

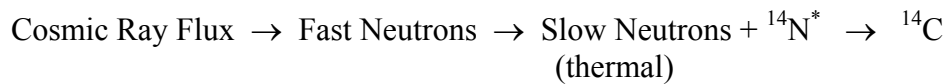
I. Cosmic Ray Production

Cosmic ray interactions produce a wide range of nuclides in terrestrial matter, particularly in the atmosphere, and in extraterrestrial material accreted by the earth. These nuclides have a wide range of chemical properties and half lives which enable them to be applied to a wide variety of geochemical problems. A summary of these isotopes, along with relevant information such as their mode of production and decay, half lives and pre-nuclear global inventory are summarized in Table 17-1.

Carbon-14 and tritium (³H) are the most well known of these isotopes. The main cosmic ray produced isotopes used on oceanography are listed below with their half-lives and pre-nuclear inventory.

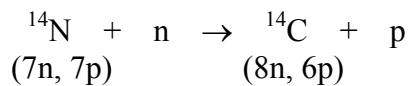
<u>Isotope</u>	<u>Half-life</u>	<u>Global inventory</u>
³ H	12.3 yr	3.5 kg
¹⁴ C	5730 yr	54 ton
¹⁰ Be	1.5 x 10 ⁶ yr	430 ton
⁷ Be	54 d	32 g
²⁶ Al	7.4 x 10 ⁵ yr	1.7 ton
³² Si	276 yr	1.4 kg

Carbon-14 is produced in the upper atmosphere as follows:



Cosmic rays (protons traveling through space with energies in the BEV range) hit the atmosphere and interact with molecules to produce fast-moving neutrons. These are slowed down by collisions so that by the time they reach about 50,000 ft they are slowly moving thermal neutrons. These thermal neutrons react with ¹⁴N (the most abundant nuclide in the atmosphere) to produce ¹⁴C. A neutron enters a ¹⁴N nucleus, knocking out a proton and remaining in its place. The loss of one proton converts the atom of nitrogen into carbon. The atomic weight remains the same at 14.

The reaction is written:



Since almost all the neutrons produced by cosmic rays are consumed to produce ¹⁴C the production rate is 2 atoms cm⁻² min⁻¹ and the pre-nuclear inventory (determined from the

balance between production and decay) was 54 tons. The production rate varies with the 11 yr sunspot cycle (Stuiver and Quay, 1981).

Tritium is produced from cosmic ray interactions with N and O. After production it exists as tritiated water (H - O - ^3H), thus it is an ideal tracer for water. Tritium concentrations are TU (tritium units) where

$$1 \text{ TU} = 10^{18} ({}^3\text{H} / \text{H})$$

Thus tritium has a well defined atmospheric input via rain and H₂O vapor exchange. Its residence time in the atmosphere is on the order of months.

In the pre-nuclear period the global inventory was only 3.5 kg which means there was very little ^3H in the ocean at that time.

II. Bomb Fallout

Nuclear weapons testing and nuclear reactors (e.g. Chernobyl) have been an extremely important sources of nuclides used as ocean tracers. The artificially produced radionuclides that have been detected in the marine environment are listed in Table 17-2. Some nuclides are conservative (^3H) while others are nonconservative (e.g. ^{95}Zr , ^{95}Nb). Others are only partially removed from seawater by adsorption or biological uptake (e.g. ^{65}Zn , ^{60}Co). In addition to ^3H and ^{14}C the main bomb produced isotopes have been:

Isotope	Half Life	Decay
^{90}Sr	28 yrs	beta
^{238}Pu	86 yrs	alpha
$^{239+240}\text{Pu}$	2.44×10^4 yrs	alpha
	6.6×10^3 yrs	alpha
^{137}Cs	30 yrs	beta, gamma

Nuclear weapons testing has been the overwhelmingly predominant source of ^3H , ^{14}C , ^{90}Sr and ^{137}Cs to the ocean. Although nuclear weapons testing peaked in 1961-1962 there were significant deliveries of radionuclides beginning as early as 1954. Fallout nuclides act as "**dyes**" that are spread over the sea surface, thus they have a well defined surface input.

Another group of man-made tracers that fall in this category but are not bomb-produced and are not radioactive are the chlorofluorocarbons (CFCs).

A. Tritium

Atmospheric testing of nuclear weapons increased the global inventory of ^3H from 3.5 kg to over 200 kg. The fusion bombs tested in the 1960's were much dirtier (2.0 kg ^3H / megaton) with ^3H than the fission bombs (0.07 kg ^3H / megaton) that had been tested earlier. Most of this tritium was injected into the stratosphere. The exchange between the

stratosphere and troposphere occurs mostly during "spring leaks" but once in the stratosphere the residence time of tritium is 30-60 days.

The time history of ^3H in rain from Adak, Alaska (52°N) to Johnston Island (17°N) from 1962 to 1967 is shown in Fig 17-1. The maximum in 1963 is clear as are the annual oscillations in input due to the "spring leak" between the stratosphere and troposphere. Most ^3H input occurred in the northern hemisphere where bombs were tested. The short residence time of atmospheric H_2O prevented inter-hemisphere transport of ^3H . The latitude dependence of ^3H input was maximum at about 50°N where the stratosphere/troposphere mixing occurs (Fig 17-2).

The key points about ^3H are that:

1. ^3H is input to the surface ocean
2. there is an input maximum at high latitude in the northern Hemisphere
3. there was a pulse input in 1963.

Once in the ocean, tritiated water follows the circulation and mixing patterns of the surface ocean (a man-made "dye" experiment). Surface ^3H in the Pacific Ocean determined in the 1974 GEOSECS Program (Fig 17-3) showed:

1. highest ^3H in the north Pacific
2. ^3H decreases southward.

The north-south section of ^3H through the Pacific (0-1000m) (Fig 17-4)(GEOSECS) shows how far the ^3H had penetrated into the ocean after only about 10 years.

1. ^3H was mostly in the northern hemisphere
2. most ^3H was within 1000m of the surface
3. there was a subsurface maximum of ^3H near the equator.

The implications of these distributions are that:

1. The equator is an effective barrier to N-S transport in the upper ocean.
2. Transport into the thermocline region occurs on a decadal time scale.
3. The sub-surface peak in ^3H implies rapid horizontal transport from high latitude can dominate over vertical transport.

The activity of ^3H appears to follow density surfaces (σ_t). The N-S sections of σ_t in the Atlantic, Indian and Pacific Oceans are shown in Fig 17-5. The dashed line shows the penetration of ^3H to the ^3H detection limit. We can test the ideas that transport of ^3H is along density or isopycnal surfaces by plotting ^3H versus density rather than depth (Fig 17-6). Note that when we do that we see that the maximum in ^3H at 12°N is on the same density as the high latitude maximum. Also note that the densest surface water in the north Pacific has a density of $\sigma_t = 26.8$ and this is approximately the boundary of ^3H penetration.

Conclusions:

1. Transport along isopycnal (σ_t) surfaces is an important process.
2. Surface ocean properties (e.g. ^3H , O_2 , etc) are input into the thermocline at the region of isopycnal "outcrop", where the σ_t surface intersects the mixed layer. Denser (and deeper) isopycnal surfaces outcrop at higher latitudes.
3. One-dimensional vertical advection-mixing models are not realistic in most thermocline situations.
4. Because of its half-life of 12 years, the presence of ^3H in the thermocline indicates that "ventilation" is rapid (on the order of 10's of years).

Tritium Ventilation Model:

We can apply a simple box-model to calculate the rate of exchange between the surface ocean and the thermocline (Sarmiento, 1983). The schematic of the model shows density surfaces from the thermocline ($\sigma_t = 26.0$ to 26.6) that outcrop in the mixed layer (Fig 17-7). The exchange rate constant between the thermocline and mixed layer is K_{TS} (yr^{-1}), the volume of mixed layer and thermocline boxes are V_S and V_T (m^3), respectively and λ is the decay constant for ^3H (yr^{-1}). The mass balance is then:

$$(\delta^3\text{H}/\text{dt})_{\text{thermocline}} = K_{ST}V_S^3\text{H}_S - K_{TS}V_T^3\text{H}_T - \lambda V_T^3\text{H}_T$$

Unfortunately tritium is not at steady state. To calculate K_{TS} we need to:

1. Determine V_S and V_T
2. Measure $(\delta^3\text{H}/\delta t)_T$
3. Estimate $(\delta^3\text{H}/\delta t)_S$

The results are shown in Table 17-3 where K_{TS} has been estimated for the thermocline of the north Atlantic. The fourth column gives values of τ for density surface from $\sigma_t = 26.2$ to 27.4 and $\tau = K_{TS}^{-1}$. The renewal times (τ) increase from 7 years at $\sigma_t = 26.2$ to 39 years at $\sigma_t = 27.4$.

Tritium in the Deep-Ocean.

Tritium is also a tracer for deep-water formation. In the western north Atlantic, tritium was determined in 1972 as part of GEOSECS and in 198X during that Transient Tracer Program (TTO). Comparison shows extensive invasion of ^3H into the deep north Atlantic in a relative short period of time (Fig 17-8)

B. Chlorofluorocarbons (CFC)

There are three anthropogenic CFCs that are also useful as ocean dye tracers.

Trichlorofluoromethane	F-11
Dichlorofluoromethane	F-12
Trichlorotrifluoroethane	F-113

The sources of these tracers include spray can propellants, refrigerator coolants and generators.

1. The production rate is known as a function of time.
2. The atmospheric concentration is known versus time.
3. Input to the ocean is by gas exchange.
4. The CFCs are thought to be non-reactive thus these are conservative tracers.
5. The ratios of F-11/F-12 in the atmosphere varied with time and this enables use of the CFCs to date water, and calculate ventilation rates (i.e., the time since last contact with the atmosphere).

The use of F-11/F-12 for "dating" is limited to the period between 1945 and 1977. Before 1945 the concentrations were too low and after 1977 the atmospheric ratio has been constant.

6. CFCs can be detected by a sensitive electron capture technique.

The atmospheric history is shown in Fig 17-9. Measured data only exist since the 1970's but the record has been extrapolated back in time using the production history.

The ratios of F-11/F-12 increased in the atmosphere from 1945 to 1977 (Fig 17-10)

The vertical distributions of F11/F12 and F113 versus density in the north Atlantic are shown in Fig 17-11.

The predicted F11/F12 and F113/F11 ages versus density are shown in Fig 17-12.

C. Carbon-14 (^{14}C)

Atmospheric nuclear testing also greatly perturbed the global inventory of ^{14}C . There are some important differences between the inputs to the ocean of ^3H and ^{14}C .

1. Atmospheric peak in 1963 and 1964 is not a single strong pulse like for ^3H . Removal of $^{14}\text{CO}_2$ from the atmosphere is not as fast as for ^3H (Fig 17-13).
2. Atmospheric $^{14}\text{CO}_2$ is well mixed and there is no interhemispheric gradient as for ^3H . There is a more homogeneous source of ^{14}C to the ocean.

Bomb ^{14}C input dominates over "pre-bomb" ^{14}C (Fig 17-14).

1. The highest ^{14}C in the surface ocean is at mid-latitudes
2. There is a minimum in ^{14}C at the equator because of dilution by upwelling of older, low ^{14}C water.

Applications of ^{14}C

A. Ocean Two-Box Models: Solve for V_{mix}

We can write the mass balance equations for water, stable carbon and ^{14}C (Radiocarbon). The equations can be combined to solve for V_{mix} using the pre-bomb distributions of ^{14}C . Before atmospheric nuclear testing the deep water of the Pacific had about 19% less ^{14}C than the surface water. We assume the mean depth is 3200m and we know the mean life of ^{14}C is 8200 yrs.

B. Ocean Mixing: Multi-Box Thermocline

C. Gas Exchange - Stagnant Boundary Layer Thickness

D. Deep-Ocean Residence Times

E. Use of ^{14}C to estimate the rate of Deep-Ocean Circulation

<to be added>

References:

Sarmiento J.L. (1983) A tritium box model of the north Atlantic thermocline. *J. Phys. Ocean.* 13, 1269-1274.

Stuiver M. and P. Quay (1981) *Earth Planet. Sci. Lett.*, 53, 349.

CFCs

CFC-11 and CFC-12 are gases of purely anthropogenic origin, whose atmospheric concentrations have increased from zero since the 1930s

CFC-11 and CFC-12 enter the surface ocean via gas exchange and are carried within the ocean as passive and conservative tracers of circulation and mixing processes that occur on decadal timescales. Thus CFCs are qualitatively similar to anthropogenic CO₂ and this has motivated the use of CFCs to evaluate the ability of ocean circulation models to simulate the uptake and redistribution of anthropogenic CO₂ in the oceans. The CFCs can be measured with high precision and measurements are available globally. The World Ocean Circulation Experiment greatly extended the coverage. The global CFC data set has proven to an excellent standard against which to assess the performance of the various models used to model ocean carbon distributions. Thirteen different models were compared with CFC-11 as part of the second stage of the Ocean Carbon-cycle Model Intercomparison project (OCMIP-2) (Dutay et al 2002).

Dutay J.-C. and 28 others (2002) Evaluation of ocean model ventilation with CFC-11: comparison of 13 global ocean models. *Ocean Modeling* 4, 89-120.