Dispersal of Tritium in Southern Ocean Waters

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ABSTRACT. Depth-distribution of tritium in Southern Ocean waters is reported. Decrease of tritium in surface waters from c 2 TU at 38°S. to 40°S. latitude, to 1.3 TU and 1.1 TU at 45°S., 55°S., 60°S. and 64°S. latitudes, are very likely to be associated with the presence of convergence and divergence areas at subtropical and subantarctic latitudes in the Southern Ocean. A tritium-free layer has been found to exist at intermediate depths. Significant increase of tritium in waters at and below 4 km. strongly suggests a younger age, a minimum of the order of 25 years for the bottom waters circulating in the entire region to the south of South Australia.

RÉSUMÉ. Dispersion du tritrium dans les eaux de l'océan Austral. L'auteur rend compte de la distribution en profondeur du tritium dans les eaux de l'océan Austral. La diminution du tritium dans les eaux de surface, d'environ 2 TU de 38°S à 40°S de latitude, à 1.3 TU et 1.1 TU à 45°S, 55°S, 60°S et 64°S de latitude, est très probablement associée à la présence d'aires de convergence et de divergence aux latitudes subtropicales et subantarctiques dans l'océan Austral. Aux profondeurs intermédiaires, on trouve un niveau sans tritium. Une augmentation importante du tritium dans les eaux qui se trouvent à 4 km et plus de profondeur suggère fortement que ces eaux sont plus jeunes, au minimum dans l'ordre de 25 ans pour les eaux de fond qui circulent dans toute la région au sud de l'Australie méridionale.

РЕЗЮМЕ. Распространение трития в водах Южного океана. Приводятся данные о распределении трития по глубине в водах Южного океана. Отмечается падение содержания трития в поверхностных водах от примерно 2 Т.Е. в зоне 38-40° ю.ш. до 1,3 — 1,1 Т.Е. на 45°, 55°, 60° и 64° ю.ш., которое, по всей вероятности, связано с наличием участков конвергенции и дивергенции течений в субтропических и субантарктических широтах Южного океана. Установлено, что на промежуточных глубинах существует слой воды с нулевым содержанием трития. Повышенное содержание трития на глубине 4 км и более дает серьёзные основания полагать, что придонные воды, циркулирующие во всей области к югу от Южной Австралии, имеют молодой — порядка 25 лет — возраст.

INTRODUCTION

Since tritium was discovered in nature during the fifties (Faltings and Harteck 1950; Grosse et al. 1951), its importance in the study of the natural hydrologic cycle has become increasingly evident. A year-round program of monitoring tritium in precipitation samples is maintained today by the International Atomic Energy Agency (IAEA), Vienna. Systematic study of the distribution of tritium along north-south transects of the three major oceans has begun under the Geochemical Sections (GEOSECS) program of the International Decade of Ocean Exploration (IDOE). Results of tritium levels in ocean water have been published earlier by the Scripps Institution of Oceanography (1967), La Jolla. Most recent study comprises that of Roether and Münnich (1972) on the depth profile of tritium at the GEOSECS intercalibration station.

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The following study of the distribution of tritium at depth in the Southern Ocean was conducted jointly by the author as a part of a Ph.D. project completed in 1972 at the Flinders University of South Australia and the Commonwealth Scientific and Industrial Research Organisation (C.S.I.R.O.) Division of Soils, Glen Osmond, South Australia. The tritium data alone of this project appeared in a joint cruise report, SIO 72-19 put out by the Scripps Institution of Oceanography, La Jolla in 1972. Also in this paper, a few salient points are discussed on the basis of the observed data.

INVESTIGATION AND RESULTS

The area of investigation along 132°E. longitude in the Southern Ocean (Fig. 1) was chosen in order to investigate tritium-regimes in areas of rapid water-mass displacements such as are characterised by frontal zones namely, Subtropical Convergence (around 45°S.), Antarctic Convergence (around 53°S.), Antarctic Divergence (around 64°S), as well as to make an estimate of the age of the Antarctic Bottom Water found in the Indian-Antarctic and South Australian basins. Details of the area are shown in Fig. 2.

Altogether 39 Niskin-samples each of 6 litres were collected at selected levels and stations (Figs. 1 and 2) during USNS *Eltanin* Cruise 41 in December 1969 and January 1970. The samples were brought back to shore laboratory in screwcapped, high density polyethylene bottles having minimal transpiration characteristics for the walls and the narrow necks. Storage of the seawater samples in these

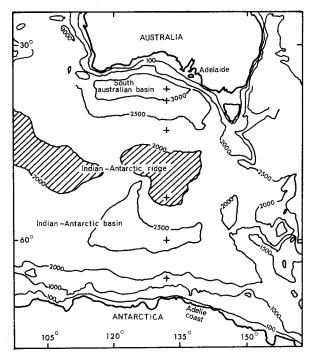


FIG. 1. Area of investigation and large volume Sampling Stations on USNS *Eltanin's* Cruise 41 along 132°E. indicated by + Contours are in fathoms.

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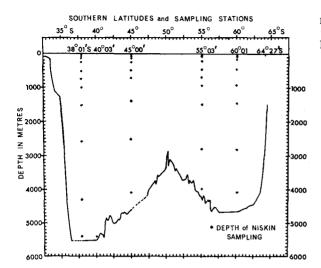


FIG. 2. Details of area investigated along 132°00'E. longitude.

containers for periods up to a maximum of 3 months was necessary since the processing time required for the laboratory analysis of tritium was limited by a rate of 5 to 7 samples per working week. The transpiration characteristics and vapour losses through the walls and the narrow necks of these containers were first tested by storing similar seawater samples and monitoring their salinities at fortnightly intervals with an Autolab Inductively Coupled Salinometer 601 MKIII. Variations in salinities were, however, not detectable during 3 months' storage either in a cellar under constant environmental conditions or in the laboratory at ambient temperatures.

As regards ambient exchange of tritium, tests made by the IAEA laboratories, Vienna, on samples stored in polyethylene bottles have been reported (IAEA 1967). Relatively small volume samples in a large volume container with most of the volume of the free air space above showed a maximum rate of increase of tritium in the water samples amounting to approximately 1.5 TU per year per 100 TU of ambient tritium. It was suggested in the same report that the change of tritium content in samples in completely filled bottles was considerably less. Furthermore, some of the results from experiments conducted at the University of Heidelberg and included in the above report showed 5 mg./year leakage rate of water vapour from tightly stoppered glass bottles as against 1000 mg./year, from wide-necked bottles.

Ambient tritium in Adelaide, South Australia, where the samples of this project were analysed is quite low ranging from 10 to 60 TU in rain water (Hughes and Holmes 1971). There have been relatively few and less powerful thermonuclear explosions in the southern hemisphere, and exchange of tritium by atmospheric mixing between the two hemispheres is slow (Bierly 1967).

The seawater samples were stored completely filled in the narrow-necked polyethylene bottles; they were withdrawn one at a time and handled carefully to avoid any accidental contamination by ambient tritium during processing. Since all of the tritium-free deep water samples gave on processing tritium levels within the limits of zero level concentration (<0.2 TU), no contamination was ever suspected.

Initially a volume of at least 3.25 litres of each sample was vacuum-distilled in a Büchi Rotavapor Unit, and subsequently 3 litres were processed using the analytical facility developed by the Soil Physics Section of the C.S.I.R.O. Division of Soils for the determination of low-level tritium in southern hemisphere natural waters (Hughes and Holmes 1971). In short, the analytical procedure consisted of the following treatment of the samples: 1) desalting by vacuum distillation; 2) electrolysis in 3 stages of 10X, 5X, and 10X volume reduction to effect enrichment of tritium; 3) reduction of the electrolysed water sample to hydrogen in a Mg-furnace at 600° C.; 4) tritium labelling by catalytic hydrogenation of tank ethylene at ambient temperature; and 5) gas proportional counting in anti-coincidence of tritiated ethane.

The results are presented in Table 1. Following Taylor (1963), the statistical error curve in Fig. 3 was constructed. Total 2-sigma error is less than 40 per cent at 0.2 TU level. Values lower than 0.2 TU are considered not to be significantly different from zero tritium concentration. Reproducibility and any contamination by ambient tritium including laboratory vapour were taken into account according

TABLE 1. Tritium content in TU* (Tritium Unit) in Southern Ocean water samples

Long. E Lat. S	131°59′ 38°01′	132°07′ 40°03′	131°57′ 45°00′	129°46′ 55°03′	132°13′ 60°01′	132°03′ 64°27′
Sample depth normal to		TU (at exact sample depth in meters)				
Surface	1.73 (<i>13</i>)	_	1.34 (5)	1.14 (<i>12</i>)	1.33 (24)	1.17 (5)
50 m.	1.90 (61)	_	1.28 (47)	1.27 (50)	1.38 (67)	, -
100 m.	1.67 (110)	_	1.48 (103)	_	_	-
200 m.	1.20 (248)	_	1.27 (253)	0.88 (250)	0.38 (174)	_
500 m.	0.65 (506)	 .	0.72 (493)	0.26 (486)	0.11 (<i>435</i>)	_
700 m.	0.30 (730)	_	0.92 (727)	0.38 (714)	_	-
1000 m.	0.21 (998)	_		0.21 (951)	0.31 (<i>910</i>)	-
1500 m.	0.15 (<i>1514</i>)	_	0.21 (<i>1407</i>)	0.28 (1507)	0.20 (1467)	_
2500 m.	0.13 (2578)	_	0.20 (2501)	0.32 (<i>2780</i>)	0.07 (2823)	_
4000 m.	0.20 (4300)		0.38 (4100)	0.35 (4004)	0.25 (4113)	_
5000 m.	0.23 (5394)	0.17 (<i>5337</i>)	_			_

^{*1} TU = 1 Tritium atom/ 10^{18} Protium atoms = c. 3.24 pCi/L of water = c. 7.2 dpm/L of water.

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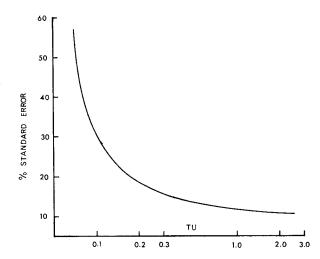


FIG. 3. Standard error for TU measured.

to the outline discussed by Cameron (1967). The laboratory blank found in this manner was incorporated in the determination of the background counting, the error of which did not exceed what was assigned as the total experimental error in Fig. 3.

The standard error displayed in Fig. 3 appeared during counting to have been associated more with the Poisson nature of the distribution of radioactivity both from sample and background. Overestimate of the error is therefore quite likely since the samples were counted for a limited time-period dependent on the timelimited availability of the tritium processing facility at the C.S.I.R.O. laboratories. Personal communications (Professor J. W. Holmes, The Flinders University of S.A., and Dr. Graham Allison, C.S.I.R.O. Division of Soils, Australia) confirm that with recent experience with the same counting facility about 8 per cent should be assigned as 1-sigma error for sample activities at 1 TU, 7 per cent at 2 TU, and thereafter, 5 to 7 per cent standard errors, irrespective of sample activity. The systematic errors for the analytical system contributing to the overall standard error were found to be less than half of what is presented in Fig. 3 (cf. Hughes and Holmes 1971). It has been suggested that the error at ≥ 1 TU activity could have been reduced to 5 per cent by counting the samples for longer periods of time. In this project, the total counting time needed for each measurement to have a standard error of about 3 per cent was predetermined after knowing the approximate activity of each sample at the end of the first 100 minutes' counting.

The representative depth-tritium plot of the results in Table 1 is drawn in Fig. 4. In order to be able to demonstrate the existence of the tritium-free layer at variable depths corresponding to salinity maximum (34.7 parts per thousand) upper and lower deep waters (Sverdrup 1940) in the Southern Ocean, areas of less than 0.2 TU tritium activity are shown in shaded contours in the same plot. It is likely that with sampling at closer depth intervals more than one tritium-free depth could have been detected on the profile at 55°S. Latitudinal variation of tritium content in the surface waters can be seen in Fig. 5 in which also are entered the salinity variations of the surface waters at the respective latitude points.

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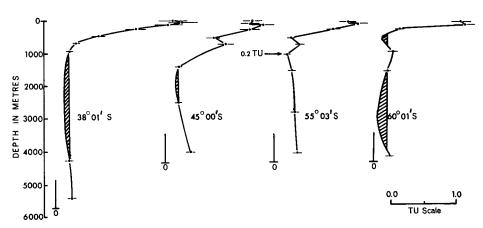


FIG. 4. Depth-tritium profiles at four Southern Latitudes (along 132°E.).

DISCUSSION

Presence of deep-sea tritium in concentrations significantly higher than zero tritium implies faster movement of bottom waters in this region adjacent to the Antarctic Continent. Tritium tracing may reveal more information regarding areas of bottom water formation around Antarctica. The disappearance of tritium at intermediate depths and the gradual displacement of this tritium-free layer upwards toward higher latitudes may provide complementary evidence of the expected countercurrent flow of the deep waters (Deacon 1937; Sverdrup 1940); this will in fact confirm that downward mixing does not contribute to the tritium income at the sea bottom. Although from a different standpoint, Roether and Münnich (1972) interpreted in a similar manner their account of tritium hydrography and subsurface water movement in the North Atlantic Ocean. Brown and Grummitt as far back as 1956 reported indications of tritium differentiation and limit of rapid mixing between surface and subsurface waters on the Canadian continental shelf southeast of Halifax, and suggested tritium tracing of subsurface water layers consistent with the southwesterly movement of surface waters in the region.

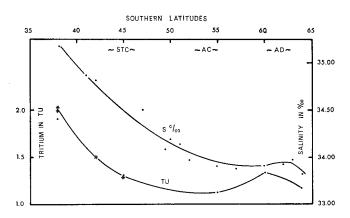


FIG. 5. Latitudinal variation of salinity and tritium content in surface ocean water (along 132°E.). Asterisks = values in the Australian Bight (see text).

Four additional values indicated by star-marks for tritium in surface water samples of the Australian Bight (Horace Lamb Centre 1969-70) are added to the plot in Fig. 5. Steady decrement of tritium content in surface waters from 38°S. latitude through 45°S., 55°S., 60°S., and 64°S. latitudes indicates rapid mixing of the waters in the frontal zones.

The difference in the concentration of tritium in surface waters can be regarded to be statistically real particularly when the concentration gradients between the reference tritium value of c. 2 TU around 38°S. latitude and the values at the four other latitudes are taken into account. The area around the lower latitude at 38°S, should be representative of an ocean surface which is relatively free of any rapid vertical mixing of waters, and therefore should be considered a reference area for comparison with respect to rapid water mixing and renewal at such other points characterised by the fronts such as the Subtropical Convergence (STC) around 44°S. to 45°S., Antarctic Convergence (AC) around 52°S. to 55°S., and Antarctic Divergence (AD) around 64°S. along 132°E. meridian. In this way highest and lowest differences observed in the tritium content of the surface waters are, 2 - 1.14 = 0.86 TU, and 2 - 1.34 = 0.66 TU respectively and significantly above the 1-sigma error of c. 11 per cent given in Fig. 3. The variation of surface water tritium may be looked at only in terms of a steady decrement from 38°S, through to 64°S, latitude, a traverse from subtropical through subantarctic to antarctic waters.

Horizontal salinity and temperature variations in the surface water layers and at least up to a depth of 150 m. to 200 m. provided systematic indications of areas of convergences and divergences in the Southern Ocean since the "Discovery II" expedition in 1932-34 (Deacon 1937). The observed tritium gradients in the surface waters appear to follow salinity gradients as shown in Fig. 5. If the standard error in Fig. 3 could be overlooked momentarily as an overestimate, it can then be emphasised that the general tendency of tritium variations in Fig. 5 markedly reflects that of salinity variations. However, it is apparent that additional data will be required to substantiate validity of tritium tracing of the water masses to past interpretations on surface and subsurface circulatory features in the Southern Ocean. Examination of tritium variations at frequent sampling intervals in the Southern Ocean could describe closely rapidly mixing areas of ocean surfaces and also differences in their mixing rates (Suess 1970).

Furthermore, the surface tritium values were analysed in an attempt to estimate the turnover or removal times of the upper mixed layer waters, τ_0 , using a balance equation (Bainbridge 1963; Eriksson 1965; Shannon *et al.* 1970) in conjunction with 10 years' IAEA (1969, 1970) data (1960-69) on rainfall tritium recorded for three representative island-stations in the southern hemisphere: Campbell Island 52°55'S., 169°15'E.), Invercargill (46°42'S., 168°32'E.), and Kaitoke (41°10'S., 175°17'E). The equation relates tritium input and removal as follows:

$$\begin{aligned} \frac{dN}{dt} &= I - RN - \lambda N \\ &= I - aN \end{aligned} \tag{1}$$

where N = concentration of tritium in the mixed layer, TU

I = input or rate of deposition of tritium into the defined 100 m.depth mixed layer, calculated from IAEA (1969, 1970) data (1960-69), TU year⁻¹

 $\lambda = \text{decay constant of tritium, } 0.056 \text{ year}^{-1}$

 R^{-1} = turnover or removal time, τo , of tritium through the mixed layer, year

and
$$a = (\lambda + R)$$
 or $(a - \lambda)^{-1} = R^{-1} = \tau o$

I, the tritium input by precipitation only to the ocean surface mixed layer of 100 m. depth was calculated from the following relationship,

$$I = \frac{P. TUP}{10^4} \frac{TU}{year}$$
 (2)

where P = mean annual precipitation, cm./year over the ocean

TUP = tritium concentration, TU, in precipitation

Average precipitation, P, over the ocean areas between 40°S. and 60°S. latitudes taken from Wüst's tables (1954) was 105 cm./year. Mean annual rainfall tritium for the area of investigation was calculated by correlating IAEA (1969, 1970) data (1960-69) recorded for the above three ocean-island stations in the southern hemisphere.

Regarding definition of a mixed layer in the Southern Ocean, it has to be remembered first of all that the samples for this project were collected during high southern summer of December 1969 to January 1970. Oceanographic observations do indicate that a shallow mixed layer of about 20 m. can be produced by a seasonal thermocline resulting from surface heating and an amplitude of variation of surface water temperature as great as 8°C. in areas above 35°N. latitude in the Pacific Ocean. In the absence of a seasonal or permanent thermocline at still higher latitudes e.g. polar areas, the mixed layer can be attributed to the depth subjected to mixing by wind (Ivanoff 1972). The 100 m.-depth surface layer was assumed to be typical of what may be called the upper mixed layer in the Southern Ocean. On a global scale this layer is said to be not less than 75 m. above the thermocline. Oceanographic studies since Deacon (1937) suggest a 150 m. to 200 m. depth as the limit of the upper waters both in the subantarctic and subtropical areas in the Southern Ocean. Fairhall (1969) assumed a 100 m.-depth mixed layer in the Antarctic seas to account for the suggested tritium-³He balance in deeper waters. The tritium results in Table 1 reflect such a gradient to exist at and below 100 m. depth at the respective sampling points along 132°E. longitude. Isobaths of 0 m., 50 m., 100 m., and 200 m. with tritium activities at the five surface points are shown in Fig. 6. Except at 45°S. where mixing of the waters probably reaches 200 m. depth, the tritium gradient appears to be quite evident below 100 m. By tritium gradient it is suggested that the tritium content abruptly decreases below the 100 m. level specified as the limit of the depth of the upper mixed layer.

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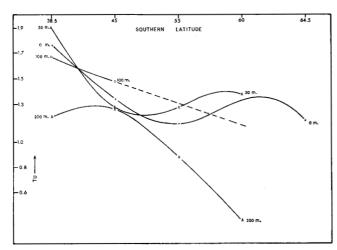


FIG. 6. Upper mixed layer isobaths with tritium activities at the five Southern Ocean latitudes.

It was assumed that tritium input by precipitation in 1969 largely contributed to the tritium concentration, N, observed in the samples of 1969-70, in addition to contributions by variable inputs from earlier years since 1961. Higher inputs of tritium by several orders of that by precipitation to the ocean surface by vapour-exchange were suggested by models in recent times (Eriksson 1965; Craig and Gordon 1965; Taylor 1968; Gat 1970). Experimental evidence, however, of the magnitude of molecular vapour-exchange of tritium into the ocean is believed to be lacking until now. Inventory methods were applied in the past to the study of tritium balance in the oceans. A recent study, comparable to this project, of precipitation tritium from 1954-67 balanced out with measured tritium in 1967 in the Crater Lake, Oregon (Simpson 1970) suggests that "approximately 70 per cent of the tritium present in Crater Lake in August 1967 can be explained by precipitation alone", and therefore questions the magnitude of the proposed aspects of tritium transfer by vapour-exchange i.e. much higher input than by precipitation alone, not only over lake surfaces but also over the oceans. In the project of this paper, however, with limited data only precipitation input of tritium was thought to be the dominant transfer mechanism in the middle latitudes of the Southern Ocean.

Initial steady-state was assumed between tritium input and removal in 1960 in the southern remisphere,

$$I - aN = 0 (3)$$

where I, the tritium input was calculated by equation (2) with rainfall tritium of 8 TU (IAEA 1969, 1970); this activity may have been at equilibrium state since the recent increase of bomb-tritium in the southern hemisphere rainwater, observed only in 1961, rose to a peak in the year 1965-66 followed by a slow decrease to 1969-70. The surface ocean concentration of tritium in the southern latitudes was reported at 1 TU in the year 1960 by Dockins (Scripps Institution of Oceanography, 1967). Junge (1963) suggests on the other hand a global tritium activity of 1 TU in seawater above the thermocline. A straightforward substitution of the above values in the steady-state equation (3) gives a τ_0 of 35 years which is not

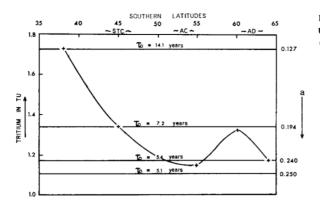


FIG. 7. Removal time of the upper mixed waters (100 m. depth and along 132°E.).

necessarily an indication of the removal time of the waters at 38°S. latitude. By inventory method Eriksson (1965) suggested a turnover time of 22 years for a mixed layer of 75 m.-depth in the pelagic ocean. By this estimate a 100 m.-depth mixed layer would overturn in about 30 years.

Under the assumed equilibrium conditions above, the balance equation (1) was solved numerically by a step-by-step computation for the period 1960-69 and by varying the constant "a" at fixed intervals. This procedure was adopted to account for any residual tritium from preceding years in the ocean's upper mixed layer. The computed quantities of N thereby obtained for the year 1969 in 100 m.-depth mixed layers were then compared with the observed quantities of N for 1969-70 at the five different latitudes. τ_0 s were then calculated directly from the comparable "a" s according to relations defined in the equation (1).

Fig. 7 shows the removal times, τ_0 s, of tritium and therefore, of the waters of the mixed layers at the respective positions. On the basis of the surface values alone, these are found to be of the order of 7 years for the waters at 45°S. and 60°S., 5 years at 55°S. and 64°S., about 15 years at 38°S. and probably more at lower latitudes north of 38°S., in pelagic areas which are free from up or downwellings.

Other workers using radioisotope data in box-models predicted residence times of the surface waters of the world oceans to be about 10 years (Pritchard *et al.* 1971). Montgomery's box-model (1959) does not use radioactive data but is based on salinity distribution and gives residence times of the subtropical surface waters in the southern hemisphere. Some of Montgomery's results relevant to what is proposed in this paper are reproduced in Table 2.

TABLE 2. Residence times of subtropical surface waters in the southern hemisphere (Montgomery 1959).

Water-mass	Exchanges with	τ0 years
South Pacific	High-latitude surface water, low-latitude	12
surface (< 230 m.) South Atlantic	surface water, deep water	9
surface (< 190 m.)		,
South Indian	44	6
surface (< 170 m.) Average (< 196 m.)		9

On the basis of the above approximation, mixed waters to 100 m. depth in the subtropical latitudes should renew within a period of the order of 5 years. Bainbridge and O'Brien (1962) further concluded from tritium abundance in a region of convergence off New Zealand that the waters there reached a depth of 2 km. in a 12-year time-period. Houterman's (1966) tentative interpretation of tritium results in the Pacific Ocean surface waters (1959-65) indicates an upper limit τ_0 of 12 years for the waters in the top 75 m. Geochemically speaking, the residence time of a relatively non-reactive chemical element such as Na⁺, Cl⁻, K⁺ in the mixed layer for the world ocean seems to be about 20 years when only the physical mixing processes are to cause removal of the element to deeper waters (Goldberg et al. 1971).

Reviewing the past cosmic-ray history of tritium, it should be assumed that in the southern hemisphere,

- an equilibrium state in tritium exchange existed between the tritium reservoirs;
- cosmic-ray tritium was supplied through production in the stratosphere and upper troposphere;
- according to the theory of the source and abyssal flow of the Antarctic Bottom Water (Brennecke 1921; Mosby 1934; Deacon 1937), present bottom waters in the two basins of the Southern Ocean (Fig. 2) were at the surface before the beginning of the thermonuclear input of additional tritium to the northern hemisphere atmosphere in 1954; and as a consequence, during residence at the surface the waters contained tritium in concentrations similar to the equilibrium pre-bomb level of 1 TU for surface ocean waters (Begemann and Libby 1957).

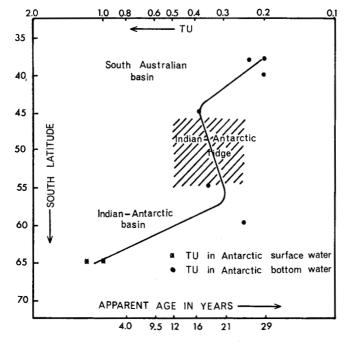


FIG. 8. Tritium content (TU) and apparent age of the bottom waters (≥ 4000 m. depth) along 132°00'E.

In the light of the present knowledge of stratosphere-troposphere mixing, general atmospheric circulation, and inter-hemispheric exchange of post-1954 northern hemisphere bomb-tritium (Libby 1958; Taylor 1966; Bierly 1967), it seems to be too complicated at this stage to predict an assessment if some of the northern bomb-tritium has already found its way into the upper waters of the Southern and Antarctic Oceans at high latitudes.

Consistent with the above assumptions, the present data suggest a minimum age of the order of 25 years for the bottom waters circulating in the entire region south of South Australia. The apparent tritium gradient of the bottom water samples from 60°S, to 38°S, latitudes is an indication in fact of an age gradient in relation to the observed (c. 1.2 TU) and the assumed (1 TU as above) tritium levels in the Antarctic Surface waters at c. 65°S, and higher latitudes (Fig. 8). In terms of northward movement of the water-mass at the bottom, a mean rate of about 1 cm./sec. seems quite probable. This is an estimate derived from tritium decay only for the meridional spreading of the bottom waters from antarctic areas into the Australian sector of the Southern Ocean. Without regular and frequent observations of tritium in the waters at depth and on a larger scale, it is difficult at present to account for the associated diffusional and mixing aspects of the Antarctic Bottom Water in its simultaneous movement to the east and to the north. Several theoretical and applied treatments on the subject of rate and direction of Antarctic Bottom Water circulation exist in the literature (Stommel 1958; Kort 1964; Munk 1966; Wright 1970).

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