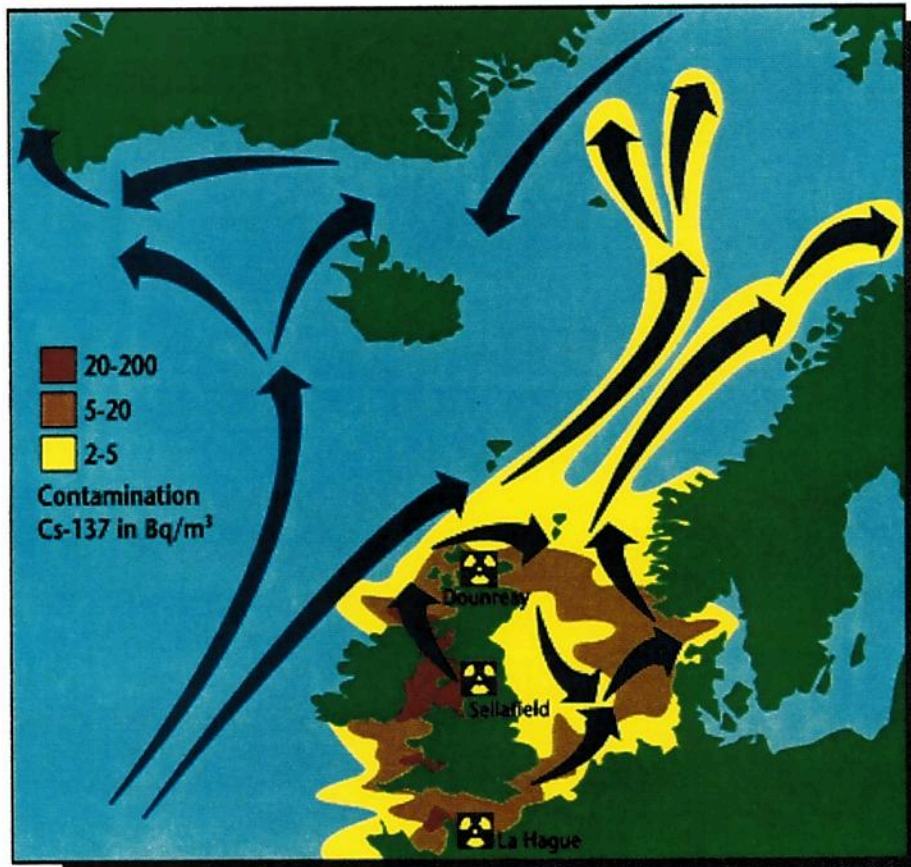


LIQUID DISCHARGES FROM EUROPEAN REPROCESSING FACILITIES

A REPORT BY GREENPEACE INTERNATIONAL



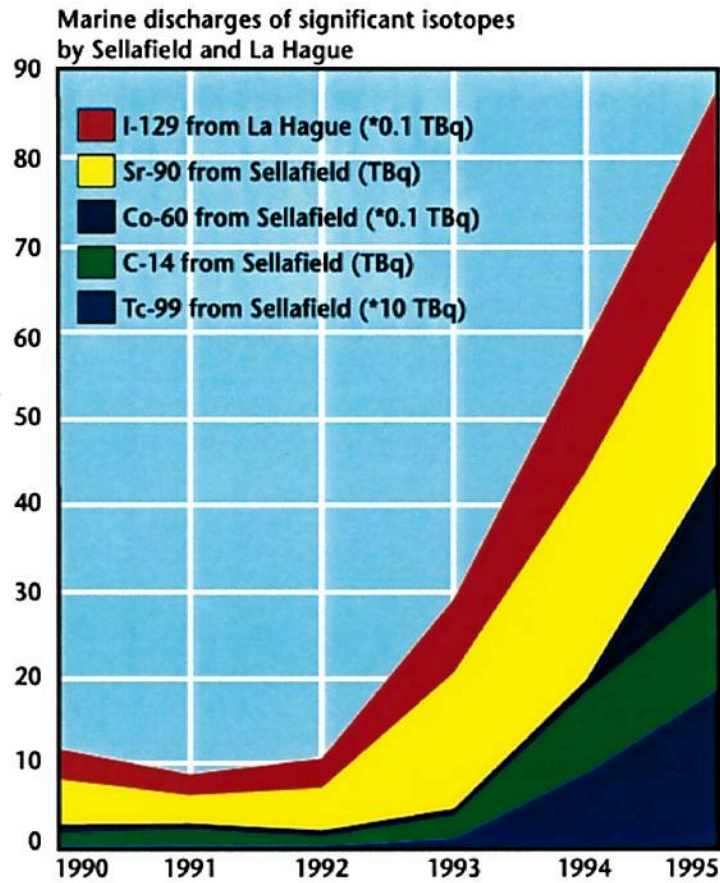
LIQUID DISCHARGES FROM EUROPEAN REPROCESSING FACILITIES

A REPORT BY GREENPEACE INTERNATIONAL

Written and researched

by

Diederik Samsom



OSPAR Final Minister's declaration 1992

“recognized .. need to reduce radioactive discharges from nuclear installations to the marine environment”

1. INTRODUCTION

- 1.1 In contrast with the legal obligation set out in the Paris (1974) and OSPAR (1992) conventions to “take all possible steps to reduce and eliminate pollution and [to] take the necessary measures to protect the [North East Atlantic] against the adverse effects of human activities so as to safeguard human health and to conserve marine ecosystems”¹ and the recognised “need to reduce radioactive discharges from nuclear installations to the marine environment”², discharges and emissions of radioactive wastes from nuclear reprocessing facilities show a significant increase, which is already resulting in new increases of contamination levels in the waters of the North West European shelf as well as the Arctic region. Moreover it is likely that the ongoing trend will continue for at least the coming years, given the expected increase in throughput of spent nuclear fuel.
- 1.2 Over the last four decades discharges of radioactive substances from reprocessing facilities have played a major role in the contamination of the North-West European shelf (Channel, North Sea, Irish Sea, North-East Atlantic). Especially during the 1970s the reprocessing industry contributed very large amounts of fission and activation products, like Cs-137, Sr-90, Pu-239 etc., to the waters of the English Channel and the Irish Sea. These isotopes are spread throughout the whole Atlantic Ocean and can still be detected in the Nordic Seas and South-West of Greenland.
- 1.3 During the 1980s reprocessing discharges were reduced following the application of new techniques and changes in the types of fuel reprocessed. However radioactive discharges from reprocessing plants in the 1990s show the opposite trend.
- 1.4 This report gives an overview of the discharges from reprocessing facilities, their dispersion through the oceanic waters and the radiological consequences. Special attention will be paid to the specific problems of the recent increase in discharges.

2. GENERAL DESCRIPTION OF THE REPROCESSING TECHNOLOGY

- 2.1. Reprocessing is the chemical separation of plutonium, uranium and fission products from spent nuclear fuel. Each type of fuel determines the exact technology used for reprocessing. Commercial reprocessing, as undertaken in France and the UK, mostly consists of reprocessing of LWR (Light Water Reactor)-oxide fuels or AGR-fuels (Advanced Gas-cooled Reactor)
- 2.2. The technology used for reprocessing these fuels is called the PUREX-process (Plutonium Uranium Recovery by Extraction). A PUREX-process includes the following installations³⁴:
 1. Reception and charging (dry or under water) facilities for the transport containers,
 2. Several spent fuel storage pools,
 3. Workshops for mechanical and chemical processes which follow the PUREX reprocessing procedure,
 4. Plants for the treatment of liquid effluents,
 5. Storage facilities for solid and liquid wastes,
 6. Laboratories, maintenance departments, energy and fluid production units, providing logistical support and security measures.
- 2.3. The different stages of the process are:

Shearing: the nuclear materials are contained in the fuel elements in the form of sintered oxide pellets stacked in zircaloy rods. The rods have to be sheared to gain access to the oxides - usually to a length of a few centimetres, leaving both ends open. These fuel rod sections are put into a dissolver.

Dissolution in nitric acid: This dissolves the uranium, plutonium and most of the fission products, but does not attack the zircaloy cladding. This dissolution is followed by a clarification of the solution by centrifugation in order to separate the insoluble parts and small fragments of cladding

Extraction of uranium and plutonium: the first step consists of eliminating the fission products from the solution. The uranium and plutonium are extracted together, most of the fission products staying in the aqueous solution. Due to the high decontamination factors necessary, several extractions are performed, usually three. During one of them the plutonium is separated from the uranium, the uranium staying in the organic phase and the plutonium migrating to the aqueous one.

Preparation of final products (oxides of Pu and U): the purified plutonium nitrate solution is concentrated by evaporation, then the plutonium is precipitated by oxalic acid. The oxalate is then filtered and dried, and when roasted at 450 C decomposed into PuO₂ and is stocked. The uranium nitrate solution can also be concentrated by evaporation, but is usually left in a nitrate form because of lack of demand for this product.³

3. LIQUID EFFLUENTS INTO THE MARINE ENVIRONMENT FROM REPROCESSING.

- 3.1 During all steps in the reprocessing process, liquid wastes are formed. These wastes are collected and sent to one or more effluent treatment centres. Due to the very aggressive (both chemical and radiological) nature of the effluents produced in the reprocessing process, these treatment centres consist of complex chains of consecutive stages in which chemicals like nitrates and phosphates are partly removed and a part of the radioelements in the effluent are removed by precipitation or flocculation before releasing the effluents into the sea. In fact, the efficiency of extraction of the radionuclides is much less than 100% and quantities of the principal elements undergoing treatment remain in the liquid discharges into the sea.⁵
- 3.2 Since the principle of the reprocessing process is based on the dissolution of spent fuel it is not surprising that the radioactive effluents which are treated in the facilities and ultimately discharged into the marine environment are mainly contaminated with actinides and fission products.
- 3.3 A typical list of the isotopes and the amounts released by the three currently operating reprocessing facilities in North West Europe is shown in Table 1⁶.

Table 1: Liquid discharges in 1995 from reprocessing facilities

<i>Nuclide</i>	<i>Sellafield (TBq)</i>	<i>La Hague (TBq)</i>	<i>Dounreay (TBq)</i>
Tritium	2700	9610	1,1
C-14	12	not available (!)	not available
Co-60	1,3	0,548	0,027
Sr-90	28	14,8	0,6
Tc-99	190	0,1	not available
Ru-106	7,3	7,6	0,76
Sb-125	9,3	2,95	not available
Cs-137	12	4,62	3,7
Mn-54	0,08	0,0306	not available
Total Beta	191	53	7
Total Alpha	0,4	0,08	0,09
Pu-alpha	0,31	not available (!)	not available
Pu-241	7,7	0,48	0,55
Am-241	0,11	0,001	not available
Uranium (kg)	1300 (kg)	not available	not available

- 3.4 These inputs form by far the largest source of artificial radionuclide inputs into waters of the European continental shelf. Table 2 gives an overview of the total input from different types of facilities from the start of their operation until 1986⁷.

Table 2: Site contributions to the total discharges

Type of facility	Alpha emitters (%)	Beta emitters (%)	H-3 (%)
Reprocessing	97,4	98.6	69,7
Power Stations	0,2	0,5	19,7
Research	0,01	0,02	10,8
Other	2,4	0,9	0

- 3.5 Although the reprocessing industry claims to have applied all the possible measures to reduce the contamination levels of the discharged effluents and states that filtering techniques are highly effective⁸⁹, the discharges of reprocessing facilities are orders of magnitudes higher than other parts of the nuclear fuel cycle (e.g. storage of spent fuel, electricity production).
- 3.6 A comparison between different stages of the nuclear fuel cycle shows clearly that even when reprocessing discharges are corrected for the total electricity production of the handled fuel (in MBq/TWh = the *normalised* discharges), these normalised reprocessing discharges into the environment are at least two orders of magnitude higher than the normalised discharges from other parts of the fuel cycle (Table 3).

Table 3: Comparison of releases of radioactivity from different stages of nuclear fuel cycle

Activity	liquid releases MBq/TWh	Total releases MBq/TWh	fraction
Electricity generation	1,6 E 6	4,2 E 6	0,5%
Dry Storage	7,3	9	1,2E-6 %
Reprocessing	3 E 7	7,4 E 8	99,45 %

- 3.7 Reprocessing is just one way of managing the back-end of the nuclear fuel cycle before trying to find a final disposal option for nuclear waste. As shown in Table 3, the most commonly used alternative, *interim storage*, releases approx. 1/100000000 of the radioactivity to the environment compared to reprocessing. In this connection, The IAEA Principles of Radioactive Waste Management, adopted in March 1995, are relevant. According to the IAEA, “*The preferred approach to radioactive waste management is concentration and containment of radionuclides rather than dilution and dispersion in the environment.*”; the IAEA does recognise that “*however, as part of radioactive waste management, radioactive substances may be released within authorised limits*”, but they warn that “*for all practical purposes this is an irreversible action and is considered suitable only for limited amounts of specific radioactive waste*”.¹⁰ Table 3 shows that reprocessing is in breach of the IAEA Principles given that dry storage limits considerably dilution and dispersion into the environment.

4. TRENDS IN DISCHARGES

SELLAFIELD

- 4.1 Annual discharges from Sellafield have shown a wide variety over the past decades. After peaking in the mid- to late-1970s with some remarkable inputs from Cs-137 and actinides¹¹ the mid 1980s show an overall decrease in discharged radioactivity from the Sellafield site.
- 4.2 However, since the beginning of the 90s new developments at the site reversed this trend: the total amount of radioactivity in the discharge streams is now steadily rising. In 1993 BNFL, the operator of Sellafield, was given a new discharge authorisation, allowing increases in discharges due to the start up of its new Thermal Oxide Reprocessing Plant (THORP) and the Enhanced Actinide Removal Plant (EARP).
- 4.3 Since the start up of EARP a backlog of stored wastes on site is again released to the sea. The activity of some isotopes, like Pu and Am have been reduced in the waste streams¹⁸, but the same plant causes a significant increase in the release of other isotopes, particularly Sr-90 and Tc-99. EARP is used for the treatment of routine and decay-stored waste streams from the Magnox-reprocessing plant. The removal technique in this facility applies well to Cs-137, Ru-106 and actinides, but the efficiency of the removal of Sr-90 is very low and Tc-99 is in fact not treated at all in this plant. Diversion of the C-14 stream from the gaseous into the liquid waste resulted in an increased discharge from 2 TBq/y to about 12 TBq/y in 1995.¹²
- 4.4 Other changes in the liquid discharges from Sellafield are resulting from a greater throughput of Magnox fuel, higher burn-up levels in reactors and the decommissioning of wastes from numerous redundant plants at the site.
- 4.5 The changes in the authorisation are listed in Table 4. Note that the actual increases are greater than reflected in the limits because a greater proportion of those limits is now being used or planned to be used.

Table 4: BNFL Sellafield - Authorisation Annual Liquid Discharge limits

<i>Radionuclide</i>	<i>Limit TBq/year</i>	
	Previous Authorisation	1994 revision ¹
H-3	3500	31000
C-14	4	20,8
Co-60	8	13
Sr-90	35	48
Zr-95/Nb-95	180	9
Tc-99	10	200
Ru-106	170	63
I-129	0,4	2
Cs-134	10	6,6

Cs-137	110	75
Ce-144	22	8
Pu-alpha	7	0,4
Pu-241	170	27
Am-241	3	0,3

¹ after commissioning of EARP

- 4.6 As expected the discharge figures of the last five years show the increase of important isotopes like C-14, Sr-90, Tc-99, I-129, H-3 and Co-60 in liquid discharges from Sellafield. Table 5 shows the actual amounts from 1991 to 1995⁶.

Table 5: Discharges for some important isotopes in the marine environment 1991-1995

<i>Isotope</i>	<i>1991(TBq)</i>	<i>1992(TBq)</i>	<i>1993(TBq)</i>	<i>1994(TBq)</i>	<i>1995(TBq)</i>
C-14	2,4	0,8	2	8,2	12
Sr-90	4,1	4,2	17,1	28,9	27
Tc-99	3,9	3,2	6,1	72	190
H-3	1883	1199	2310	1680	2700
Co-60	0,09	0,07	0,09	0,11	1,4

- 4.7 It should be noted that no active chemical separation took place in THORP in 1994 and only limited separation took place in 1995. The recent changes in the discharges will therefore be amplified if the throughput of THORP reaches its anticipated levels. Especially the discharges of tritium, C-14 and I-129 are expected to increase significantly when full commissioning of THORP takes place.
- 4.8 Moreover, the processing of 'old' Medium Active Wastes in EARP will continue for at least another ten years. According to BNFL, the 'historical backlog' of Tc-99 on 1 July 1996 amounted to 720 TBq; if discharged uniformly over the projected life of the Magnox programme (about 12-13 years) this would be about 55 TBq/year. Arisings from Magnox reprocessing over the period 1996/97 - 2008/9 have been estimated to give rise to a further 760 TBq technetium-99; this would produce an annual discharge of 45 TBq/year. BNFL conclude that an average annual discharge rate of around 100 TBq could be achieved (although there are uncertainties in the projections of Tc-99 discharges) and have asked for a reduction in the annual limit to 150 TBq/year¹³. Bearing in mind that before 1992 the average discharge of technetium-99 was about 3-5 TBq per year, the proposed reduction in the limit from 200 TBq to 150 TBq seems rather cosmetic when compared to the increase in the level of actual discharge.
- 4.9 In any case, since a fixed amount of technetium-99 remains in storage, it will presumably just be discharged over a longer time period if the limit is reduced. This would not significantly reduce the total amount of radioactivity discharged nor its environmental effects because the half-life of technetium-99 is so long. In addition, Magnox reprocessing is projected to continue and to cause annual

Tc-99 discharges comparable to those arising from the backlog. It is thus clear that the problem is by no means a temporary one.

LA HAGUE

- 4.10 Discharges from La Hague have been somewhat more stable over the past three decades than the Sellafield discharges although this facility also shows large variations of about one order of magnitude (whereas Sellafield discharges vary over more than two orders of magnitude) with peak values during the 70s and early 80s.
- 4.11 As with Sellafield the downward trend from the late 80s stops in the beginning of the 1990s and in fact the total activity discharged into the marine environment is rapidly increasing. This is due to the large increase in throughput of spent fuel at the reprocessing factories at La Hague (UP2 and UP3) and the lack of filtering techniques applied for some principal isotopes.
- 4.12 The operators of the La Hague installations, COGEMA admit that in fact tritium is not filtered at all from the liquid nor gaseous releases. It is therefore not surprising that the releases of this isotope show a significant increase over the last few years⁹.
- 4.13 A similar effect is observed with I-129, where a significant fraction of the I-129, trapped in the spent fuel, is released to the environment. It can be shown that this fraction varies from 40% to almost 90 %^{14 15}. Typical values of 50% are reported by Raisbeck¹⁶.
- 4.14 In fact, when looking at the possibilities to remove I-129 from the waste streams, releasing 40% to 90% of the total into the environment is unacceptable. It represents a breach of the agreement reached within the framework of the Paris convention (1974) and the OSPAR Convention for the Protection of the North East Atlantic (1992) to apply the Best Available Techniques. In its Technical report No. 276 IAEA concludes that removal of I-129 from the waste stream to about 99% is practicable. The Karlsruhe reprocessing plant (WAK) for instance operated with a removal of 99,9% by using silver-absorber filters, which were after use immobilised in cement.⁴⁵
- 4.15 Although COGEMA do not give any figure for C-14 releases to the environment in its reports, the discharged amounts of this isotope are also estimated to rise rapidly in accordance with the spent fuel throughput³. Figure 1 shows the throughput and the discharges of C-14, H-3 and I-129 to the marine environment.

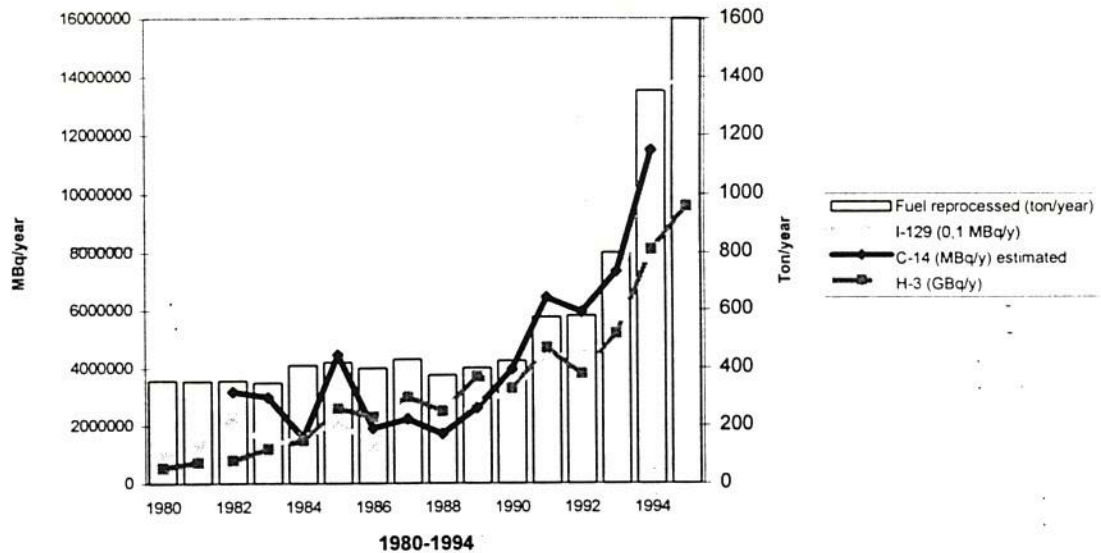


Figure 1: Throughput and discharges of significant isotopes.

DOUNREAY

4.16 This is the third reprocessing facility in Western Europe. Although the operators of Dounreay, UKAEA, like to present it as a 'research' facility (it is even listed as such in the OSPAR submissions made by the UK) amongst other activities, Dounreay commercially reprocesses nuclear fuel, mostly originating from research reactors and containing Highly Enriched Uranium

4.17 Figures for discharges in the last 5 years show a significant increase in discharges of beta emitters and alpha emitters. Beta emitters going up from 4,3 TBq in 1991 to 9 in 1994. The trend in discharges of alpha emitters is shown in Figure 2⁶.

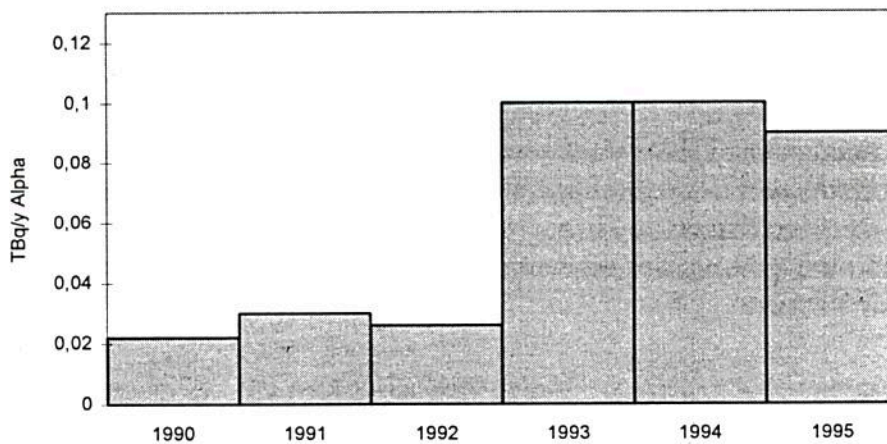


Figure 2: Alpha discharges from Dounreay in recent years

4.18 Moreover the increases of discharges are going to be even more radical when Dounreay is granted the new discharge authorisation that it applied for in 1993. According to UKAEA's own documents the discharges are likely to increase between 3 and 22 times the amounts discharged in the recent past.¹⁷ Table 6 shows some of these projected increases in liquid discharges.

<i>Nuclide</i>	<i>Average 1989-1994 (TBq)</i>	<i>Expected (TBq)</i>	<i>increase</i>
Total alpha	0,053	0,27	
Total Beta	6,6	49	
H-3	1,46	20,5	
Co-60	0,026	0,46	
Sr-90	1,39	7,7	
Cs-137	3,42	23	
Pu-241	0,75	2,3	
Ce-144	0,039	0,42	

Table 6: Average and expected discharges from Dounreay

4.19 If these projected discharges are authorised, the Dounreay releases will for some important isotopes be in the same order of magnitude as those from the Sellafield installations, although Dounreay's throughput is some 200 times lower than that of Sellafield.

OVERVIEW

4.20 To give an overview of recent trends Figure 3 shows the increases in discharges of tritium from La Hague and Sellafield in the last five years. Figure 4 shows the total beta-discharges from all reprocessing facilities and Figure 5 shows the increase of some of the significant isotopes from La Hague and Sellafield.

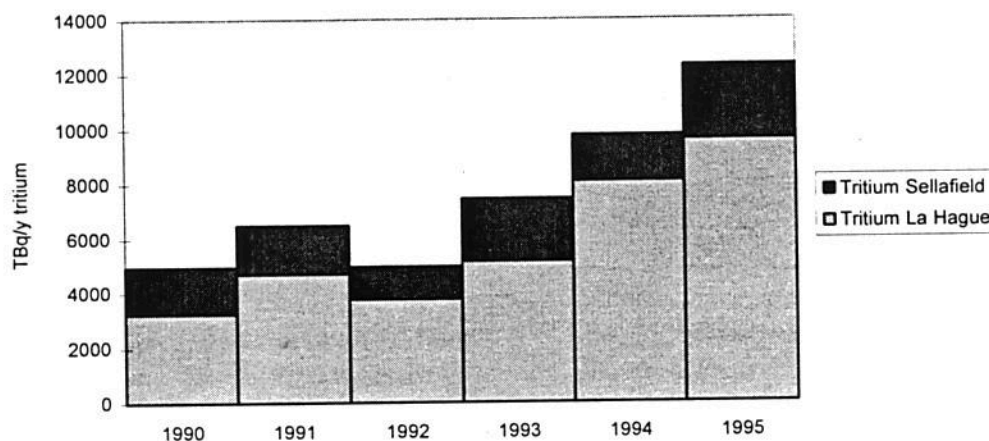


Figure 3: Discharges of tritium by Sellafield and La Hague

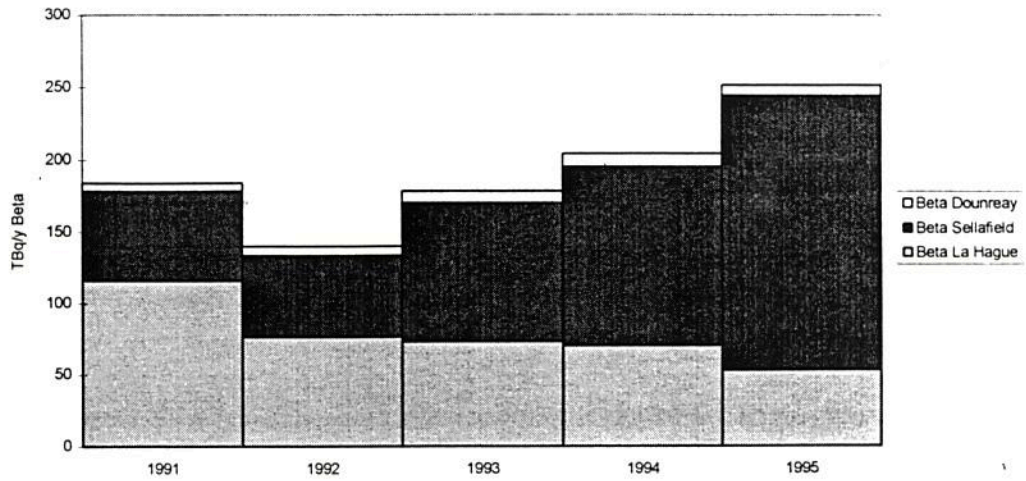


Figure 4: Discharges of Beta activity from reprocessing facilities

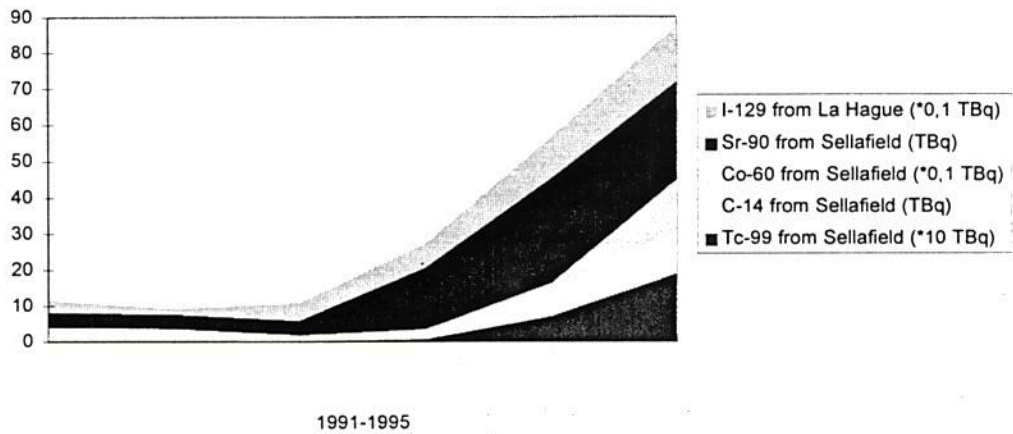


Figure 5: Discharges of some significant isotopes by reprocessing facilities in recent years

5 TRANSPORT PROCESSES OF RADIONUCLIDES FROM LA HAGUE AND SELLAFIELD

SELLAFIELD.

- 5.1 The initial dispersion of radionuclides from Sellafield is influenced by a number of factors including variations in the discharge rate, the chemical form of the radionuclides in the effluent, local hydrographic conditions and the distribution and composition of bottom sediments.
- 5.2 Data collected over two decades show that the soluble elements (Tc-99, C-137, Sr-90 and H-3) are transported principally to the North and the West, leaving the Irish Sea via the North Channel, with a mean transit time of about one year (a much smaller proportion is carried South via St. Georges Channel).
- 5.3 To illustrate, the influx of fresh sea water pushing up through the Irish sea from the South is clearly evident from Cs-137 contamination levels in the water leaving the Irish Sea. It is well established that Caesium discharged from Sellafield mixes with Sea water transported in a northerly direction, exits through the North Channel and follows a well defined path around the West and North coast of Scotland, where it enters the North Sea¹⁸.
- 5.4 Studies of the distributions of Tc-99, Cs-134, Cs-137 and also Pu-238, Pu-239,240 and Am-241 in inshore waters around Ireland carried out in 1985-1986 have shown that the distribution of soluble isotopes are strongly asymmetric with highest concentrations being recorded in the general vicinity of the North-east coast where they approached their maxima just South of Bangor¹⁹.
- 5.5 In contrast, non-soluble nuclides, such as plutonium and americium, are quickly removed from the water by direct precipitation or adsorption on suspended particular matter. Thus, the behaviour and distribution of these nuclides is closely linked to that of the finer seabed sediments. The mechanism by which these sediment-bound nuclides are subsequently dispersed are complex and not yet fully understood.
- 5.6 However it is clear that nuclides associated with sediments can be transported by the sediment flow near the seabed. In the North-Eastern Irish sea, this flow is predominantly northward and results in the accumulation of transuranics in estuaries along the Cumbrian coast and the South-West coast of Scotland²⁰. Indeed, it has been shown that by the mid-1980s, the movement of contaminated silt as the dominant mechanism of supply of Sellafield-discharges Plutonium to the South-West coast of Scotland²¹.
- 5.7 Resuspension of contaminated sediments can also result in transport within the water column as suspended particulate. Moreover resolubilization and subsequent solution transport can play a significant role in the dispersion of previously sediment-bound radionuclides²².

5.8 As a result of these processes, small fractions of the discharged plutonium and americium have been transported over considerable distances e.g. the Norwegian Sea²³. Studies, conducted on soil samples taken in West Cumbria, UK, since the late 1970s, have shown that a part of the Pu and Am discharges into the Irish Sea from Sellafield has been transferred into the atmosphere and returned to land²⁴. Preliminary estimates have been made which suggest for instance that 40-80 GBq of excess Pu had been deposited in a coastal strip approximately 5 km wide and 40 km long by the year 1980 and an identical effect has been observed along the North-east coast of Ireland²⁵. These sediment transports were not predicted when the discharge authorisations were made.

LA HAGUE

5.9 La Hague releases are transported Eastwards forming a characteristic distribution pattern in the Channel and the Southern North Sea. This includes a near-coastal "plume" and a distinct boundary between waters contaminated by La Hague and by Sellafield.

5.10 Generally there is a northerly-bound coastal current from the English Channel along the French, Belgian, Dutch, German and Danish coasts. En route the coastal current receives a significant part of the aquatic discharges from continental Europe via major river outputs such as the Seine, the Rhine, the Weser and the Elbe. As a result of transport processes, the radionuclide contamination of inshore waters along the North-West European continental coast is, in general, found to be higher than that of offshore waters. Typical transit times have been estimated from La Hague to the Straits of Dover (4-7 months), to the German Bight (11-14 Months), Danish waters (15-20 months) and to Utsira on the Norwegian SW coast (17-18 Months)^{26 27 28}.

5.11 Continuous monitoring has revealed peak values of contamination in the environment to be linked with peaks in discharges from La Hague and with fluctuations in water flow rates through the English Channel.^{26 29}

5.12 In contrast with the relatively extensive studies on movements of soluble nuclides, very little is known about the contamination of sediments with radioactive nuclides and how these sediments move. Nevertheless new research on plutonium in sediment samples in the Roads of Cherbourg shows striking results. The ratio of Pu-238 and Pu-239,240 show a remarkable increase from the early 70s up to 1990. This is consistent with the increase of the Pu isotopic ratios in the release of the La Hague reprocessing plant since it started.³⁰ Transit times on the sediments throughout the Eastern Channel range ca. 10 to 50 years, which are to be compared to 4-8 months for the water masses.

5.13 In these conditions, it is not surprising that the sediment Pu inventory is of the same order of magnitude as the Pu cumulated release from the La Hague plants. In other words, most of the Pu of industrial origin remains on the seabed of the

Channel where it remains a slow but stable source of contamination of the water column³⁰.

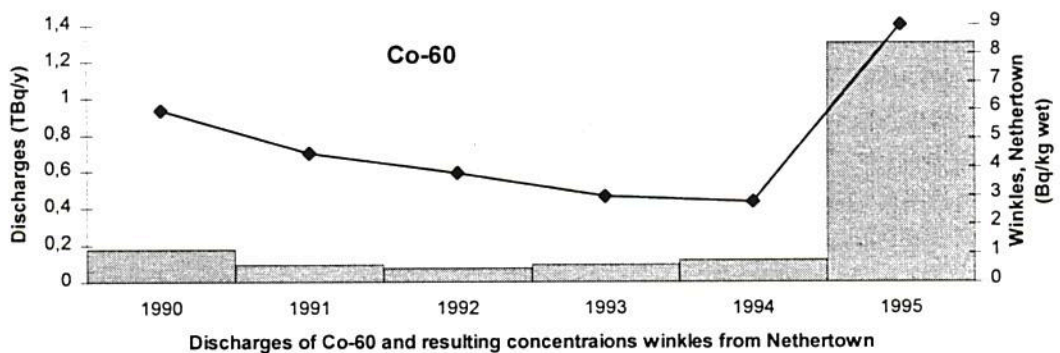
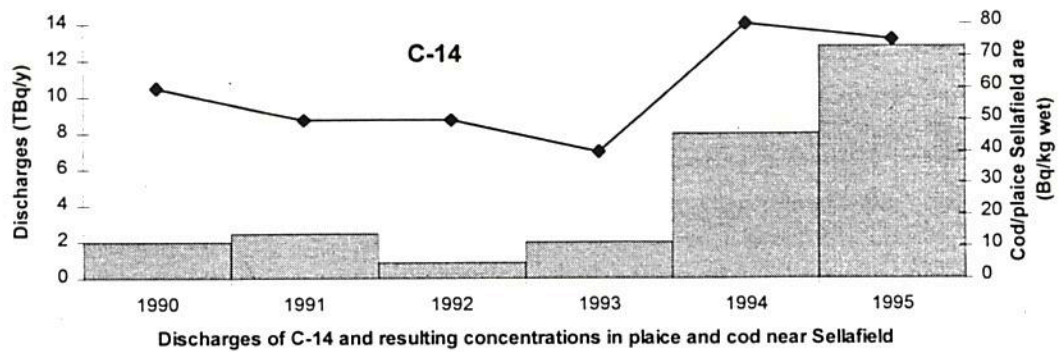
FINALLY: DISPERSION TO THE NORDIC WATERS

- 5.14 The currents from the La Hague, Dounreay and Sellafield installations all flow into a Northerly direction towards Greenland and the Barents Sea and even further into the Nordic waters. Reprocessing is now the single largest source of radioactivity throughout the complete Arctic Oceans: for instance its caesium input in this area is between two and three times more than the input originating from the Chernobyl fall-out.³¹

6 RECENT CHANGES IN CONTAMINATION LEVELS OF WATER AND BIOTA IN THE ENGLISH CHANNEL, IRISH SEA AND NORTH SEA.

IRISH SEA

- 6.1 Contamination levels in the Irish Sea used to be largely dominated by the very high discharge levels of caesium, ruthenium and actinides in the 70s and 80s from Sellafield. With the decrease of the releases of these isotopes, declining contamination levels could be observed.
- 6.2 However, as expected, the recent increase in the release of different isotopes now start to have a significant effect on the marine environment. New research reveals that the contamination levels of biota in the vicinity of the Sellafield outlet (distance between 5 and 15 km) are affected by these radiologically significant isotopes that now become more dominant in the Sellafield liquid discharges.



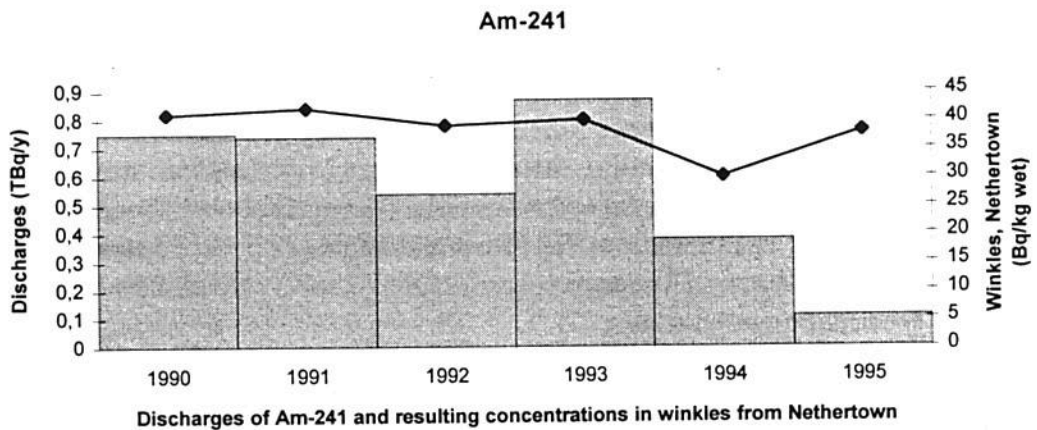
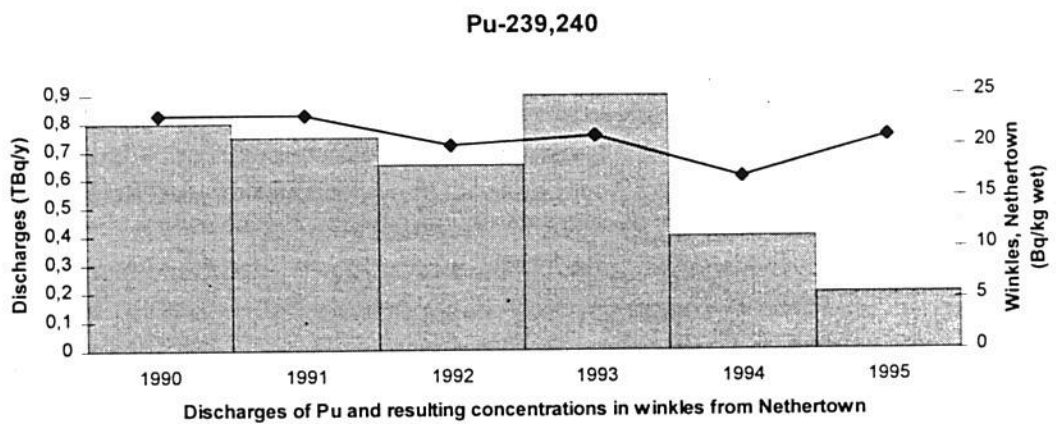
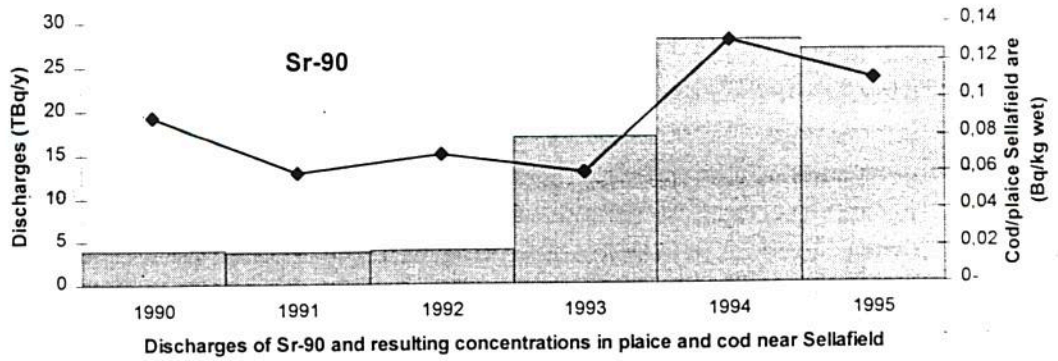


Figure 6: Impact of recent discharges on biota in Irish Sea¹²

6.3 As can be seen from Figure 6 the contamination levels of Pu and Am in winkles do not decrease as a result of the decreasing discharges. This is consistent with the known adsorption of these radionuclides on sediments. The contaminated

sediments now act as a continuous source of Pu and Am due to resuspension. A decrease in contamination levels in biota is therefore not anticipated in the coming years.

- 6.4 The C-14 levels in cod and plaice near Sellafield have been corrected for the known natural levels (approx. 23 Bq/kg). The real impact of the C-14 discharges on species of fish and shellfish is still unclear as environmental sampling fluctuations result in some discrepancies between discharges and contamination levels. The picture may become clearer in the coming years when even higher discharge levels are anticipated.¹²
- 6.5 Technetium-99 forms a special case when looking at contamination levels in the Irish Sea. Tc-99 is a long lived radionuclide formed in significant quantities in the fission process of nuclear reactors. It emits β -radiation and has a half life of 213.000 years.
- 6.6 The disposal of Tc-99 therefore causes long term environmental problems that must be addressed with a detailed understanding of its environmental behaviour. Until recently, Tc-99 has been present in low concentrations in most environmental materials and this factor, combined with difficulties inherent in its measurement, has resulted in few data being currently available. Despite this lack in understanding of the behaviour of Tc-99, Sellafield has increased its Technetium discharges by two orders of magnitude. Over the period 1981-1993 discharges of Tc-99 were approximately 3-6 TBq/y. After the commissioning of EARP in 1994 the level raised to 72 TBq in 1984 and to 190 TBq in 1995.
- 6.7 It is only after these discharges occurred that comprehensive studies have started to assess the impact of Tc-99 on the environment. Recent studies however already show some remarkable results indicating that the radiological impact from Tc-99 might be higher than expected.
- 6.8 It is shown for instance that the uptake of Tc-99 by mussels and winkles is a factor of 10 to 100 greater than formerly estimated. Laboratory studies led to an estimation of concentration factors of about 45 for winkles and of 1 to 2 for mussels^{32 33} while recent environmental research showed concentration factors for winkles and mussels in the Ravenglass area (12,5 km South of Sellafield outlet) of 640 and 330 respectively. Table 7 shows the results of these recent investigation on molluscs³⁴.

Table 7: Tc-99 activity and concentration factors in molluscs

<i>Date</i>	<i>location</i>	<i>Sample</i>	<i>Tc-99 activity</i>	<i>Conc. Factor</i>
Jun-95	St. Bees	Winkles	210	390
		Mussels	230	420
Jun-95	Ravenglass	Winkles	770	640
		Mussels	390	330
Mar-95	Off Sellafield	Whelks	350	150

- 6.9 A similar result is obtained for the concentration factors of Tc-99 in seaweed. Figure 7 shows the impact of the Tc-99 discharges on the other side of the Irish Sea.

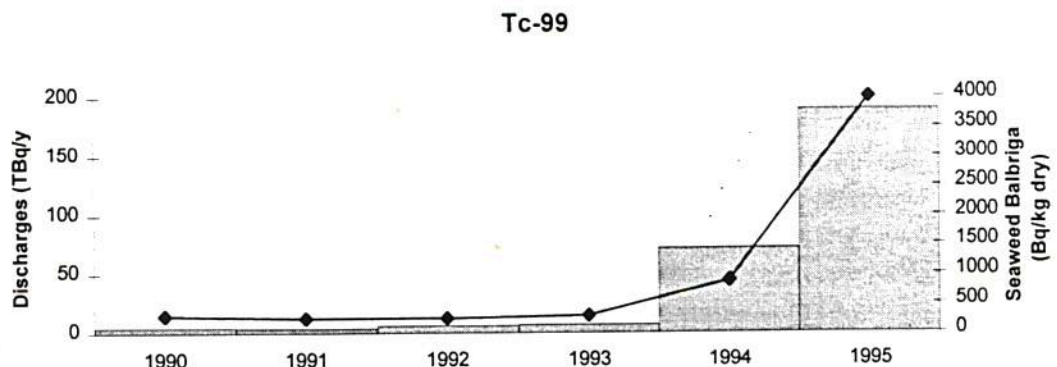


Figure 7: Discharges of Tc-99 and resulting concentrations in seaweed from Balbrigan (Irish Coast)³⁵

- 6.10 The results of a recent study in the Irish Sea reveal that the laboratory values are generally much lower than the values based on field-studies. The authors conclude that: *"The results of this study confirm that the uptake of Tc-99 by marine organisms far exceed that which would be expected from laboratory studies. Given the potential radiological significance of this nuclide (it was the largest contributor (47%) to the committed effective dose of 0,06 mSv/y to the Sellafield critical group of high seafood consumers due to discharges in 1994 and is also expected to be a radiologically important constituent of high level waste) it would seem desirable to obtain more accurate information on the environmental behaviour of this nuclide"*.³⁶
- 6.11 It can only be concluded that, far from the regulators claim that these discharges are harmless, the discharge of radionuclides is being allowed as part of a massive experiment. Indeed a UK government report in 1992 states that: *"The likely increased discharge of Technetium-99 from Sellafield... from 1993 onwards will provide an opportunity to use this tracer to study water movements, and validate transfer models, within the Irish Sea, and exchange in the North Channel, Malin Shelf and along the Scottish coastal current"*³⁷

NORTH SEA

- 6.12 In the past the main source of radioactivity in the North Sea has corresponded to the releases of Cs-137 from Sellafield, with the maximum impact being observed between 1975 and 1985. Data collected throughout the North Sea show that the zone influenced by the influx of Irish Sea with contamination from Sellafield plays an important role over most of the North Sea³⁸.

- 6.13 In the remaining parts of the North Sea, corresponding to inshore waters between the Netherlands and Denmark, the currents from the Irish Sea play a less dominant role and the activities due to Sellafield are clearly lower. However the plume of English Channel waters is known to run along this coastal belt and therefore affects the contamination levels of these waters.
- 6.14 These waters are characterised by specific radionuclides present in the La Hague discharges, with tritium playing a dominant role. It is shown that tritium contamination in the German Bight is rapidly increasing. The initial value of contamination was 1,9 Bq/l in 1980 and decreased to 1,1 Bq/l in 1992. After 1992 the amount of tritium increased rapidly and now shows average contamination levels of 2,8 Bq/l, significantly higher than the 1980 value ³⁹.
- 6.15 It is well known from previous studies that the majority of the radionuclides from Sellafield and La Hague come together in the North Sea, near the Southern coast of Norway and are then trapped into the Arctic seas via the Norwegian Coastal Current ^{40,41}. Therefore the I-129 and Tc-99 'signals' coming from La Hague and Sellafield respectively will be of significant importance to these waters.
- 6.16 Since the discharges of I-129 have been relatively constant from 1975 to 1990 and increased rapidly from 1990 onwards, the next few years will show a rapid increase in the I-129 levels in the currents north of the North Sea.
- 6.17 Similarly Dahlgard concludes in a report on Tc-99 and Cs-137 in algae at the Norwegian coast that "a very distinct pulse of Tc-99 is now underway from Sellafield to the Norwegian coast".⁴²

ENGLISH CHANNEL

- 6.18 Waters from the English channel are largely affected by the La Hague reprocessing installations which augmented their discharges drastically in the recent years due to increased throughput of spent fuel. (see para 4.13 above) I-129 and tritium discharges are increasing rapidly.
- 6.19 I-129 is a radioactive isotope with a half -life of 16 Million years. Before the nuclear era, when I-129 on Earth was principally due to spontaneous fission of U-238 and cosmic ray induced spallation of Xenon in the atmosphere, the ratio of I-129/I in the ocean is estimated to have been 10^{-12} ⁴³. Since the development of nuclear programmes, anthropogenic releases of I-129, principally from nuclear reactor operations and bomb explosions, overwhelmingly dominates natural production. The total release of I-129 in nuclear testing is estimated to have been $2 \cdot 10^{26}$ atoms ⁴⁴, from which the resulting ratio of I-129/I in the oceans would be $2 \cdot 10^{-11}$, i.e. a twenty fold increase above natural levels.
- 6.20 Beginning in 1990 when a new plant was put into operation at La Hague the discharges of I-129 and the measured ratios of I-129/I in seaweed both

increased. Figure 8 shows the amounts of I-129 discharges and the measured ratio at Goury a few miles from the outlet of La Hague¹⁴

- 6.21 I-129 is of particular concern because it has been identified as one of the isotopes most likely to cause radiation exposure to large numbers of people. This results from its long half-life, its ready dispersion and transport in air and water and the possibility of biological concentration.⁴⁵ See also chapter 7.
- 6.22 It should be noted that COGEMA does not provide any information on I-129 levels in the environment in their reports.⁴⁶

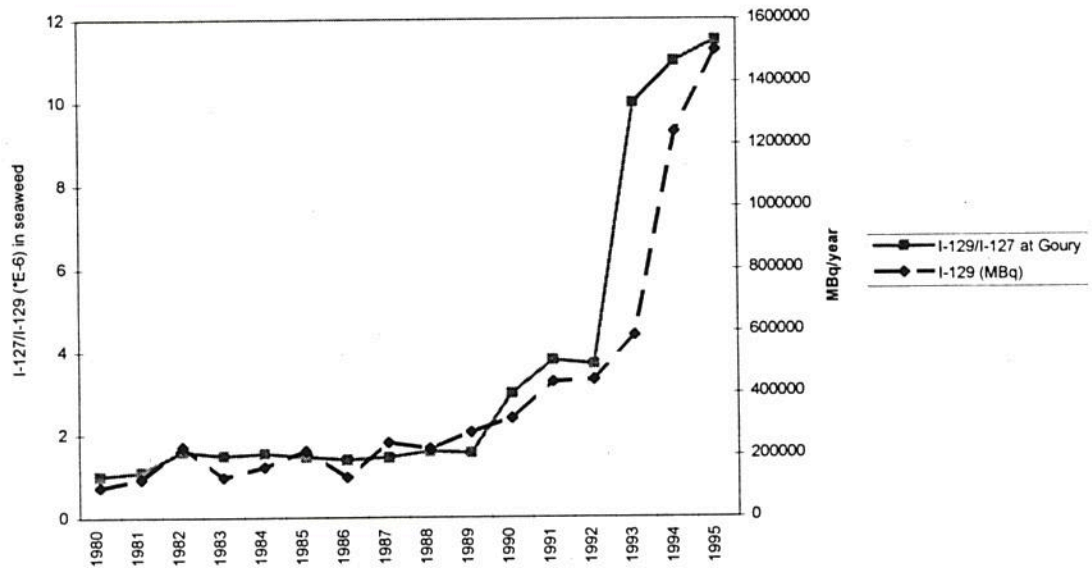


Figure 8: I-129/I ratio in seaweed from Goury, quantity of fuel reprocessed at La Hague, and marine discharges of I-129 based on official data and estimates (1975-1983)

- 6.23 There is very little data available on tritium contamination in the English Channel. It is obvious that increased discharges have led to a significant increase in contamination levels which is reflected by the tritium levels in the German Bight in the North-East part of the North Sea (6.14). In fact the actual contamination of waters near the outlet of La Hague are orders of magnitudes higher than in other parts of the Channel and the North Sea.

7. RADIOLOGICAL CONSEQUENCES

- 7.1 Since the increases of radioactive inputs in the European waters have just begun, it is impossible now to undertake a detailed assessment of their actual long term consequences. The best scientific understanding, though, provides sufficient insight on this issue to justify an *a priori* conclusion that these increases may well have an effect on human and environment. In these circumstances, continued discharges amount to a massive irreversible experiment on the environment. For this reason alone the precautionary principle should be applied.
- 7.2 Moreover, based on the large amount of data which is available on radiological consequences of specific isotopes in the marine environment, an estimate of the possible consequences can be done.
- 7.3 The recent increases in discharges from La Hague and Sellafield are particularly important from a radiological point of view since some of the isotopes concerned are of special significance for the individual dose and collective dose impacts from reprocessing (I-129, Tc-99, C-14, Co-60).
- 7.4 Tc-99 discharges from Sellafield were in 1994 by far the largest contributor to the individual dose to critical groups (seafood consumers in Cumbria) with 47% of the 0,06 mSv/y in 1994³². The near three fold increase of Tc-99 discharges in 1995 is therefore expected to have an even larger contribution (60%-70%) to these individuals, significantly raising the doses suffered due to the liquid discharges of Sellafield.
- 7.5 It is also remarkable that the other isotopes which play a dominant role in the total effective dose to critical groups (C-14 and Co-60) are the ones that have been increased significantly in the discharges of recent years. The individual critical group dose due to C-14 discharges was increased from 5 μ Sv/y in 1990 to 35 μ Sv/y in 1995 and is now with Tc-99 the largest contributor to the total dose.
- 7.6 A similar conclusion can be drawn when the recent trends are considered with respect to the collective dose to the world population. The C-14 in the discharge effluents is responsible for a large part of the *collective* doses to the world population in the next 500 years (96%) due to the liquid discharges from Sellafield.⁵ An increase in the discharge of this isotope is therefore almost linearly reflected in an increase in the projected collective dose. Increasing the C-14 discharges will inevitably raise the total radiological burden of the facility considerably.
- 7.7 At La Hague the collective doses calculated over the next 500 years to the local and regional population by consumption of seafood and other pathways are dominated by C-14 discharges, contributing more than 50% to these doses. As COGEMA gives no data for C-14 in its liquid effluents, it is derived from the C-14 amounts in the gaseous discharges. This C-14 amount in the gaseous

discharged is estimated, based on the assumption that it is a fixed fraction of the Kr-85 releases in the gaseous effluents. Without the official data from COGEMA its difficult to make an assessment on C-14, but given the recent increases in Kr-85 discharges in the gaseous releases, it is very likely that the total C-14 activity in liquid releases is also raising significantly³. This increase has a large effect on the collective dose to the regional population. It is justified to conclude that any increase in C-14 discharges is to be avoided.

- 7.8 For the collective dose to the global population integrated over 100,000 years the models for COGEMA take only a few isotopes into account, with the rest having a relatively very small impact according to those models. These few isotopes are H-3, Kr-85, I-129 and C-14. It is notable that exactly these isotopes have shown significant increases in the last few years. Figure 9 shows the resulting increase in the projected global collective dose due to the increase in these isotopes

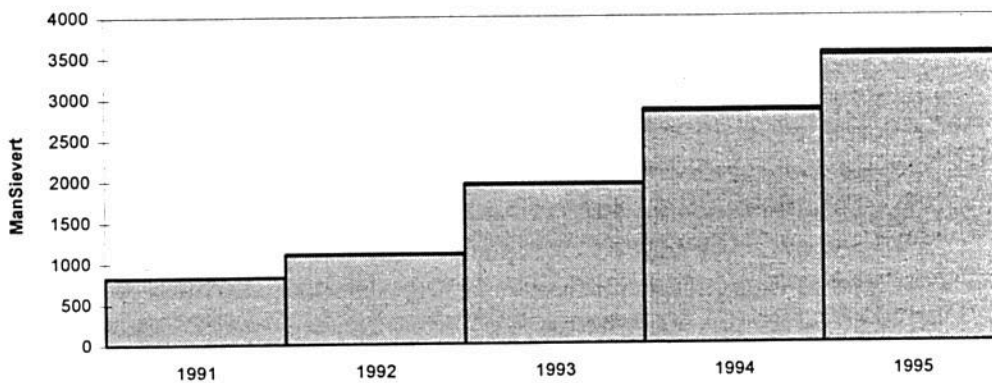


Figure 9: Projected collective dose (integration time 100,000 y) from La Hague discharges

- 7.9 In the light of the available data on discharges and their expected dose implications it is clear that the radiological impact on human health and the environment of reprocessing is by far the largest of all elements in the nuclear fuel cycle. It is therefore not surprising that the OSPAR Convention area (plus the Baltic), e.g. the North East Atlantic, in which the biggest reprocessing facilities are located, clearly stands out as the ocean area giving the highest collective doses in the world from the consumption of seafood containing man-made radionuclides.⁴⁷
- 7.10 These radiological consequences will undoubtedly have their impact on human health. In the last few decades a lot of epidemiological research has been conducted on the effects of radiation and it is now scientifically understood that at low doses of radiation (below 100 (mSv)) the linear-no-threshold-theory is the only acceptable tool to describe detrimental effects of radiation to human populations⁴⁸. This means that the currently occurring increase in discharges from reprocessing facilities will result in a dramatic increase in radiation induced cancer mortalities and probable environmental impacts. With the

current knowledge of radiation risk factors the collective dose due to La Hague's discharges for the year 1995 can be expected to result in more than 170 cancer fatalities.

- 7.11 Indications for harmful radiation effects to local population around the reprocessing facilities are described in the scientific literature. All three reprocessing facilities - at Dounreay, La Hague and Sellafield have leukaemia clusters nearby, and the most recent study at La Hague has concluded that there is statistical evidence for a causal link⁴⁹⁵⁰⁵¹. It can be shown simply that the link between radioactive discharges and these effects is unlikely to be proven with epidemiological methods, because of the complex mechanism behind cancer-induction and the relative high yield of cancer, due to other causes than radiation. To dismiss these indications as 'not proven and thus not valid' is therefore scientific nonsense. Further research into these indications is urgently needed but its completion should in no way be used to block progress on reducing discharges.
- 7.12 As reflected above in this section, it is mostly the effects of radiation to humans that are taken into account. However, the detrimental effects or damage to the environment are likely to be far greater than on humans, due to the fact that the environment is *directly* affected by the discharges. The surroundings of the outlets from reprocessing facilities are for instance submitted to relative very high doses of radiation and radioactive contamination far beyond levels regarded as 'acceptable'. Doses are estimated to reach as much as 10 - 100 Gy per year, a dose area classified as having 'dramatic irreversible detrimental effect on the marine ecosystem'⁵². Ecosystem impacts of radioactive discharges have not been systematically or comprehensively evaluated to-date.

8. CONCLUSIONS

- 8.1 Reprocessing is by far the largest contributor to the total radioactive discharges in the North East Atlantic. In absolute terms reprocessing releases have attributed 97,4 % of the total site inputs of alpha emitters and 98,6 % of the Beta emitters. These discharges are clearly avoidable by adopting other waste management techniques or options, in particular dry storage of spent fuel.
- 8.2 The reprocessing industry has repeatedly stated that it is effectively reducing its discharges. In fact the opposite is true. Since the beginning of the 90s the liquid discharges of reprocessing facilities have increased significantly, in terms of released activity as well as in terms of their radiological impact on human health and potential impacts on marine ecosystems.
- 8.3 This increase is inconsistent with the legal obligation set out in the Paris (1974) and OSPAR (1992) conventions to *"take all possible steps to reduce and eliminate pollution and [to] take the necessary measures to protect the [North East Atlantic] against the adverse effects of human activities so as to safeguard human health and to conserve marine ecosystems"* and the recognised *"need to reduce radioactive discharges from nuclear installations to the marine environment"*. The on-going reprocessing discharges of at least some of the most persistent isotopes is also inconsistent with the legal obligation set out in the OSPAR convention to apply the Best Available Techniques (BAT).
- 8.4 In the Irish Sea, the impact of the increased releases from Sellafield is clearly measurable in biota, with for example a massive increase in Tc-contamination levels in several species. At La Hague the increase of I-129 discharges is clearly visible, for example, in contamination levels of this radiologically significant isotope in algae.
- 8.5 The unexpected increase in Tc-99 concentrations in lobsters in the Irish Sea is a clear example of the shortcoming of laboratory-values for Concentration Factors. It shows how dangerous it is to set discharge limits based on such limited knowledge of the environmental behaviour of nuclides.
- 8.6 By the use of Best Available Techniques the unprecedented increase in I-129 releases from La Hague is clearly avoidable in terms of practicable techniques to filter I-129 from the waste streams. Absent the use of such available filtering techniques, La Hague should stop its activities immediately, in line with France's legal obligation under OSPAR.
- 8.7 The increase of radioactive discharges are expected to cause adverse health effects both to people living near the plants and worldwide. The recent increases in routine liquid discharges from reprocessing facilities are some of the worst possible in terms of the radiological burden on humans and the environment.

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- ¹ OSPAR, art. 2.1 (a) of the OSPAR convention for the protection of the NE Atlantic.
 - ² OSPAR (1992) Final Declaration of the Ministerial Meeting of the OSPAR commission, Part III.
 - ³ M.Dreicer et al., The Nuclear Fuel Cycle, Estimation of physical impacts and monetary valuation for priority pathways, CEPN, 1995
 - ⁴ C. Macic, From reactor to waste disposal: the back end of the nuclear fuel cycle with emphasis on France, Rad. Prot. Dos., 26, 174, 1989, p 15-22
 - ⁵ BNFL, Application for a variation to the certificate of Authorisation for the disposal of low level liquid waste from the marine pipeline at the Sellafield Site, 1996
 - ⁶ Draft Annual report on Liquid discharges from nuclear installations, 1995, RAD 97/12/1, Draft Annual report on Discharges from nuclear installations, 1995, PRAM 97/6/4, Corrections to the draft annual report on Discharges from nuclear Installations, 1995, PRAM 97/6/4 add 1.; Further corrections to the Draft Annual report on Discharges from nuclear installations, 1995, PRAM 97/6/4 add 2.
 - ⁷ F. Nyffeler et al (1996) The atlantic ocean, Radionuclides in the oceans, Les Edition de physique, 1-28
 - ⁸ Implementation report on OSPAR recommendation 91/4, Submitted by the UK, RAD 96/5/5
 - ⁹ Implementation report on OSPAR recommendation 91/4, Submitted by France, RAD 97/5/3
 - ¹⁰ IAEA (1995) The principles of Radioactive waste management. Safety Series 111-F.
 - ¹¹ P.J. Kershaw et al (1992) A review of radioactivity in the Irish Sea, Aquatic Environment monitoring report, 32, 65 pp
 - ¹² G.J. Hunt (1997) Recent changes in Liquid radioactive waste discharges to the Irish Sea from Sellafield, Radioprotection Colloques, 32 17-22
 - ¹³ BNFL (December 1996) Application for a Variation to the Certificate of Authorisation for the Disposal of Low Level Liquid Waste from the Marine Pipeline at the Sellafield Site.
 - ¹⁴ G.M. Raisbeck et al (1997) Marine Discharges of I-129 by the nuclear reprocessing facilities of La Hague and Sellafield, Radioprotection - Colloques, 32, C2, 91- 95
 - ¹⁵ R. Castaign et al (1984) Du groupe de travail sur les recherches et developpements en matière de gestions des rejets radioactifs, conseil supérieur de la sûreté nucléaire, étude pour le Ministère du redéploiement industriel et du commerce extérieur.
 - ¹⁶ G.M. Raisbeck et al (1995). I-129 from nuclear reprocessing facilities at Sellafield and La Hague. J. Marine. Syst. 6, 561-570
 - ¹⁷ UKAEA (1995) Further information in support of an application for authorisation for the disposal of radioactive wastes from AEA-Technology Dounreay.
 - ¹⁸ P.I. Mitchell et al. (1996) Radionuclides in the Oceans, Les Edition de physique, 155-175
 - ¹⁹ P.I. Mitchell et al (1989) The impact on Irish coastal waters of long-lived radioactive waste discharges to the Irish Sea, The Irish Sea - a resource at Risk, geographical society of Ireland, Special Publication 3, 124-147

-
- ²⁰ P.J. Kershaw et al. (1984) The incorporation of Plutonium, Americium and curium into the Irish Sea seabed by biological activity. *Sci. Total. Environ.* **40**, 61-81
- ²¹ P. Mc Donald et al. (1990) Radionuclide transfer from Sellafield to south-west Scotland. *J. Environ. Radioactivity*, **12**, 285-298
- ²² G.J. Hunt and P.J. Kershaw (1990) remobilisation of artificial radionuclides from the sediments of the Irish Sea. *J. Rad. Prot.*, **10**, 147-151
- ²³ P.J. Kershaw and A. Baxter (1995) The transfer of reprocessing wastes from North-west Europe to the Arctic. *Deep Sea Res. II*, **6**, 1413-1448
- ²⁴ D.H. Peirson et al (1982), Environmental radioactivity in Cumbria. *Nature*, **300**, 27-31
- ²⁵ J.A. Garland et al (1989) Artificial radioactivity on the coasts of Northern Ireland, Report DOE/RW.89.055
- ²⁶ P. Guegueniat et al., Radiotracers measurements: a contribution to the dynamics of the English Channel and North Sea, *J. Mar. Syst*, **6**, 1995, p 483-494
- ²⁷ H. Dahlgard et al., A traces study of the transport of coastal water from the English Channel through the German Bight to the Kattegat, *J. Mar. Syst*, **6**, 1995, p. 571-578
- ²⁸ M. Masson et al., Time series for sea water and seaweed of Tc-99 and Sb- 125 originating from releases at La Hague, *J. Mar. Sys.*, **6**, 297-413, 1995
- ²⁹ P. Bailly du Bois et al., A quantitative estimate of English Channel water fluxes into the North Sea from 1987 to 1992 based on radiotracer distribution, *J. Mar. Sys.*, **6**, 1995, p. 457-481
- ³⁰ D. Boust et al (1997) Distribution and transit times of plutonium bearing particles Throughout the Channel, *Radioprotection - Colloques*, **32**, C2, 123-128
- ³¹ P. Strand et al., The Arctic, Radioanuclides in the Oceans, les Editions de Physique, 95-119
- ³² D.J. Swift (1989), *Journal of Environmental Radioactivity*, **9**, 31-52
- ³³ S.W. Fowler et al (1981) Impacts of Radionuclide releases into the marine environment. IAEA-Vienna, 319-339
- ³⁴ R. Busby et al (1997) Technetium concentration factors in Cumbrian seafood. *Radioprotection - Colloques*, **32**, C2, 311-316
- ³⁵ V. Smtih et al (1997) Temporal and geographical distributions of Tc-99 in inshore water around Ireland following increased discharges from Sellafield, *Radioprotection - Colloques*, **32**, C2, 71-77.
- ³⁶ M. Mc Cartneu and K Rajendran (1997), Tc-99 in the Irish Sea, recent trends. *Radioprotection - Colloques*, **32**, C2, 119
- ³⁷ PJ Kershaw, RJ Pentreath DS Woodhead, and GJ Hunt. A review of radioactivity in the Irish Sea. MAFF, 1992
- ³⁸ P Guegueniat et al (1996) Artificial Radioactivity in the English Channel and the North Sea, Radionuclides in the Oceans, Les Edition de physique, 121-154.
- ³⁹ C. Wedekind (1997) Tritium in the German bight 1980 to 1995. *Radioprotection - Colloques*, **32**, C2, 119

-
- ⁴⁰ H. Dahlgard. (1993) Anthropogenic radioactivity in the Arctic seas: time trends and present levels. New research and policy priorities in the arctic and north Atlantic, 49-63
- ⁴¹ P. J. Kershaw and A.J. Baxter (1993) Sellafield as a source of radioactivity to the Barents Sea. New research and policy priorities in the arctic and north Atlantic, 91-104
- ⁴² H. Dahlgard et al (1997) Technetium-99 and Cesium-137 timeseries at Norwegian Coast monitored by the brown algae *Fucus Vesiculosus*. Radioprotection - Colloques, **32**, C2, 353-358
- ⁴³ Kilius et al. (1990) Nuclear Instrum. and Methods., **b52**, 357-365
- ⁴⁴ Chamberlain et al. (1991) Radioactive Aerosols, Cambridge university press.
- ⁴⁵ IAEA (1987) Treatment, conditioning and disposal of Iodine 129, Technical reports series no 276.
- ⁴⁶ COGEMA (1996) Surveillance de l'environnement
- ⁴⁷ OSPAR (1997) Report summarising the most important information on radioactive discharges to, and contamination with radioactive substances in, Different marine compartments, RAD 97/6/3-E
- ⁴⁸ United Nations (1994) Sources and effects of Ionising Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, Report to the General Assembly.
- ⁴⁹ D. Pobel; and J.F. Viel (1997) Case-control study of leukaemia among young people near La Hague nuclear reprocessing plant: the environmental hypothesis revisited. *BMJ*, **314**, 101-106
- ⁵⁰ M.J. Gardner et al. (1984) Mortality in Cumberland during 1959-1984 with reference to cancer around young people around Windscale. *Lancet*, 217-218
- ⁵¹ JD Urquhart et al. (1991) Case-control study of leukaemia and non Hodgkin's lymphoma in children in Caithness near the Dounreay nuclear installation., *BMJ*, **302**, 687-692
- ⁵² V.N. Lystsov (1997) Marine ecosystems: how clean is clean enough ?, oral presentation at the international symposium "radionuclides in the oceans", Norwich 7-11 April 1997.