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NUCLEAR FUEL REPROCESSING AND RELATED DISCHARGES

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Introduction

Oceanographers have for some decades made use of human perturbations to the environment as tracers of ocean circulation and of the behavior of similar substances in the oceans. The study of the releases of anthropogenic radionuclides $-$ by-products of the nuclear industry $-$ by nuclear fuel reprocessing plants is an example of such an application. Other examples of the use of anthropogenic substances as oceanographic tracers addressed in this Encyclopedia include tritium and radiocarbon, largely resulting from atmospheric nuclear weapons testing in the 1950s and 1960s, and chlorofluorocarbons (CFCs), a family of gases which have been used in a variety of applications such as refrigeration and polymer manufacture since the 1920s. As outlined in this article, the source function of nuclear fuel reprocessing discharges, which is very different from that of either weapons test fallout or CFCs, is both a blessing and a curse, defining the unique applicability of these tracers but also requiring much additional and careful quantification.

The two nuclear fuel reprocessing plants of most interest to oceanographers are located at Sellafield, in the UK, which has been discharging wastes into the Irish Sea since 1952, and at Cap de la Hague, in north-western France, which has been discharging wastes into the English Channel since 1966. These releases have been well documented and monitored in most cases. It should be noted, however, that the releases of some isotopes of interest to oceanographers have not been as well monitored throughout the history of the plants because they were not of radiological concern, were difficult to measure, or both. This is particularly true for $99Tc$ and $129I$, which are the isotopes currently of greatest interest to the oceanographic community but for which specific, official release data are only available for the last two decades or less. The radionuclides most widely studied by ocean scientists have been ^{137}Cs , $134Cs$, $90Sr$, $125Sb$, $99Tc$, $129I$, and Pu isotopes. The Sellafield plant has dominated the releases of most of these, with the exception of 125Sb and, particularly in the 1990s, ¹²⁹I. In general, the Sellafield releases have been particularly well documented and are the easiest for interested scientists and other parties to obtain. The figures of releases presented in this article are based primarily on the Sellafield data, except where sufficient data are available for Cap de la Hague.

The location of the Sellafield and Cap de la Hague plants is important to the oceanographic application of their releases. As discussed below, the releases enter the surface circulation of the high latitude North Atlantic and are transported northwards into the Norwegian and Greenland Seas and the Arctic Ocean. They have been very useful as tracers of ventilation and deep water formation in these regions, which are of great importance to global thermohaline circulation and climate. In addition, the study of the dispersion of radionuclides from the reprocessing plants at Sellafield and Cap de la Hague serves as an analog for understanding the ultimate distribution and fate of other wastes arising from industrial activities in northwestern Europe.

Much of the literature on reprocessing releases in the oceans has focused on, or been driven by, concerns relating to contamination and radiological effects, and they have not found the same broad or general oceanographic application as some other anthropogenic tracers despite their great utility and the excellent quality of published work. This may be attributable to a variety of factors, including perhaps (1) the contaminant-based focus of much of the work and its publication outside the mainstream oceanographic literature, (2) the complications of the source function as compared with some other

tracers, and (3) the comparative difficulty of measuring many of these tracers and the large volumes of water that have typically been required. This article addresses the historic and future oceanographic applications of these releases.

A Note on Units

In most of the literature concerning nuclear fuel reprocessing releases, amounts (both releases and ensuing environmental concentrations) are expressed in activity units. The SI unit for activity is the becquerel (Bq), which replaced the previously common curie (Ci), a unit which was based on the activity of one gram of radium. These two units are related as follows:

> 1 Bq = 1 disintegration per second $1 \text{ Ci} = 3.7 \times 10^{10} \text{Bg}$

The conversion from activity to mass or molar units is dependent on the half-life of the isotope in question. Activity (*A*) is the product of the number of atoms of the isotope present (*N*) and its decay constant (λ): $A = \lambda N$. The decay constant, λ , is related to the half-life by $t_{1/2} = (\ln 2)/\lambda$. Using this equation, it is simple to convert from Bq to mol, given the decay constants in units of Bq to mol, given the decay constants in units of s^{-1} , and Avogadro's number of 6.023 × 10²³ atoms mol^{-1} .

It is also worth noting that the total activity of naturally occurring radionuclides in sea water is approximately 12 kBq m^{-3} . Most of this activity is from the long-lived naturally occuring isotope,
⁴⁰K ($t_{1/2} = 1.25 \times 10^9$ years). Activities contributed ⁴⁰K $(t_{1/2})$ from anthropogenic radionuclides greatly exceed this (millions of $Bq m^{-3}$) in the immediate vicinity of the Sellafield and La Hague outfall pipes. However, most oceanographic studies of these releases involve measurements of much lower activities: up to 10s of Bg m^{-3} in the case of ^{137}Cs , and generally even lower for other radionuclides, for instance mBq m^{-3} or less for plutonium isotopes and 129 I.

Nuclear Fuel Reprocessing and Resulting Tracer Releases

Origin and Description of Reprocessing Tracers

Nuclear fuel reprocessing involves the recovery of fissile material (plutonium and enriched uranium), and the separation of waste products from 'spent' (used) fuel rods from nuclear reactors. In the process, fuel rods, which have been stored for a time to allow short-lived radionuclides to decay, are dissolved and the resulting solution is chemically purified and separated into wastes of different composition and activity. Routine releases from the plants to the environment occur under controlled conditions and are limited to discharge totals dictated by the overseeing authorities of each country. The discharges have varied over the years as a function of the amount and type of fuel processed and changes in the reprocessing technology. The discharge limits themselves have changed, in response to monitoring efforts and also spurred by technological advances in waste-treatment capabilities. As a general rule, these changes have resulted in decreasing releases for most nuclides (most notably cesium and the actinide elements), but there are exceptions. For instance, the Enhanced Actinide Removal Plant (EARP) was constructed at the Sellafield site in the 1990s in order to enable the additional treatment and subsequent discharge of a backlog of previously stored wastes. Although the new technology enabled the removal of actinide elements from these wastes, it is not effective at removing ⁹⁹Tc. Therefore an allowance was made for increased discharge of 99 Tc, up to 200 TBq per year. The resulting pulse of increased ⁹⁹Tc discharges from Sellafield beginning in 1994 is currently being followed with great interest, as is discussed in more detail below.

The end result of these processes is the availability of a suite of oceanographic tracers with different discharge histories (e.g., in terms of the timing and magnitude of spikes) and a range of half-lives, and thus with a range of utility and applicability to studies of oceanographic processes at a variety of spatial and temporal scales. A brief summary of the reprocessing radionuclides most widely applied in oceanography is shown in **Table 1**, and examples of discharge histories are given in **Figure 1**. It has also been particularly useful in some cases to measure the ratio of a pair of tracers, for instance $^{134}Cs^{137}Cs$, $^{137}Cs^{90}Sr$, or potentially, $^{99}Tc^{129}I$. The use of isotope ratios can (1) provide temporal information and aid the estimation of rates of circulation, (2) mitigate the effects of mixing which will often alter individual concentrations more than ratios, and (3) aid in distinguishing the relative contributions of different sources of the nuclides in question. Finally, differences in the chemical behavior of the different elements released can be exploited to study different processes. Although most of the widely applied tracers are largely conservative in sea water and therefore serve as tracers of water movement, the actinides, particularly Pu,

Isotope	Half-life (years)	Sources	Applications
137Cs	30	Weapons testing, reprocessing (mostly Sellafield), Chernobyl	The 'signature' reprocessing tracer, used in the earliest studies of Sellafield releases. Has been applied in European coastal waters, the Nordic Seas, Arctic Ocean, and deep North Atlantic
90Sr	28	Weapons testing, reprocessing (mostly Sellafield)	In combination with ¹³⁷ Cs, early tracer for Sellafield discharges. The ¹³⁷ Cs/ ⁹⁰ Sr ratio was used to distinguish reprocessing and weapons sources
134 _{Cs}	2.06	Reprocessing and Chernobyl	In combination with ¹³⁷ Cs, this short-lived isotope has been used in the estimation of transit times, e.g., from Sellafield to the northern exit of the Irish Sea
125 Sb	2.7	Reprocessing, mostly Cap de la Hague	Circulation in the English Channel and North Sea, into the Skaggerak and Norwegian coastal waters
99Tc	213000	Reproducing - large pulse from Sellafield beginning in 1994. Some weapons testing	Surface circulation of European coastal waters, the Nordic Seas, Arctic Ocean, and East and West Greenland Currents
129	15.7×10^{6}	Reprocessing, mostly Cap de la Hague since 1990, some weapons testing	Deep water circulation in the Nordic Seas, Arctic Ocean, and Deep Western Boundary Current of the Atlantic Ocean. It has also been measured in European coastal waters and the Gulf of Mexico
241 Pu	14	Weapons testing, reprocessing (mostly Sellafield)	Not widely used as circulation tracers but often measured in conjunction with other reprocessing radionuclides. Other transuranic
$^{239}P_{11}$	24 000		
240P _U 238 Pu	6000		
	88		elements that have been measured include 241 Am and 237 Np

Table 1 Summary of the major reprocessing tracers and their applications

Adapted in part from Dahlgaard (1995).

have a high affinity for particulate material and accumulate in the sediments. These tracers are then useful for studying sedimentary processes.

Figure 1 Examples of the source functions of reprocessing tracers to the oceans, illustrating some of the differences in magnitude and timing of the releases. 137Cs data, from Gray et al. (1995) and ⁹⁹Tc data, courtesy of Peter Kershaw, are for liquid discharges from Sellafield. 129I release information is courtesy of G. Raisbeck and F. Yiou, and combines available information for Sellafield from 1966 and Cap de la Hague from 1975. Note the different scales used for the releases of the three isotopes.

The Reprocessing Tracer Source Function

The primary difference between the north-western European reprocessing releases and other anthropogenic tracers used in oceanography is the nature of their introduction to the oceans. Reprocessing releases enter the oceans essentially at a point source, rather than in a more globally distributed fashion as is the case for weapons test fallout or the chlorofluorocarbons. Thus, reprocessing tracers are excellent, specific tracers for waters originating from north-western Europe. Because these waters are transported to the north into the Nordic Seas and Arctic Ocean, reprocessing releases are particularly sensitive tracers of the climatically important deep water formation processes that occur in these regions.

There are several complications associated with this point source tracer introduction, however. Comparison with CFCs indicates some of these difficulties. Within each hemisphere (northern or southern), CFCs are well mixed throughout the troposphere, and the time history of their concentrations is well known. Their entry into the oceans occurs by equilibration with surface waters, and the details of their solubilities as a function of temperature and salinity have been well characterized. The primary complication is that in some areas equilibrium saturation is not reached, and so assumptions must be made about the degree of equilibration. Where they have been necessary, these assumptions appear to be fairly robust. In the case of reprocessing tracers, the discharge amounts have not always been as well known, although this situation has improved continuously. One major difficulty arises in translating a discharge amount, in kg or Bq per year, or per month, to a concentration in sea water some time later. In order to do this, the surface circulation of the coastal regions must be very well known. This circulation is highly variable, on daily, seasonal, interannual and decadal timescales, further complicating the problem. Also, other sources of the radionuclides under consideration must often be taken into account. Depending on the tracer, these sources may be important and/or numerous, and due to the secrecy involved in many aspects of the operation of nuclear installations the necessary information may not always be available.

Other Sources of Anthropogenic Radionuclides

Other sources of radionuclides to the oceans complicate the interpretation of Sellafield and Cap de la Hague tracers to varying extents, depending on the isotope under consideration, the location, and the time. First there is the need to understand the mixing of signals from these two plants. Other sources may include fallout from nuclear weapons tests, uncontrolled releases due to nuclear accidents, dumping on the seabed, other reprocessing plants, atmospheric releases from Sellafield and Cap de la Hague, and unknown sources. Many of these other sources are small compared to the Sellafield and Cap de la Hague releases. The primary complicating source for many isotopes is nuclear weapons test fallout, which peaked in the 1950s and again, more strongly, in 1962–63. This source has been particularly important for ^{137}Cs and ^{90}Sr . The Chernobyl accident in 1986 also released significant amounts of radioactive materials which have themselves found use as oceanographic tracers. In terms of comparison to releases from Sellafield, the Chernobyl accident has been most important with respect to ¹³⁴Cs and ¹³⁷Cs.

In addition, it has recently been noted that in the case of some nuclides, particularly $137Cs$ and Pu isotopes, the sediments of the Irish Sea have become a significant ongoing source of tracers to the North Atlantic. Although generally considered a conservative tracer, ^{137}Cs exhibits some affinity for particulate material and a significant amount has accumulated in the sediments around Sellafield. With the continuing reduction of ¹³⁷Cs activities in liquid effluents from Sellafield, release from the sediments, either by resuspension of the sediments or by reequilibration with the reduced seawater concentrations, has become a relatively large (though still small in an absolute sense compared to the liquid discharges of the 1970s) contributor to the current flux out of the Irish Sea, and will continue to be such for years to come.

A further complication of the use of some tracers derived from Sellafield and Cap de la Hague, but one that cannot be lamented, is the continuing reduction of the releases, as well as the radioactive decay of those with the shorter half-lives, such as $137Cs$ which was released in large quantities over 20 years ago. With respect to many of these complications, and for several other reasons, the reprocessing radionuclides attracting the most attention in the oceanographic community today are ⁹⁹Tc and ¹²⁹I. Both are long lived and have fairly small relative contributions from weapons testing and other sources. Unlike most radionuclides, their releases increased in the 1990s, and the recent releases of each are largely dominated by a single source: ⁹⁹Tc by Sellafield and 129 I by Cap de la Hague. Significant advances have been and are being made in the measurement of these isotopes, allowing their measurement on smaller sample volumes. The activity of 129 I is so low that it is measured by accelerator mass spectrometry. This technique allows measurement of 129I on 1 liter seawater samples. Advances in ⁹⁹Tc measurement, using both radiochemical and mass-spectrometric (ICP-MS) techniques, are continuing. The primary limitation of 99Tc studies continues to be the comparative difficulty of its measurement. This is also true to some extent for 129 I, for although the sample sizes have been greatly reduced the measurement requires highly specialized technology. A further complication for 129 I is its volatility, and the fact that as much as 10% of the reprocessing discharges have been released directly to the atmosphere. Nevertheless, the promise of both tracers is such that these difficulties are likely to be overcome.

Regional Setting and Circulation of Reprocessing Discharges

A summary of the regional circulation into which the liquid effluents from Sellafield and Cap de la Hague are released is presented in **Figure 2**. It is particularly important to note that studies of the

Figure 2 Map of the northern North Atlantic and Nordic Seas, indicating the locations of the Sellafield and Cap de la Hague reprocessing plants (stars), and the major circulation pathways relevant to the discussion of the dispersal of reprocessing wastes. Surface currents are indicated with thin solid lines, and deep waters, (deep overflows from the Nordic Seas, the resulting Deep Western Boundary Current (DWBC) and Labrador Sea Water (LSW)), with heavy dashed lines. Curled arrows indicate the two major areas where deep waters are formed by convective processes. Additional abbreviations: NCC, Norwegian Coastal Current; WSC, West Spitsbergen Current; EGC, East Greenland Current; FBC, Faroe Banks Channel; ISOW, Iceland Scotland Overflow Water; NEADW, Northeast Atlantic Deep Water; DSOW, Denmark Strait Overflow Water.

reprocessing discharges have contributed greatly to the development of this detailed picture of the regional circulation. Briefly, from the Sellafield site the waste stream is carried north out of the Irish Sea, around the coast of Scotland, through the North Sea, and into the northward-flowing Norwegian Coastal Current (NCC). Transport across the North Sea occurs at various latitudes: some fraction of the reprocessing releases 'short-circuits' across the northern part of the North Sea, while some flows farther south along the eastern coast of the UK before turning east and north. Recent studies of the EARP ⁹⁹Tc pulse from Sellafield have suggested that the rate and preferred transport path of Sellafield releases across the North Sea into the NCC may vary in relation to climatic conditions in the North Atlantic such as the North Atlantic Oscillation (NAO).

Radionuclides discharged from the Cap de la Hague reprocessing plant flow north-east through the English Channel and into the North Sea, following the coast and joining the Sellafield releases in the NCC. A small amount of the Sellafield releases and some of the Cap de la Hague releases, which flow closer to the coast, flow east through the Skaggerak and Kattegat to enter the Baltic Sea.

The NCC is formed of a mixture of coastal waters (containing the reprocessing tracers) and warm, saline North Atlantic surface waters. Dilution of the reprocessing signal with Atlantic water continues along the northward flow path of the NCC. There is evidence from reprocessing tracers that turbulent eddies between the NCC and Atlantic water result in episodic transport westward into the surface waters of the Norwegian Sea, in addition to a fairly well-defined westward advective transport towards Jan Mayen Island. The NCC branches north of Norway, with one branch, the North Cape (or Nordkap) Current, flowing eastward through the Barents Sea and thence into the Kara Sea and Arctic Ocean, and the remainder flowing north and west as part of the West Spitsbergen Current (WSC). This latter flow branches in the Fram Strait west of Spitsbergen, with some recirculation to the west and south joining the southward flowing East Greenland Current (EGC), and the remainder entering the Arctic Ocean. The majority of surface outflow from the Arctic is through the Fram Strait into the EGC, thus the bulk of the reprocessing nuclides entering the Arctic Ocean will eventually exit to the Nordic (Greenland, Iceland, and Norwegian) Seas and the North Atlantic. The presence of high concentrations of radionuclides derived from reprocessing in the surface waters of the Barents and Nordic Seas is also an indication of their utility as tracers of deep-water ventilation and formation in these regions, as discussed below.

The Use of Reprocessing Releases in Oceanography

Historical Background

The first papers reporting measurements of Sellafield-derived $137Cs$ in coastal waters appeared in the early 1970s. Much of this early work arose from monitoring efforts by the Division of Fisheries Research of the UK's Ministry of Agriculture, Fisheries, and Food, which holds a joint oversight role on the Sellafield discharges and which continues to this day, now as the Centre for Environment, Fisheries and Aquaculture Science, to be a leader in studies of the distribution and oceanographic application of reprocessing radionuclides. In the late 1970s scientist at the Woods Hole Oceanographic Institution published the first of a number of papers detailing the unique utility of the reprocessing tracers and demonstrating their application. Leading studies of reprocessing tracers in the oceans have been undertaken by researchers in numerous other countries affected by the radiological implications of the releases, including France, Germany, Denmark, Canada, Norway, and the former Soviet Union.

Coastal and Surface Circulation

A great deal of work has been published using the documented releases of radioisotopes from Sellafield and Cap de la Hague to study the local circulations of the Irish Sea, North Sea, and English Channel. Early studies of the Sellafield releases examined a variety of isotopes, including ^{134}Cs , ^{137}Cs , ^{90}Sr , and Pu isotopes. Most attention focused on ¹³⁷Cs and 90 Sr, and their activity ratio, because: (1) the releases of these two isotopes were well documented, (2) they had been studied extensively since the 1950s and 1960s in weapons test fallout; and (3) the $137Cs/90Sr$ ratio in reprocessing releases (particularly those from Sellafield) was significantly higher than in global fallout and thus could be used to distinguish the sources of these isotopes in a given water sample. The use of the $^{134}Cs/^{137}Cs$ ratio enabled estimates of transit times, assuming the initial ratio in the releases was constant and making use of the short half-life of ^{134}Cs . The short-lived isotope ^{125}Sb has been used as a specific tracer of the circulation of Cap de la Hague discharges through the English Channel and North Sea, into the Baltic and the Norwegian Coastal Current.

Summaries of transit times and dilution factors for the transport of Sellafield and Cap de la Hague discharges to points throughout the North Sea, Norwegian Coastal Current, Barents and Kara Seas, Greenland Sea, and East and West Greenland Currents have been published in recent reviews. Numbers are not included in this article because they are currently under revision. In terms of transport to the NCC, where the two waste streams are generally considered to merge, the consensus has been that the transit time from Sellafield to about 60° N is three to four years, and that from Cap de la Hague to the same area is one to two years. Compilations of 'transfer factors,' which relate observed concentrations to the discharge amounts, and factor in the transit times, suggest that the two reprocessing waste streams meet in approximately equal proportions in the NCC. In other words, if Sellafield and Cap de la Hague released equal amounts of a radionuclide, with the Cap de la Hague release two years later, they would make equal contributions to the NCC.

Recently, detailed studies have been undertaken of the dispersal of the EARP ⁹⁹Tc pulse from Sellafield, which began in 1994. These studies have suggested substantially shorter circulation times for Sellafield releases to northern Scottish coastal waters and across the North Sea to the NCC than previously accepted. For instance, the $99Tc$ pulse reached the NCC within 2.5 years, rather than three to four. It has been suggested that this is a real difference between sampling periods, resulting from climatically induced circulation changes in the North Atlantic. Some of the difference may also be related to the fact that much more detailed data, both in terms of seawater sampling and regarding the releases, are available on this recent event. The continuing passage of the EARP ⁹⁹Tc signal promises to be very useful in the Nordic Seas and Arctic Ocean as well.

Deep-water Formation in the North Atlantic and Arctic Oceans

In addition to surface water flows, deep-water formation processes within the Arctic Ocean and

Nordic Seas have been elucidated through the study of reprocessing releases. In a classic presentation of reprocessing tracer data, Livingston showed that the surface water distribution of ¹³⁷Cs in the Nordic Seas in the early 1980s was marked by high concentrations at the margins of the seas, highlighting the delivery of the isotope in the northward-flowing NCC and WSC to the west and the return flow in the southward-flowing EGC to the east. The opposite distribution was found in the deep waters, with higher concentrations in the center of the Greenland Basin than at the margins, as a result of the ventilation of the Greenland Sea Deep Water by deep convective processes in the center of the gyre.

In the Arctic Ocean, elevated ¹³⁷Cs and ⁹⁰Sr concentrations and $^{137}Cs/^{90}Sr$ ratios at 1500 m at the LOREX ice station near the North Pole in 1979 indicated that deep layers of the Arctic Ocean were ventilated from the shelves. Similar observations in deep water north of Fram Strait provided early evidence suggesting a contribution of dense brines from the Barents Sea shelf to the bottom waters of the Nansen Basin. Reprocessing tracers, particularly those, like ^{129}I and ^{99}Tc , which have only a small contribution from other sources such as weapons fallout, hold great promise for illuminating eastern Arctic Ocean deep water ventilation processes from the Barents and Kara Sea shelves. The NCC delivers reprocessing tracers to the Barents and Kara relatively rapidly (\sim 5 years) and more importantly in high concentration. With high tracer concentrations in the area of interest as a source water, the reprocessing tracers may be particularly sensitive tracers of a contribution of dense shelf waters to the deep Arctic Ocean.

In addition to the deep waters formed through convection in the polar regions (most notably in the Greenland Sea) which fill the deep basins of the Nordic Seas, intermediate waters are formed which subsequently overflow the sills between Greenland, Iceland, and Scotland and ventilate the deep North Atlantic. The presence of ^{137}Cs and ^{90}Sr from Sellafield was reported in the overflow waters immediately south of the Denmark Straits sampled during the Transient Tracers in the Ocean (TTO) program in 1981. Later, it was demonstrated that reprocessing cesium and strontium could be distinguished in the deep waters as far as TTO Station 214, off the Grand Banks of Newfoundland. No samples were taken for reprocessing radionuclides further south as part of that study, but it was clear in retrospect that the reprocessing signal had traveled even further in the Deep Western Boundary Current (DWBC). Recent work on 129I has highlighted the utility of reprocessing radionuclides as tracers of northern source water masses and the DWBC of the Atlantic. Profiles in stations south of the overflows show much clearer tracer signals for ¹²⁹I than for the CFCs, and ¹²⁹I has been detected in the DWBC as far south as Cape Hatteras. As with the cesium studies of the 1980s, it is likely that sampling further south will reveal the tracer there as well.

Conclusions

Releases of radionuclides from the nuclear fuelreprocessing plants at Sellafield and Cap de la Hague have provided tracers for detailed studies of the circulations of the local environment into which they are released, namely the Irish Sea, English Channel, and North Sea, and for the larger-scale circulation processes of the North Atlantic and Arctic Oceans. These tracers have very different source functions for their introduction into the oceans compared to other widely used anthropogenic tracers, and in some cases compared to each other. The fact that they are released at point sources makes them highly specific tracers of several interesting processes in ocean circulation, but the nature of the releases has complicated their quantitative interpretation to some extent.

Recent advances have been made in the measurement of ^{129}I and ^{99}Tc , long-lived tracers whose releases have increased in recent years and which experience little complication from other sources. These two tracers hold great promise for elucidating deep water formation and ventilation processes in the North Atlantic, Nordic Seas, and Arctic Ocean in the years to come.

See also

Arctic Basin Circulation. CFCs in the Ocean. North Sea Circulation. Radioactive Wastes. Water Types and Water Masses.

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