bioturbation and ^{226}Ra migration", Geochem.J., 20/1986/ 29-40.

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