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Radioactivity in the Marine Environment 2008 and 2009

Results from the Norwegian Marine Monitoring Programme (RAME)





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Key words:

Radioactivity, marine environment, RAME, monitoring, Norway

Abstract:

This report presents results of monitoring of radioactivity in seawater and biota collected along the Barents Sea, the Norwegian coast, the North Sea and the Skagerrak in 2008 and 2009. An overview of discharges from Norwegian sources and data concerning the long-range transport of radionuclides from European nuclear facilities is included.

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Emneord:

Radioaktivitet, marin miljø, RAME, overvåking, Norge

Resymé:

Rapporten inneholder resultater fra overvåkningen av radioaktivitet i sjøvann og biota i 2008 og 2009 langs norskekysten og i Barentshavet, Nordsjøen og Skagerrak. En oversikt over utslipp fra norske kilder og utslippsdata fra europeiske nukleære anlegg som er relevante for langtransport av radioaktivitet til norske havområder er inkludert i rapporten.

Head of project: Torbjörn Gäfvert *Approved*:

Per Strand, director, Department for Emergency Preparedness and Environmental Radioactivity.

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Radioactivity in the Marine Environment 2008 and 2009

Results from the Norwegian Marine Monitoring Programme (RAME)

Statens strålevern

Norwegian Radiation Protection Authority Østerås, 2011

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1 Introduction

The issue of present and potential radioactive contamination in the marine environment has received considerable attention in Norway. In the late 1980s several accidents and incidents nuclear-powered involving submarines. demonstrated that the risk of the release of radionuclides into the Barents Sea should be considered more carefully. In particular, it became evident that better documentation concerning the radioactivity levels in fish and other seafood was important for the seafood export industries. Furthermore, in the early 1990s, information concerning the dumping of nuclear waste emerged through bilateral environmental cooperation between Norway and Russia. In the years that followed, concern grew regarding the safety of military and civil nuclear installations in the northwest of Russia. This concern was associated not only with possible reactor accidents, but also with the prolonged or sudden release of radio-nuclides from radioactive waste facilities.

In addition to the potential threats outlined above, radionuclides originating from nuclear weapons fallout, the Chernobyl accident and waste discharged from European reprocessing facilities have been detected in the Norwegian marine environment. In 1994 and 1995, the discharge of ⁹⁹Tc from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced, it continued at a high level up to 2003. There has been much public concern about the consequences of such kinds of release, as the radionuclides discharged to the Irish Sea are transported by ocean currents via the North Sea into the Norwegian coastal current and to the Barents Sea. In response to this concern, programmes for the monitoring of radioactivity in the marine environment have been established. Due to the economic importance of the fishing industry and its vulnerability to contamination, as well as any rumours of radioactive contamination, one of the main objectives of these programmes is to document levels and trends of radionuclides in the Norwegian marine environment.

Other industrial activities, such as mining and oil production, may change the distribution of

naturally occurring radionuclides in the environment. The discharge of radium from water produced by oil installations is one area that has received special attention.

Recently, there have been an increased number of transports involving spent nuclear fuel in Norwegian marine waters. Spent nuclear fuel is shipped to Murmansk for further transport and processing at Mayak. Accidents during these transports may lead to releases of radioactivity in the marine envirionment.

The marine monitoring programme (RAME) is funded by the Ministry of the Environment and focus on monitoring of radioactivity both in coastal areas and in the open sea. The marine monitoring programme also includes the compilation of discharge data from Norwegian sources, in addition to the collection of discharge data relevant for the long-range transport of radionuclides from various sources. Liquid discharge data for 2008 and 2009 from nuclear installations are summarised in Chapter 2.

During 2008 and 2009, samples for monitoring radioactivity in the marine environment were collected mainly in the Barents Sea, the North Sea, the Skagerrak and at permanent coastal stations along the Norwegian coastline. Results from the analysis of these samples are presented in Chapters 4 and 5. In Chapter 6, a summary of the findings and the conclusions is given. In the Appendix, technical information regarding sample preparation techniques and analytical methods employed the in laboratories are presented.

2 Sources of radionuclides in the marine environment

This chapter provides updated information and an overview of radionuclide discharges and other sources that are relevant to the Norwegian marine environment. The collection and updating of this information is an integral part of the marine monitoring programme RAME (Radioactivity in the Marine Environment), the main purpose of which is to present recent trends in radionuclide discharges and other sources of radioactivity in the marine environment. Information on discharges from Norwegian sources and on long-range transport of radionuclides from various distant sources is included. Anthropogenic radionuclides are discharged from the nuclear facilities of the Institute for Energy Technology, IFE Kjeller and IFE Halden. Discharges from these facilities are authorised by the NRPA. Unsealed radioactive materials used in medicine and science will primarily be released to the marine environment via sewage treatment plants. The utilisation of such unsealed sources is regulated through guidelines issued by the NRPA.

In recent years, increased attention has been devoted to releases containing elevated levels of naturally occurring radionuclides. Such releases may occur in connection with offshore oil production and, probably, leaching of mine tailings.

The discharge of radionuclides from Norwegian sources is only detectable in the local environment, near the discharge point. The long-range transport of radionuclides originating from fallout from atmospheric nuclear weapons tests (conducted mainly in the 1950s and 1960s), the Chernobyl accident in 1986 and from reprocessing of nuclear fuel are still the main contributors to the general levels of anthropogenic radionuclides found in Norwegian marine waters.

2.1 Discharges of anthropogenic radionuclides from Norwegian sources

2.1.1 IFE Kjeller and GE Healthcare

IFE Kjeller is located about 20 km east of Oslo. The facilities include a heavy-watercooled and moderated research reactor with a thermal effect of 2 MW, called JEEP II, a metallurgical laboratory, production facilities for medical radioactive isotopes, and a radioactive waste treatment plant for low-level and intermediate-level waste (LLW and ILW). The liquid LLW originates from these facilities. Liquid effluent is discharged through a designated pipeline to the River Nitelva about 100 km from the sea. The river empties into Lake Øyeren where the water is mixed with the water from the Glomma River. The River Glomma empties into the Oslofjord at the city of Fredrikstad.

A plant manufacturing radiopharmaceutical products, managed by the private company GE Healthcare (former Amersham Health). operated at the IFE Kjeller site up to 2009. Authorisation for IFE Kjeller discharges also included the discharge from this production facility. The production of radiopharmaceuticals was closed down in 2008/2009 and the contribution to the discharge from GE Healthcare in 2009 was very low.

Table	<i>2.1</i> .	Liquid	disch	arges	(MBq)	from	IFE
Kjeller	· (IFE	2010a,	; IFE	2009a	ı; IFE	2008a;	IFE
2007a;	IFE	E 2006a	ı), 20	005-200	09 (inc	cluding	GE
Health	care).						

Nuclide	2005	2006	2007	2008	2009
²⁴¹ Am	0.03	0.006	0.007	0.907	0.0022
¹²⁴ Sb	< 0.1	< 0.3	< 0.2	< 0.2	< 0.02
¹²⁵ Sb	< 0.7	0.7	< 0.7	<0.6	< 0.2
¹⁴⁴ Ce	0.054	2.7	<1.3	0.38	< 0.2
¹³⁴ Cs	0.08	5.3	0.5	4.1	< 0.04
¹³⁷ Cs	0.51	53.6	9.5	32.1	0.398
³ H	$4.1 \cdot 10^{5}$	$1.43 \cdot 10^{6}$	$4.99 \cdot 10^{5}$	$1.69 \cdot 10^{6}$	400
⁵⁹ Fe	< 0.2	< 0.5	< 0.3	< 0.5	< 0.03
^{125}I	50	42.8	44.1	44.9	25.0
131 I	39	4.2	2.0	1.26	0.005
⁵⁸ Co	< 0.1	<0.3	< 0.2	< 0.2	< 0.02
⁶⁰ Co	6.7	74	71.7	33.1	1.56
⁵¹ Cr	<1.3	<0.9	< 0.5	<1.7	< 0.04
⁵⁴ Mn	< 0.2	< 0.4	< 0.3	< 0.2	< 0.03
⁹⁵ Nb	< 0.1	0.04	< 0.1	< 0.2	< 0.01
²³⁸ Pu	0.007	0.003	0.002	0.0469	0.0001
²³⁹⁺²⁴⁰ Pu	0.15	0.04	0.07	7.65	0.0141
¹⁰³ Ru	< 0.3	0.02	< 0.1	< 0.2	< 0.01
¹⁰⁶ Ru	<2.2	<2.6	<2.0	<1.7	< 0.3
⁹⁰ Sr	0.34	1.2	0.50	12.2	0.356
^{110m} Ag	1.2	0.4	0.05	< 0.2	< 0.04
⁶⁵ Zn	8.4	<0.9	< 0.7	2.75	< 0.07
⁹⁵ Zr	< 0.2	< 0.5	< 0.2	< 0.4	< 0.02
²² Na	-	-	0.56	< 0.2	< 0.03
²³⁴ U	-	0.007	0.0024	0.879	0.003
²³⁵ U	-	$3 \cdot 10^{-4}$	$2.5 \cdot 10^{-5}$	0.0346	9.10-5
²³⁸ U	-	0.005	0.0018	0.924	0.0029

The discharge limit authorised by the NRPA is based on the annual dose to any member of a critical group of the population along the River Nitelva, and shall not exceed 1 μ Sv. Each year, IFE Kjeller reports discharge data, the results of their environmental monitoring programme and calculations of effective doses resulting from discharges to NRPA. The reported discharges for the period 2005-2009 are summarised in Table 2.1.

Table 2.2. Effective doses (μ Sv) to any member of the critical group from river water exposure pathways as reported by IFE Kjeller (IFE 2010; IFE 2009; IFE 2008a; IFE 2007a; IFE 2006a).

2005	2006	2007	2008	2009
0.02	0.48	0.09	0.304	0.0053

Effective doses have been calculated by IFE for an individual in the hypothetical critical group exposed through the annual consumption of 20 kg of fish from the river and 100 hours per year presence on the riverbanks. In 2008 and 2009, the calculated effective dose to this critical group was 0.30 μ Sv and 0.0053 μ Sv, respectively, corresponding to 30 % and 0.53 % of the annual dose limit of 1 μ Sv. The effective dose to the critical group varies, as can be seen in Table 2.2, corresponding to between 0.53 and 30 % of the dose limit, with an average of 18 % for the period 2005-2009.

2.1.2 IFE Halden

The Halden Boiling Water Reactor (HBWR) is located in the town of Halden, in the southeast of Norway, close to the Swedish border. The HBWR is heavy-water-cooled and moderated with a thermal effect of 20 MW. The reactor was commissioned in 1959.

IFE Halden is authorised to discharge a limited amount of radioactivity from the research reactor to the River Tista (which empties into the Iddefjord). The discharge limit for liquid waste to the marine environment is based on a dose limit of 1 μ Sv per year to a hypothetical critical group, and the actual annual discharge is typically 1-3 % of the authorised limit. Liquid discharges in the period 2005-2009, and the corresponding doses to the critical group are presented in Tables 2.3 and 2.4.

Table 2.3. Liquid discharge (MBq) from IFE Halden, 2053 - 2009 (IFE 2010b; IFE 2009b; IFE 2008b; IFE 2007b; IFE 2006b).

Nuclide	2005	2006	2007	2008	2009
¹²⁴ Sb	0.003	-	0.0093	0.0038	0.014
¹²⁵ Sb	0.037	0.0064	0.023	0.007	0.025
¹⁴¹ Ce	0.61	0.25	1.5	1.9	0.57
¹⁴⁴ Ce	8.8	3.0	17	22	4.9
¹³⁴ Cs	18	25	29	39	8.1
¹³⁷ Cs	130	190	240	260	91
³ H	$5.3 \cdot 10^{5}$	$3.0 \cdot 10^{5}$	$9.8 \cdot 10^{6}$	$2.2 \cdot 10^{6}$	$1.8 \cdot 10^{6}$
⁵⁹ Fe	0.88	-	0.55	2.8	0.44
131 I	0.42	21	1.8	25	2.4
¹⁰⁹ Cd	-	-	0.13	0.024	-
⁵⁸ Co	41	4.0	3.6	18	3.9
⁶⁰ Co	99	86	49	46	40
⁵¹ Cr	180	240	25	30	43
⁵⁴ Mn	2.5	1.1	0.45	0.98	0.45
⁹⁵ Nb	12	8.3	12	11	9
¹⁰³ Ru	0.42	0.16	0.86	1.9	0.37
¹⁰⁶ Ru	-	-	2.6	2.6	0.73
⁹⁰ Sr	4.9	4.4	6.8	5.2	1.0
^{110m} Ag	0.94	0.21	3.2	2.2	2.1
⁹⁵ Zr	5.3	3.8	4.8	5.8	3.9

The calculation of the effective dose to the critical group is based on the following assumptions:

- An annual consumption of 30 kg of fish from the Iddefjord,
- 200 hours per year exposure on the shore of the fjord,

- 50 hours per year bathing in the fjord, and

- 1000 hours per year boating on the fjord.

 Table 2.4. Effective doses (µSv) to the critical group from marine exposure pathways as reported by IFE Halden, 2005 - 2009 (IFE 2010b; IFE 2009b; IFE 2008b; IFE 2007b; IFE 2007b; IFE 2006b).

 2009
 2006
 2007
 2008
 2009

0.025 0.02	0.017	0.016	0.011

2.2 Unsealed radioactive substances in the medical sector

Unsealed radioactive substances used in medicine dominate the anthropogenic radioactive discharges to the sewage system. Radioactive solutions are given to patients for diagnostic and therapeutic purposes, and enter the sewage systems mainly by the excretion of urine and faeces. The two most used radionuclides in nuclear medicine for diagnostic and the rapeutic purposes are $^{99\mathrm{m}}\mathrm{Tc}$ (which decays to 99 Tc with a half-life of 6 h) and 131 I (T_{1/2} = 8.04 d). The most important radionuclide concerning dose to the public after discharge is ¹³¹I. The discharge of ¹³¹I radionuclides has been estimated according instructions published by OSPAR. The percentage of the administered doses that is discharged as liquid waste is:

Ablation therapy: 100% of the administered dose.

Thyrotoxicosis treatment: for in-patients, 50% of the administered dose; for out-patients, 30% of the administered dose; for patients who cannot be classified as in-patients or out-patients, 50% of the administered dose.

Under these assumptions the total discharged activity of 131 I in 2008 and 2009 from the

medical sector was 1094 GBq and 1177 GBq, respectively.

2.3 Discharge of produced water containing ²²⁶Ra and ²²⁸Ra

Large volumes of produced water, containing dissolved ²²⁶Ra and ²²⁸Ra, are discharged into the sea during oil exploitation. Produced water may be formation water trapped in the reservoirs for millions of years or a mixture of formation water and sea water, if sea water has been injected in order to maintain pressure in the reservoirs during oil and gas production. Although the levels of ²²⁶Ra and ²²⁸Ra are not technologically enhanced. the natural concentrations of these isotopes in the saline formation water are generally much higher (about 3 orders of magnitude) than the background concentration in sea water (a few Bq m^{-3}). The concentration of radium in the produced water may change over the lifetime of the well. Injection of large volumes of sea water, to maintain the pressure within the well, may lead to dilution of the radium in the produced water, but at the cost of greater water-to-oil ratios later in the production process.

In 2008, 149 10⁶ m³ of produced water was discharged to the marine environment from the Norwegian oil and gas industry, while 30 10⁶ m³ was reinjected in the reservoirs (OLF, 2009). The total activity of ²²⁶Ra and ²²⁸Ra discharged was about 460 GBq and 370 GBq, respectively. In Figure 2.1 the activity of ²²⁶Ra and ²²⁸Ra discharged from Norwegian oil fields are presented. About 40 % of the total discharged activity of ²²⁶Ra and ²²⁸Ra was discharged from the Troll B and C platforms.

In 2009, 134 10⁶ m³ of produced water was discharged to the marine environment from the Norwegian oil and gas industry, while 30 10⁶ m³ was reinjected in the reservoirs (OLF, 2010). The total activity of ²²⁶Ra and ²²⁸Ra discharged was about 480 GBq and 360 GBq, respectively. In Figure 2.2 the activity of ²²⁶Ra and ²²⁸Ra discharged from Norwegian oil fields are presented. About 40 % of the total discharged activity of ²²⁶Ra and ²²⁸Ra was discharged from the Troll B and C platforms. Discharge of ²²⁶Ra and ²²⁸Ra from Norwegian

platforms in the period 2005 to 2009 is presented in Figure 2.3. Reported annual discharges of ²²⁶Ra from all offshore oil and gas installations in the North Sea in the period 2005-2008 have been in the range from 0.78 to 0.90 TBq (OSPAR, 2010a).



Figure 2.1. Estimated discharged activity of ²²⁶Ra and ²²⁸Ra from Norwegian oil and gas fields in 2008.



Figure 2.2. Estimated discharged activity of ²²⁶Ra and ²²⁸Ra from Norwegian oil and gas fields in 2009.



Figure 2.3. Total annual discharge of ²²⁶Ra and ²²⁸Ra via produced water and discharged volume of produced water from the Norwegian oil and gas industry in the period 2005 to 2009.

2.4 Long-range transport of radionuclides

There are several real and potential sources of radioactive contamination of the northeast Atlantic and the Arctic marine environment, as shown in Figure 2.4.



Figure 2.4. Sources of radionuclides in the northern marine environment: Chernobyl in the Ukraine, the reprocessing plants at Sellafield, Cap de la Hague and Dounreay, the dumping sites for nuclear waste in the Kara Sea, the sites of the sunken submarines Komsomolets and K-159 and Russian nuclear installations (Mayak, Tomsk and Krasnoyarsk) releasing radionuclides to the Russian rivers Ob and Yenisey.

The primary sources are fallout from atmospheric nuclear weapons testing during the 1950s and 1960s, discharged radionuclides from reprocessing plants and fallout from the Chernobyl accident.

Radioactive contaminants discharged into northern European marine waters can reach Norwegian waters by ocean current transportation. The main ocean currents of the area are presented in Figure 2.5. Some elements, such as Pu and Am, are particlereactive and will to a large extent sink and end up in the sediments. From the sediments the radionuclides can later be remobilised and away transported local by currents. Remobilised plutonium and ¹³⁷Cs from Irish Sea sediments contaminated by previous Sellafield discharges are one of the main sources of these elements in Norwegian marine waters. Other elements, such as ⁹⁹Tc and ⁹⁰Sr, are not particle-reactive and will follow currents and can be transported large distances away from the discharge point.



Figure 2.5. Overview of the main current system in the North Sea, Norwegian Sea, Greenland Sea and Barents Sea (Source: IMR).

2.4.1 Discharge of radionuclides from European nuclear installations

Spent nuclear fuel can be managed in different ways. In a few countries, for instance in France and the UK, spent nuclear fuel is reprocessed. The major part of the waste from these facilities, such as fission products, is stored pending final management and disposal, but a small part is discharged as liquid waste to the marine environment. Discharge limits are set by the relevant national authority.

Sellafield (formerly Windscale) is located on the west coast of Cumbria in the UK. Further information about this reprocessing plant can be found in Amundsen et al., (2003). The other reprocessing plant in Europe affecting Norwegian marine waters is located at Cap de la Hague in France. Except for tritium (³H), the discharge of liquid radioactive waste has generally been lower here than from Sellafield. In 1994 and 1995, the discharge of ⁹⁹Tc from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced, it was continued at a high level up to 2003. The discharge of ⁹⁹Tc has been substantially reduced since 2004, when a new chemical treatment process that removes ⁹⁹Tc was implemented.

The discharges of alpha- and beta-emitting (excluding tritium) radionuclides to marine

environment of the North-East Atlantic from all European nuclear installations have decreased significantly in the period 1990-2008. The reported discharges in 2008 were the lowest recorded by OSPAR in this period (OSPAR, 2010b).

In addition to the direct discharges from reprocessing, the remobilisation of ¹³⁷Cs and plutonium from contaminated sediments in the Irish Sea acts as a secondary source of radiothe marine environment. nuclides in Substantial discharges of ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, ²⁴¹Am and ¹³⁷Cs and other radionuclides in the 1970s and early 1980s resulted in widespread contamination of Irish Sea sediments. It has been shown that these radionuclides can be redissolved and transported out of the Irish Sea. An annual loss of 1.2 TBq ²³⁹⁺²⁴⁰Pu and 86 TBq ¹³⁷Cs from the Irish Sea has been estimated by Cook et al., (1997), while Leonard et al., (1999) estimated annual losses of plutonium based on remobilisation from surface sediments to be 0.6 TBq. The half-lives of plutonium and americium in Irish Sea sediments were estimated to be 58 years and ~1000 years, respectively. This implies that remobilisation of radionuclides from Irish Sea sediments is a significant source of anthropogenic radionuclides in the North Sea and Norwegian coastal areas, and will remain so for a long time. Liquid radioactive discharges from Sellafield and Cap de la Hague in 2008 and 2009 are presented in Table 2.5.

Table 2.5. Liquid discharge of radionuclides (TBq)
from Sellafield and Cap de la Hauge to the marine
environment in 2008 and 2009 (OSPAR, 2010b;
AREVA, 2010; Environment Agency et al., 2010).

	Sellafield		Cap de la Hague	
Nuclide	2008	2009	2008	2009
³ H	778	1510	8193	9130
Total-α	0.127	0.154	0.0172	0.0162
Total-β	14.3	17.8	6.37	
¹⁴ C	7.19	8.19	6.24	6.12
⁵⁴ Mn			2.26 10-3	
⁵⁷ Co			7.31 10-5	
⁵⁸ Co			6.41 10-5	
⁶⁰ Co	0.0721	8.25 10-2	9.72 10-2	8.9 10-2
⁶³ Ni			6.44 10-2	
⁶⁵ Zn				
⁹⁰ Sr	1.7	2.86	0.159	0.115
⁹⁹ Tc	2.37	3.08	7.41 10-2	
¹⁰⁶ Ru	1.39	3.16	3.37	1.58
¹²⁵ Sb			0.38	
^{129}I	0.199	0.253	1.04	
¹³⁴ Cs	0.115	0.141	7.50 10-2	6.89 10-2
¹³⁷ Cs	5.11	4.27	0.892	1.04
¹⁴⁴ Ce	0.354	0.498	7.55 10-5	
¹⁵⁴ Eu			5.55 10-4	
¹⁵⁵ Eu			8.08 10-5	
²³⁹⁺²⁴⁰ Pu	0.108	0.12	1.69 10-3	
²⁴¹ Pu	2.44	2.87	0.12	
²⁴¹ Am	0.0297	4.63 10-2	2.74 10-3	
²³⁷ Np	0.043	5.31 10-2	4.27 10-4	
²⁴² Cm			1.14 10-5	
²⁴³⁺²⁴⁴ Cm	2.92 10-3	4.52 10-3	1.35 10-3	
U (kg)	276 kg	409 kg	19.3 kg	

2.4.2 Global fallout from nuclear weapons testing

From a global point of view, atmospheric nuclear weapons tests, conducted between 1945 and 1980, are the largest source of radioactive contamination in the environment. The most intensive test period was between 1952 and 1962, during which the USSR and USA were responsible for most of the tests. The last atmospheric nuclear detonation was conducted in China in 1980. In total, it has been estimated that 948 PBq ¹³⁷Cs, 622 PBq ⁹⁰Sr, 6.52 PBq ²³⁹Pu and 4.35 PBq ²⁴⁰Pu have been released to the environment (UNSCEAR, 2000a), the major part in the northern hemisphere, where most of the tests took place.

Fallout from atmospheric nuclear weapons tests can still be found in the marine environment. About 60 % of the released activity was deposited in the world's oceans. The distribution of the fallout today depends on parameters such as ocean currents, diffusion and sedimentation rates. Earlier expeditions in the Atlantic have shown that the concentration of ¹³⁷Cs in surface water is about a few Bq m⁻³ (about twice as much as 90 Sr), between 45° S and 45° N, with a slight increase to the north (Bourlat *et al.*, 1996). The activity concentration of $^{239+240}$ Pu shows the same trend, but the activity concentrations are about 1000 times lower. Reasons for this are that less plutonium was released, but also that plutonium is more particle reactive and has a higher sedimentation rate, which leads to a shorter residence time in the water.

2.4.3 Outflow of Chernobylcontaminated water from the Baltic Sea

The worst nuclear accident in history occurred on April 26, 1986 in Chernobyl, Ukraine. In the initial explosion and the following fire. about 3-4 % of the nuclide inventory in the core was released to the atmosphere. The major part consisted of volatile radionuclides such as inert gases, iodine isotopes and ¹³⁴Cs and ¹³⁷Cs. Many of the released nuclides had short half-lives and can not be observed in the environment today. The radionuclide that will affect the environment in a long-term perspective is ¹³⁷Cs, with a physical half-life of 30 years. In total about 85 PBq of ¹³⁷Cs was released to the environment (UNSCEAR, 2000b). The Baltic Sea was the most affected marine area, where about 5 PBq was deposited (Evans, 1991). Shortly after the accident the activity concentration of ¹³⁷Cs was very high in part of the Baltic Sea. Due to low water exchange and a constant supply from rivers it will take a relatively long time for the levels to decrease. The average activity concentration of ¹³⁷Cs in the southern Baltic Sea surface water in 2008 was reported to 35 Bq m⁻³ (Zalewska and Saniewska, 2009). Due to the water exchange through the Danish Straits it was estimated an outflow of 40 TBq from the Baltic Sea into the Kattegat in 2000. With the observed effective half-life the outflow in 2009 can be estimated to about 25 TBq (Dahlgaard, 2002). The ¹³⁷Cs will subsequently be transported by the Baltic current which continues as the Norwegian coastal current. This is one of the major sources of ¹³⁷Cs in the Norwegian marine environment, together with remobilised ¹³⁷Cs from the seabed of the Irish Sea and fallout from atmospheric nuclear weapons tests.

2.5 Other present and potential sources of radioactivity in the northern marine environment

In addition to discharges from the western European reprocessing plants, northern marine waters have also received an input of various radionuclides from Russian reprocessing plants situated on the tributaries of the Russian rivers Ob and Yenisey (see Figure 2.2). These are the Mayak Production Association in the southern Urals and the Siberian Chemical Combine (Tomsk-7) north of the town of Tomsk, and the Krasnovarsk Mining and Chemical Combine (KMCC). Uncertainty exists regarding the magnitude of radionuclide contributions from these sources, although estimates have been made. For example, between 1958 and 1993 routine discharges from the KMCC are estimated to have led to an input of 30-100 TBq of ¹³⁷Cs into the Kara Sea; a level which is of a similar order of magnitude to inputs from Sellafield-derived ¹³⁷Cs to the area (Vakulovsky et al., 1995). However, the fact that the prevailing surface currents tend to transport contamination away from the Kara Sea in predominantly easterly and northerly directions, suggests that the resultant inputs to the Barents Sea has been relatively low. Nonetheless, there is a potential for significant radionuclide contamination following large accidental releases of radioactivity from these nuclear complexes.

Several other sources exist which also represent potential sources of radionuclides in the Barents and Norwegian Seas. Among these are radioactive waste containers dumped in the Barents and Kara Seas by the Former Soviet Union and the sunken submarines the Komsomolets and the K-159 in the Norwegian Sea and the Barents Sea. The dumping areas and the locations of the sunken submarines are given in Figure 2.4. Several investigations have been conducted to detect possible leakage from these sources.

As part of the bilateral environmental cooperation with the Russian Federation, a joint Russian-Norwegian expert group has been investigating the condition of the contained dumped objects and has analysed sediment and water samples at these sites.

According to Strand *et al.* (1998), elevated levels of radionuclides in sediments collected in the vicinity of the dumped objects

demonstrated that leakage had occurred, but no significant inputs of radionuclides to the open sea from this source have been detected. Following an accident on April 7, 1989, the Russian nuclear submarine the Komsomolets, lies at a depth of 1700 metres, 180 km southwest of Bear Island in the Norwegian Sea. The submarine contains a nuclear reactor and two torpedoes with nuclear warheads. reported Kolstad (1995) low activity concentrations of ¹³⁴Cs in sea water samples collected near the submarine, showing that leakage of radiocaesium isotopes may have occurred. Samples of water and sediments collected around the submarine by the Institute of Marine Research and analysed with respect to ¹³⁷Cs are presented in Figure 2.6.



Figure 2.6. Cs-137 in samples from the position of the sunken nuclear submarine "Komsomolets".

3 Collection of samples

In 2008 and 2009 samples were collected in the Barents Sea, the North Sea, the Skagerrak in selected fjords and at permanent coastal stations. A geographic overview of the sampling area covered by the marine monitoring programme, with the fixed coastal stations marked, is shown in Figure 3.1.



Figure 3.1. Geographic overview of the sampling area covered by the marine monitoring programmes. (Fixed coastal sampling stations are indicated by yellow dots).

Sampling and analysis were carried out by the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR) and the Institute for Energy Technology (IFE).

Samples from the Barents Sea were collected in August - October 2008 by IMR and NRPA from the vessels R/V "Johan Hjort", R/V "G.O. Sars" (Figure 3.2) and R/V "Jan Mayen". In 2009 samples were collected in August – September from the same vessels. During the expeditions samples of surface water were collected which were later analysed for ⁹⁹Tc, ¹³⁷Cs, ⁹⁰Sr, ²⁴¹Am and plutonium isotopes. Sediment was also sampled and later analysed for ¹³⁷Cs.



Fig. 3.2. R/VG O Sars (Photo: NRPA).

3.1 Sea water

For ⁹⁹Tc, ⁹⁰Sr and 50-100 litres of unfiltered water was collected at each sampling station and later transported to various laboratories for analysis.

For the analysis of 137 Cs, Cu₂[Fe(CN)₆]impregnated cotton filters are used as sorbents (Roos *et al.*, 1994). The system consists of a prefilter (1 micron) and two Cu₂[Fe(CN)₆]impregnated cotton filters connected in series (Figure 3.3). Assuming identical collection efficiency for the Cu₂[Fe(CN)₆]-impregnated cotton filters, the amount of caesium passing through the prefilter can be calculated by considering the two filters as the first two terms in a geometrical series, which has the sum:

$$A(^{137}Cs) = A_1/(1-A_2/A_1)$$

where A_1 and A_2 are the ¹³⁷Cs activities on the first and second impregnated filters, respectively.



Figure 3.3. Filter system used for analysis of ^{137}Cs in sea water (Photo: NRPA).

Sea water samples of 200 litres (unfiltered) were collected for the determination of the activity concentrations of $^{239+240}$ Pu and 241 Am. To collect deep water samples a CTD/Rosette multi bottle sampler was used (Figure 3.4). Plutonium-242 and 243 Am were used as chemical yield determinants. The precipitation of plutonium and americium from the water was performed *in situ* according to the analytical procedure described by Chen *et al.* (1991).



Fig. 3.4. CTD/Rosette multi-bottle sampler(Photo: NRPA).

3.2 Sediment

Sediment samples were retrieved from the seabed using a Smøgen box corer. Profiles were collected from the box by slowly inserting PVC tubes into the sediment sample. Surface sediment samples were obtained from the upper 2 cm layer of the sediment remaining in the box-corer.

3.3 Seaweed

NRPA collected seaweed (*Fucus vesiculosus*) samples from four stations along the Norwegian coastline. At Hillesøy in northern Norway, seaweed is collected every month and at the other locations once per year. In addition, IFE performs monthly or annual seaweed sampling at eleven locations along the coastline, from the Russian border in the north to the Swedish border in the south.

3.4 Fish, crustaceans and molluscs

IMR collected fish samples from commercial fishing locations in the Barents Sea, the Norwegian Sea and the North Sea. Samples of cod were prepared twice a year from 2 locations in the Barents Sea by combining muscle samples of 5 - 50 grams from 25 individual fish. In addition samples of polar cod, capelin and shrimps were collected.

At the request of NRPA, Labora A/S in Salten collected different samples of fish species by combining muscle samples of 5 - 50 grams from 25 individual fish, and crustaceans from the northern part of Norway.

Samples of mussles (*Mytilus edulis*) were collected by the Norwegian Food safety Authority from eight different locations along the Norwegian coast.

4 Radioactivity in sea water and sediments

4.1 Technetium-99 in sea water

Tc-99 has a physical half-life of 2.13.10⁵ years, and originates from the decay of ⁹⁹Mo, which is formed either as a fission product from ²³⁵U or ²³⁹Pu or by neutron activation of ⁹⁸Mo. The main source of ⁹⁹Tc in Norwegian waters is liquid discharge from the reprocessing plant at Sellafield. In oxygenated sea water ⁹⁹Tc is present as the highly soluble pertechnetate ion (TcO_4) . Due to its conservative behaviour in sea water, TcO_4 has the ability to be transported by ocean currents without being significantly affected by sedimentation processes. From the Irish Sea, ⁹⁹Tc is transported by ocean currents to the North Sea and via the Norwegian Coastal Current up to the Barents Sea. The transit time (the time between a specific discharge and the maximum activity concentration from that discharge reaching the sampling location) for ⁹⁹Tc to reach this area (Hillesøy) from the Irish Sea has been estimated to 3-4 years (Brown et al., 2002; Dahlgaard, 1995).

During 2008 and 2009, sea water samples were collected in the North Sea. the Norwegian Sea and the Barents Sea and analysed for ⁹⁹Tc. The results are presented in Figure 4.1 and 4.2. In the North Sea the activity concentration in 2008 of 99 Tc ranged from 0.29 to 2.0 Bq m⁻³. The activity concentration of ⁹⁹Tc in the Barents Sea in 2008 and 2009 ranged from m^{-3} . The 0.03 0.44 Bq to highest concentrations were found in coastal water. The activity concentrations found in the North Sea and the Barents Sea are generally lower than those observed in 2002, 2003 and 2005 (NRPA, 2005; NRPA, 2007). The reason for this is the reduced discharge of ⁹⁹Tc from Sellafield. Monthly samples have also been collected at Hillesøy (Figure 4.3). The results from Hillesøy show that the activity concentration of ⁹⁹Tc in sea water continues to slowly decrease since the peak concentrations were recorded in 1998/1999.



Figure 4.1. Activity concentration $(Bq m^{-3})$ of ⁹⁹Tc in sea water samples collected in the North Sea and the Barents Sea in 2008.



Figure 4.2. Activity concentration $(Bq m^{-3})$ of ⁹⁹Tc in sea water samples collected in the Barents Sea in 2009.



Figure. 4.3. Activity concentration (Bq m^{-3}) of ^{99}Tc in sea water at Hillesøy from monthly sampling.

4.2 Strontium-90 in sea water

Strontium-90 is a fission product with a physical half-life of 29 years. Similar to 99 Tc, 90 Sr is a conservatively behaving element in the marine environment.

Sea water has been sampled mainly in the Barents Sea and later analysed for ⁹⁰Sr. The main sources of ⁹⁰Sr in these waters are discharge of liquid waste from reprocessing plants (mainly Sellafield), fallout from atmospheric nuclear weapons tests conducted mainly in the 1950s and 1960s and outflow of water from the Baltic Sea. The results for 2008 and 2009 are presented in Figure 4.4 and 4.5.



Figure 4.4. Activity concentration $(Bq m^{-3})$ of ${}^{90}Sr$ in surface water samples collected in the Barents Sea in 2008.

In the Barents Sea the activity concentration in surface water ranged from 0.7 Bq m^{-3} to 1.5 Bq m^{-3} . The highest concentrations were found in the Skagerrak. Typical activity concentration of 90 Sr in sea water from the southern Baltic

Sea in 2008 has been reported to be around 6 Bq m^{-3} (Zalewska and Saniewski, 2009).



Figure 4.5. Activity concentration $(Bq m^{-3})$ of ${}^{90}Sr$ in surface water samples collected in the North Sea, the Skagerrak and the Barents Sea in 2009.

Results from annual or biannual sampling at Hillesøy shows that the activity concentration in sea water in the Norwegian coastal current is slowly decreasing (Figure 4.6). One explanation for this is the reduced discharges from Sellafield over the last 10 years (Figure 4.7).



Figure. 4.6. Activity concentration (Bq m^{-3}) of ⁹⁰Sr in sea water at Hillesøy.



Figure 4.7. Annual liquid discharge of ⁹⁰Sr from Sellafield and Cap de la Hague in the period 1999 to 2009 (data from OSPAR).

4.3 Caesium-137 in sea water and sediment

Caesium-137 is a fission product with a halflife of 30 years. The main sources of ¹³⁷Cs in the North Sea and the Skagerrak are fallout from atmospheric nuclear weapons tests in the 1950s and 60s, outflowing water from the Baltic Sea and ¹³⁷Cs remobilised from Irish Sea sediments. Runoff from land, from the areas with the highest Chernobyl fallout can also contribute locally in coastal water. Like ⁹⁹Tc and ⁹⁰Sr, ¹³⁷Cs is also a conservatively behaving radionuclide in sea water.



Figure 4.8. Activity concentration $(Bq m^{-3})$ of $1^{37}Cs$ in sea water samples (surface water) collected in the Norwegian Sea, the Skagerrak and the Barents Sea in 2008.

Observed levels of 137 Cs in the North Sea and the Skagerrak in 2008 are shown in Figure 4.8. Results from 2009 are presented in Figure 4.9 and 4.10. The activity concentration in surface water sampled in the North Sea and the Skagerrak in 2008 and 2009 ranged from 1.4 to 35.4 Bq m⁻³, where the highest levels were found in the Skagerrak. The activity concentration of 137 Cs versus salinity is presented in Figure 11, showing that the highest levels are found in brackish water from the Baltic Sea.



Figure 4.9. Activity concentration (Bq m⁻³) of ¹³⁷Cs in sea water samples (surface water) collected in the Barents Sea in 2009.



Figure 4.10. Activity concentration (Bq m⁻³) of ¹³⁷Cs in sea water samples (surface water) collected in the Skagerrak and Kategatt in 2009.

Data from Hillesøy from the period 2002 and 2009 (Figure 4.12) show that the levels of ¹³⁷Cs in the Norwegian coastal current are slowly decreasing. The effective half-life of ¹³⁷Cs in the Baltic Sea, which is one of the main sources of ¹³⁷Cs in this region, has been estimated to 10 years (Ikäheimonen *et al.* 2009).

In 2009, the activity concentration of 137 Cs in the Barents Sea ranged from 1.5 to 2.3 Bq m⁻³. This is similar or lower than the observed levels in 2002, 2003, 2005 and 2006 (NRPA, 2004, NRPA, 2005; NRPA, 2007 and NRPA 2008).

Cs-137 has also been analysed in surface sediments (upper 2 cm layer) from the Barents Sea, the North Sea and selected fjords. The results are presented in Figure 4.13 and Figure 4.14 and range from 0.8 to 7.0 Bq kg⁻¹ (d.w.) in the Barents Sea. The activity concentration 137 Cs in sediments sampled in the fjords ranged from 3.1 to 323 Bq kg⁻¹ (d.w.). Highest activity concentration were found in samples from Sognefjorden.



Figure 4.11. Salinity versus activity concentration of ¹³⁷Cs in sea water samples from the Skagerrak, the North Sea and the Barents Sea in 2008 and 2009.



Figure 4.12. Activity concentration ($Bq m^{-3}$) of ^{137}Cs in sea water at Hillesøy in the period 2002-2009.



Figure 4.13. Activity concentration ($Bq kg^{-1} d.w.$) in surface sediment from the Barents Sea and selected fjords in 2008.



Figure 4.14. Activity concentration ($Bq kg^{-1} d.w.$) in surface sediment from the Barents Sea in 2009.

4.4 Plutonium-239+240 in sea water

Plutonium-239 (T_{1/2} = 24 110 y) and 240 Pu (T_{1/2} = 6563 y) belong to the transuranium elements and are mainly produced by neutron capture (with subsequent beta decay) by ²³⁸U and ²³⁹Pu, respectively. The behavior of plutonium in the marine environment is complex due to its different possible oxidation states. Under oxidizing conditions Pu is generally found in the more soluble +V and +IV state, while in reducing environments the more particlereactive +III and +IV states dominate. The main sources of ²³⁹⁺²⁴⁰Pu in northern Norwegian marine waters is global fallout from atmospheric nuclear weapons tests in the 1950s and 1960s, and remobilised plutonium from Irish Sea sediments.



Figure 4.15. Activity concentration $(mBq m^{-3})$ of $^{239+240}Pu$ in surface water samples collected in the Barents Sea in 2008.



Figure 4.16. Activity concentration $(mBq m^{-3})$ of $^{239+240}$ Pu in surface water samples collected in the North Sea, the Skagerrak and the Barents Sea in 2009.

Observed levels in the Barents Sea in 2008 and 2009 are presented in Figure 4.15 and 4.16 and range from 1.4 to 12.4 mBq m⁻³. The levels of ²³⁹⁺²⁴⁰Pu in the Barents Sea in 2008 and 2009 are similar as in 2002 and 2005 (NRPA, 2003; NRPA, 2006).

4.5 Americium-241 in sea water

Americium-241 belongs to the transuranium elements and has a physical half-life of 432 years. It is produced, for example, in nuclear reactors by successive neutron capture by ²³⁹Pu. Am-241 is finally formed by beta-decay of ²⁴¹Pu ($T_{1/2} = 14.35$ y). Main sources of ²⁴¹Am in the environment are fallout of ²⁴¹Pu from nuclear weapon tests in the 1950s and 1960s and the discharge of ²⁴¹Am and ²⁴¹Pu from reprocessing plants.



Figure 4.17. Activity concentration $(mBq m^{-3})$ of ²⁴¹Am in sea water from the Barents Sea in 2008.



Figure 4.18. Activity concentration $(mBq m^{-3})$ of ²⁴¹Am in sea water from, the Skagerrak and the Barents Sea in 2009.

Measured activity concentrations of 241 Am in the Barents Sea in 2008 and 2009 are presented in Figure 4.17 and 4.18 and range from 1.0 to 9.0 mBq m⁻³. The observed levels of 241 Am are similar as in 2002 and 2005 (NRPA, 2003; NRPA, 2006).

Radium-226 in sea water

Radium-226 is а naturally occurring radionuclide with a physical half-life of 1 600 years. As a member of the natural decay chain starting with 238 U (t_{1/2} = 4.47·10⁹ years), it is continuously produced by the decay of 230 Th, and can be found in different concentrations in the environment. In the marine environment ²²⁶Ra is naturally supplied from both the sediments (the mother nuclide ²³⁰Th is particle reactive and is mainly found attached to sediment particles) and by river water to the oceans. Typical activity concentration of ²²⁶Ra in Atlantic surface water has been reported to be about 1.3 Bq m⁻³ (IAEA, 1990).



Figure 4.19. Activity concentration (*Bq* m⁻³) of ²²⁶*Ra* in surface sea water from the Barents Sea in 2009.

The activity concentrations of 226 Ra observed in the Barents Sea in 2009 are presented in Figure 4.19. The levels range from 0.9 to 2.4 Bq m⁻³. This is similar to those found in 2005 in the same area (NRPA, 2006).

5 Radioactivity in biota

5.1 Technetium-99 in seaweed



Figure 5.1. Brown seaweed (Fucus vesiculosus) (*Photo:NRPA*).

Seaweed is a useful bioindicator for ⁹⁹Tc in the marine environment. It has a high ability to concentrate 99Tc from sea water and is easy accessible in most coastal areas. During 2008 and 2009, seaweed (Fucus vesiculosus, Figure 5.1) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for ⁹⁹Tc. At two locations, Hillesøy and Utsira, sampling was performed monthly. At the remaining sites sampling was conducted in August or September. The results of the analyses are presented in Figure 5.2 and 5.3 and range from 35 to 115 Bq kg⁻¹ (d.w.), where the highest activity concentration (147 Bq kg⁻¹ d.w.) was found in one of the monthly samples collected at Utsira. Compared with the results from 1999-2001 (Rudjord et al., 2001; Gäfvert et al., 2003), the levels of ⁹⁹Tc have decreased at most sampling sites, due to the reduced discharge of ⁹⁹Tc from Sellafield. The trend can also be seen in Figures 5.3 and 5.4 which show the annual average activity concentration of ⁹⁹Tc in Fucus vesiculosus at Utsira and Hillesøy, together with the annual discharge of ⁹⁹Tc from Sellafield.

Tc-99 was also analysed in samples of *Ascophyllum nodosum* collected at Utsira in the period 1995 - 2009 (Figure 5.5). *Ascophyllum nodosum* is known to have a relatively high concentration factor for ⁹⁹Tc. Holm and Rioseco (1986) have shown that the concentration factor for ⁹⁹Tc in *Ascophyllum nodosum* are about twice as high as for *Fucus*

vesiculosus. The highest level found at Utsira, 614 Bq kg⁻¹ (d.w.), was observed in 1999. Since then the levels have decreased and in 2008 and 2009 the activity concentration was found to be 146 and 135 Bq kg⁻¹ (d.w.), respectively.



Figure 5.2. Levels of ^{99}Tc in Fucus vesiculosus sampled along the Norwegian coastline in 2008.



Figure 5.2. Levels of ⁹⁹*Tc* in Fucus vesiculosus sampled along the Norwegian coastline in 2009.

Utsira



Figure 5.3. Annual liquid discharge of ⁹⁹Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) ⁹⁹Tc activity concentration in brown algae (Fucus vesiculosus) sampled at Utsira (data provided by IFE) in the period 1995-2009 (secondary axis).



Hillesøy

Figure 5.4. Annual liquid discharge of ⁹⁹Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) ⁹⁹Tc activity concentration in brown algae (Fucus vesiculosus) sampled at Hillesøy in the period 1997-2009 (secondary axis).



Figure 5.5. Activity concentration (Bq kg⁻¹d.w.) of ⁹⁹Tc in seaweed (Ascophyllum nodosum) sampled at Utsira in the period 1995 to 2009 (data provided by IFE).

5.2 Plutonium-239+240 in seaweed

Fucus vesiculosus has been collected and analysed for $^{239+240}$ Pu at Utsira since 1980. The results from the period 1980 to 2009 are presented in Figure 5.6. The activity concentrations in these samples were in the

range of 23 to 201 mBq kg⁻¹, with relatively large fluctuations from year to year. One can see a slowly decreasing trend in the activity concentration of ²³⁹⁺²⁴⁰Pu in the seaweed samples collected in the period from 1980 to 2009.



Figure 5.6. ²³⁹⁺²⁴⁰*Pu levels (mBq kg⁻¹ d.w.) in* Fucus vesiculosus *at Utsira in the period 1980 to 2009 (data provided by IFE).*

5.3 Caesium-137 in seaweed

Fucus vesiculosus has also been widely used as a bioindicator for 137 Cs. The accumulation of 137 Cs in brown algae is, however, not as pronounced as for 99 Tc. The uptake of 137 Cs also depends on the salinity of the surrounding sea water, with higher uptake at lower salinities (Carlsson and Erlandsson, 1991).



Figure 5.7. Levels of ${}^{137}Cs$ (Bq kg ${}^{-1}$ d.w.) in Fucus vesiculosus sampled along the Norwegian coastline in 2008.

In 2008 and 2009, samples of *Fucus* vesiculosus from the permanent coastal stations were analysed with respect to ¹³⁷Cs. The results are presented in Figure 5.7 and 5.8, and range from <0.2 Bq kg⁻¹ (d.w.) in the north to 3.3 Bq kg⁻¹ (d.w.) at Hvaler. The peak in the ¹³⁷Cs levels in seaweed collected at Vikna can be explained by run-off of ¹³⁷Cs from land, since this area was affected by fallout from the Chernobyl accident in 1986. The reason for the higher activity concentration in *Fucus*

vesiculosus in the southern part of Norway is a combination of two effects. First, the salinity in the water is lower and secondly, that the activity concentration of ¹³⁷Cs in the sea water is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident. Comparing those results with the levels found at the coastal sampling stations in the period 2000-2007 (Gäfvert et al., 2003; NRPA, 2004, NRPA 2005, NRPA 2006, NRPA 2007, NRPA 2008 and NRPA, 2009), one can see that the activity concentration of ¹³⁷Cs in *F. vesiculosus* has been relatively stable in recent years. However, data from monthly sampling at Utsira (Figure 5.8) show that the activity concentration of ¹³⁷Cs is slowly decreasing in seaweed. This is in agreement with the reported temporal trend of ¹³⁷Cs in Baltic Sea sea water (Ikäheimonen et al. 2009).



Figure 5.8. Levels of ^{137}Cs (Bq kg⁻¹ d.w.) in Fucus vesiculosus sampled along the Norwegian coastline in 2009.



Figure 5.8. Average activity concentration (Bq kg⁻¹ d.w.) from monthly sampling in seaweed (Fucus vesiculosus) from Utsira in the period 1999-2009 (data from IFE).

5.4 Caesium-137 in fish, molluscs and crustaceans

Samples of cod from the Barents Sea have been analysed for ¹³⁷Cs since the early 1990s. In Figures 5.9 and 5.10 the activity concentration of ¹³⁷Cs in muscle tissue from cod caught in two areas of the Barents Sea is shown. Almost all samples from both locations are below 1 Bq kg⁻¹ (w.w.), and in recent years below 0.5 Bq kg⁻¹ (w.w.). The results show a slightly decreasing level of ¹³⁷Cs in the period 1992-2009.



Figure 5.9. Activity concentration of ${}^{137}Cs$ (Bq kg⁻¹ w.w.) in cod from the Barents Sea (the area around Bjørnøya) sampled in the period 1992 to 2009.



Figure 5.10. Activity concentration of 137 Cs (Bq kg⁻¹ w.w.) in cod from off the coast of Finnmark sampled in the period 1991 to 2009.

Cs-137 levels in fish species, crustaceans and molluscs caught in Norwegian marine waters in 2008 and 2009 are shown in Table 5.1, 5.2,

5.3 and 5.4. All obtained results were below 1.5 Bq kg⁻¹ (w.w.).

Table 5.1. Activity concentrations ($Bq kg^{-1} w.w.$) of ^{137}Cs in fish and shrimps caught in the North Sea, the Barents and the coastal waters of Finnmark, Troms, Nordland and Trøndelag, in 2008.

Species	Location	No. of samples (total no. of fish	137 Cs in muscle tissue (Ba kg ⁻¹ w w)
species			(Dq kg w.w.)
Cod		20 (551)	0.06 ± 0.02 to 0.22 ± 0.02
(Gadus morhua L.)			
Haddock		14 (22.6)	0.000 0.000 0.014 0.04
(Melanogrammus		14 (326)	0.009 ± 0.002 to 0.14 ± 0.04
Aeglefinus)			
Saithe		1 (1)	0.20 + 0.02
(Pollachius virens)			0.20 ± 0.02
Long Rough Dab			0.07 ± 0.02 to 0.15 ± 0.02
(Hippoglossoides		12 (300)	0.07 ± 0.02 to 0.13 ± 0.03
Platessoides)			
Capelin		1.5 kg (8 pooled)	< 0.10 to 0.21 ± 0.08
(Mallotus villosuss)		1.5 kg (8 pooled)	<0.10 to 0.21± 0.08
Polar Cod		1.5 kg (5 pooled)	< 0.50 to 0.15 ± 0.03
(Boreogadus Saida)		1.5 kg (5 pooled)	<0.50 to 0.15 ± 0.05
Greenland Halibut			< 0.30 to 0.14 ± 0.04
(Reinhardtius		3 (75)	<0.50.10 0.14 ± 0.04
Hippoglossoides)			
Golden Redfish	Barents Sea	5 (125)	<0.10 to 0.06 ± 0.02
(Sebastes Marinus)	Burents Beu	5 (125)	
Golden Redfish		1.5 kg (1 pooled)	0.05 ± 0.02
(Sebastes mentella)			
Blue Whiting		2 (50)	
(Micromesistius		2 (50)	0.10 ± 0.01 to 0.13 ± 0.03
Poutassou)			
Norway pout		2 (50)	<0.50 to 0.05 ± 0.01
(Triopterus esmarkii)			
Spottet Cat-fish		1(1)	0.09 ± 0.03
(Anarnichas minor)			
(Triglops murrayi)		1.5 kg (1 pooled)	<0.10
Shrimps		$1.5 \log (10 \mod 1 d)$	$(0.10 \pm 1.1 \pm 0.2)$
(Pandalus borealis)		1.5 kg (10 pooled)	<0.10 to 1.4 ± 0.2
Starfish		1.5 kg (2 pooled)	~1.5
(Echinoderma)		1.5 kg (2 pooled)	<1.5
Cod larvae		1.5 kg (3 pooled)	< 0.40 to 0.03 ± 0.1
(Gadus morhua L.)		1.5 kg (5 pooled)	<0.40 10 0.05 ± 0.1
Cod		11 (425)	0.13 ± 0.03 to 0.44 ± 0.11
(Gadus morhua L.)			
Haddock		3 (75)	< 0.30 to 0.11 ± 0.04
(Melanogrammus		5(15)	<0.50 to 0.11 ± 0.04
Aeglefinus)	Coastal waters of		
(Dolla ching winong)	Finnmark and Troms	1 (25)	<0.30
(Pollachius virens)			
(Cluped harmony)		2 (50)	<0.10 to 0.08 ± 0.02
Long Rough Dab			
(Hinnoglassoides		2 (50)	0.12 ± 0.03
Platessoides			
i micssones/	1		

Capelin (Mallotus villosuss)		1.5 kg (1 pooled)	0.07 ± 0.04
Blue Whiting (Micromesistius Poutassou)		1 (25)	0.14 ± 0.02
Mackerel (Scomber scombrus)		1 (25)	<0.30
Shrimps (Pandalus borealis)		1.5 kg (2 pooled)	<0.40
Krill (Euphasiscea)		1.5 kg (1 pooled)	<0.30
Cod (Gadus morhua L.)	Coastal waters of Nordland	8 (200)	0.22 ± 0.08 to 0.41 ± 0.13
Salmon (Salmo salar)	Farmed fish from coastal waters of Troms and Nordland	2 (50)	0.34 ± 0.11 to 0.37 ± 0.12
Cod (Gadus morhua L.)		2 (50)	0.17 ± 0.03 to 0.21 ± 0.03
Haddock (Melanogrammus Aeglefinus.)		1 (25)	0.12 ± 0.04
Saithe (Pollachius virens)	The North Sea	2(50)	0.28 ± 0.03 to 0.32 ± 0.03
Atlantic herring (Clupea harengus)		1 (25)	<0.70
Mackerel (Scomber scombrus)		1 (25)	0.26 ± 0.06
Norway pout (Triopterus esmarkii)		1 (25)	0.11 ± 0.03

Table 5.2 Activity concentrations (Bq kg⁻¹ w.w.) of 137 Cs in fish caught in the Barents Sea and in the coastal waters of Finnmark, Troms and Trøndelag 2009.

Species	Location	No. of samples (total no. of fish muscles)	¹³⁷ Cs in muscle tissue (Bq kg ⁻¹ w.w.)
Cod (Gadus morbua L)		5 (275)	0.11 ± 0.10 to 0.20 ± 0.10
Haddock (Melanogrammus Aeglefinus)	Barents Sea	3 (75)	0.10 ± 0.10 to 0.10 ± 0.10
Atlantic herring (Clupea harengus)		1 (25)	0.08 ± 0.10
Long Rough Dab (Hippoglossoides Platessoides)		5 (125)	0.07 ± 0.07 to 0.20 ± 0.10
Capelin (Mallotus villosuss)		1.5 kg (4 pooled)	<0.10 to 0.05 ± 0.05
Polar Cod (Boreogadus Saida)		1.5 kg (2 pooled)	<0.10 to 0.06 ± 0.06
Greenland Halibut (Reinhardtius Hippoglossoides)		3 (75)	0.10 ± 0.10 to 0.25 ± 0.10
Golden Redfish (Sebastes Marinus)		2 (50)	0.13 ± 0.10 to 0.14 ± 0.10
Deepwater Redfish (Sebastes mentella)		3 (75)	<0.30 to 0.10± 0.10

Blue Whiting (Micromesistius Poutassou)		1 (25)	0.10 ± 0.10
Norway pout (Triopterus esmarkii)		1 (25)	0.06 ± 0.06
Cod larvae (Gadus morhua L.)		1.5 kg (1 pooled)	0.05 ± 0.10
Haddock larvae (Melanogrammus Aeglefinus)		1.5 kg (1 pooled)	0.06 ± 0.10
Atlantic herring larvae (Clupea harengus)		1.5 kg (1 pooled)	<0.10
Shrimps (Pandalus borealis)		1.5 kg (3 pooled)	0.06 ± 0.10 to 0.10 ± 0.10
Krill (Euphausiacea)		1.5 kg (2 pooled)	< 0.10 to 0.06 ± 0.06
Amphipoda (amphipoda)		1.5 kg (1 pooled)	<0.10
Cod (Gadus morhua L.)	Coastal waters of Finnmark and Troms	5 (275)	<0.10 to 0.20 ± 0.10
Saithe (Pollachius virens)		2 (50)	0.10 ± 0.10 to 0.17 ± 0.10
Atlantic herring (Clupea harengus)		1 (25)	0.12 ± 0.10
Blue Whiting (Micromesistius Poutassou)		1 (25)	0.10 ± 0.10 to 0.11 ± 0.10
Norway pout (Triopterus esmarkii)		1 (25)	<0.10
Shrimps (Pandalus borealis)		1.5 kg (1 pooled)	0.10 ± 0.10
Cod (Gadus morhua L.)	Coastal waters of	8 (200)	0.21 ± 0.06 to 0.35 ± 0.12
Saithe (Pollachius virens)	Nordland and Trøndelag	2 (50)	0.23 ± 0.09 to 0.27 ± 0.10
Salmon (Salmo salar)	Farmed fish from coastal waters of Finnmark and Trøndelag	2 (50)	0.22 ± 0.08 to 0.29 ± 0.11

Table 5.3. Activity concentrations ($Bq kg^{-1} w.w.$) of ¹³⁷Cs in molluscs and crustaceans caught in the Barents and the coastal waters of Finnmark in 2008 and 2009.

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Location	2008	2009
	137 Cs (Bq kg ⁻¹ w.w.)	137 Cs (Bq kg ⁻¹ w.w.)
	King crab (Paralithodes camtschaticus)	King crab (Paralithodes camtschaticus)
Coastal waters of		-0.10
Finnmark	-	<0.10
	Sea cucumber (Holothundea)	Sea cucumber (Holothundea)
Barents Sea	<0.10	-
	Sea anemone (Urticina)	Sea anemone (Urticina)
Barents Sea	<0.03	-
	Seafan (Phakellia ventilabrum)	Seafan (Phakellia ventilabrum)
Barents Sea	<0.03	-

Different species of bivalves have been sampled at 12 different location along the Norwegian coast (Figure 5.11) and analysed with respect to 137 Cs. The results are presented in Table 5.4.

Location	2008	2009
	Blue mussel	Blue mussel
	(Mytilius edulis)	(Mytilius edulis)
	137 Cs (Bq kg ⁻¹ w.w.)	137 Cs (Bq kg ⁻¹ w.w.)
Hvaler	0.26 ± 0.06	0.14±.0.3
Tønsberg	<0.10	<0.06
Egersund	<0.10	< 0.05
Stigevika	<0.10	<0.09
Vemmelsvik	<0.10	<0.05
Molde	0.08 ± 0.02	<0.09
Åfjorden	0.10 ± 0.02	<0.09
Sløvefjorden	<0.10	< 0.05
Bodø	0.28 ± 0.14	-
Kjempebakken	<0.10	< 0.05
	2008	2009
	Horse mussel	Horse mussel
	(Modiolus modiolus)	(Modiolus modiolus)
Vadsø	<0.10	<0.07
	2008	2009
	Great scallops	Great scallops
	(Pecten maximus)	(Pecten maximus)
Horva	-	0.12±.0.2

Table 5.4. Activity concentrations ($Bq kg^{-1} w.w.$) of ^{137}Cs in mussel soft tissue in 2008 and 2009.



Figure 5.11. Sampling stations for mussels in 2008 and 2009.

6 Summary and conclusions

In 2008 and 2009 samples of sea water, sediment, and seaweed were collected in the Barents Sea, the North Sea, the Skagerrak and at a number of coastal stations and fjords. Data on radioactivity levels and trends in these areas together with data on liquid discharges from nuclear installations and recent trends in such discharges are summarised in this report.

6.1 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2008 and 2009 are within authorised limits according to the estimates made by the operator. The effective dose to a member of the critical group from liquid radioactive discharge at Kjeller was estimated to be 0.30 μ Sv in 2008 and 0.0053 μ Sv in 2009, which correspond to 30 % and 0.53 % of the dose limit, respectively. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be 0.016 μSv in 2008 and 0.011 μSv in 2009, corresponding to 1.6 % and 1.1 % of the dose limit. respectively. The discharge of anthropogenic radionuclides from Norwegian sources is only detectable in the environment close to each discharge point and has no significant impact on the large-scale distribution of these radionuclides in the marine environment.

Produced water from offshore oil production may contain enhanced levels of naturally occurring radium isotopes. The reported total discharge of ²²⁶Ra and ²²⁸Ra in produced water from Norwegian platforms in 2008 was 460 GBq and 370 GBq, respectively, where about 40 % was discharged from 2 platforms, Troll B and Troll C. In 2009, the discharged activity of ²²⁶Ra and ²²⁸Ra was 480 GBq and 360 GBq, respectively.

The liquid discharges of alpha- and betaemitting radionuclides from European nuclear installations to the North-East Atlantic have, according data reported by OSPAR, decreased markedly in the period 1990 to 2008, and are now at their lowest level during this period. Data from OSPAR also shows that the annual discharge of ²²⁶Ra from all offshore oil and gas installations in the North Sea in the period 2005-2008 have been relatively stable in the range from 0.78 to 0.90 TBq

The long-range transports of radionuclides originating from nuclear weapons fallout, the Chernobyl accident and from reprocessing of spent nuclear fuel are still the main contributors to anthropogenic radionuclides found in Norwegian waters. In addition to direct discharges from reprocessing, water from the Baltic Sea and remobilisation of ¹³⁷Cs and plutonium from contaminated Irish Sea sediments act as secondary sources of radionuclides to the Norwegian marine environment.

6.2 Radioactivity in sea water and sediment

In 2008 and 2009, samples of sea water and sediment were collected in the North Sea, the Skagerrak and the Barents Sea and analysed with respect to ¹³⁷Cs, ⁹⁹Tc, ⁹⁰Sr, ²³⁹⁺²⁴⁰Pu and ²⁴¹Am. A general trend seen in most samples is that the levels of radioactivity are similar, or slightly lower than have been observed in recent years. This can be explained by reduced discharges, radioactive decay and other processes such as sedimentation and dilution.

6.2.1 Caesium-137 in sea water

Observed levels of ¹³⁷Cs in surface water in the North Sea, the Skagerrak and the Barents Sea in 2008 and 2009 ranged from 1.4 to 35.4 Bq m⁻³. The highest levels were found in brackish water in the Skagerrak. Samples from the Barents Sea in 2009 showed an activity concentration in the range from 1.5 to 2.3 Bq m⁻³. This is generally similar or lower than the activity concentrations observed in the same area in the period 2002 to 2006. Data from Hillesøy in the period 2002 to 2009 also show that the levels of ¹³⁷Cs are slowly decreasing in the Norwegian coastal current.

6.2.2 Technetium-99 in sea water

Samples of sea water were collected mainly in the North Sea and the Barents Sea. Levels of ⁹⁹Tc in surface sea water ranged from 0.03 Bq m⁻³ to 2.0 Bq m⁻³. Average activity concentration from monthly sampling at Hillesøy in 2008 and 2009 was 0.44 ± 0.13 and 0.35 ± 0.09 , respectively, which is significantly lower than the peak values observed in 1999/2000.

6.2.3 Strontium-90 in sea water

The activity concentration in surface sea water sampled in the Barents Sea and in the Skagerrak in 2008 and 2009 ranged from 0.7 Bq m⁻³ to 2.8 Bq m⁻³. The highest concentrations were found off the southern coast of Norway in the Skagerrak. The levels of ⁹⁰Sr in the Barents Sea are generally lower than those observed in 2002. Data from Hillesøy in the period 2002 to 2009 indicates that the levels of ⁹⁰Sr are slowly decreasing in the Norwegian coastal current.

6.2.4 Plutonium-239+240 and americium-241 in sea water

Observed levels of $^{239+240}$ Pu in the Barents Sea and the Skagerrak in 2008 and 2009 ranged from 1.4 to 12.4 mBq m⁻³. The activity concentrations of $^{239+240}$ Pu are similar to those found in the same area in 2002 and 2005.

The activity concentration of ²⁴¹Am in sea water from the Barents Sea and the Skagerrak ranged from 0.9 to 2.4 mBq m⁻³. This is similar to what was observed in 2002 and 2005 in the same area.

6.2.5 Radium-226 in sea water

The activity concentration of ²²⁶Ra in the Barents Sea ranged from 0.9 to 2.4 Bq m⁻³. This is similar to the levels observed in 2005 in the same area.

6.3 Radioactivity in biota

6.3.1 Technetium-99 in seaweed

Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 35 to 115 Bq kg⁻¹ (d.w.). For most stations the levels were lower in 2008 and 2009 compared to observed levels in the period 2002 - 2007. The levels in 2008 and 2009 are significantly lower than the peak values observed in the period 1999-2001. Monthly sampling at Hillesøy and Utsira show that the levels of ⁹⁹Tc in seaweed have decreased since 2005 due to the reduced discharge of ⁹⁹Tc from Sellafield.

6.3.2 Caesium-137 in seaweed

The activity concentration of 137 Cs in *Fucus vesiculosus* sampled at the coastal stations in 2008 and 2009 was in the range from < 0.2 to 3.3 Bq kg⁻¹ (d.w.), where the highest levels were found in Skagerrak. Data from monthly sampling at Utsira indicates that the levels of 137 Cs in seaweed are slowly decreasing.

6.3.3 Caesium-137 in fish

Different species of fish and, commercially important and others, have been sampled in the Barents Sea and the North Sea. The activity concentration of ¹³⁷Cs in fish from Norwegian marine waters is generally low. All analysed samples were below 1.5 Bq kg⁻¹ (w.w.).

Samples of cod from two locations in the Barents Sea have been analysed for ¹³⁷Cs since the early 1990s. These results show a slightly decreasing trend of ¹³⁷Cs in this period.

7 References

Amundsen et al., 2003

Amundsen I, Brekken A, Liland A. Utslipp av radioaktive stoffer fra Sellafield-anleggene. En gjennomgang av britiske myndigheters regulering av utslippstillatelser. StrålevernRapport 2003:2. Østerås: Statens strålevern, 2003 (In Norwegian, English summary).

AREVA, 2010

AREVA, 2010. Liquid releases annualy report 2009. <u>http://www.areva.com/EN/operations-</u> 2331/liquid-releases-annualyreport.html#tab=tab3 (28.03.11)

Bourlat et al., 1996

Bourlat Y, Milliès-Lacroix J-C, Le Petit G, Bourguignon J. ⁹⁰Sr, ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu in world ocean water samples collected from 1992 to 1994. In: Guéguéniat P, Germain P, Métivier H, eds. Radionuclides in the oceans. Les Ulis: Institute de Protection et de Surete Nucleaire / Les editions de physique, 1996: 75-93.

Brown et al., 2002

Brown J, Iospje M, Kolstad AK., Lind B, Rudjord AL, Strand P. Temporal trends for ⁹⁹Tc in Norwegian coastal environments and spatial distribution in the Barents Sea. Journal of Environmental Radioactivity 2002; 60: 49-60.

Carlsson L. and Erlandsson B., 1991 Carlsson L, Erlandsson B. Effects of salinity on the uptake of radionuclides by *Fucus vesiculosus* L. Journal of Environmental Radioactivity 1991; 13: 309-322.

Chen et al., 1991

Chen Q et al. Determination of plutonium in environmental samples by controlled valence in anion exchange. RISØ-M-2856. Roskilde: Risø National Laboratory, 1991.

Cook et al., 1997

Cook GT, Mac Kenzie AB, Mc Donald P and Jones SR . Remobilization of Sellafieldderived radionuclides and transport from the North-East Irish Sea. Journal of Environmental Radioactivity 1997; 35: 227-241.

Dahlgaard, 1995 Dahlgaard H. Transfer of European coastal pollution to the Arctic: Radioactive tracers. Marine Pollution Bulletin 1995; 31: 3-7.

Dahlgaard, 2002

Dahlgaard H. Baltic ¹³⁷Cs outflow through the Danish Straits indicates remobilisation. In: Strand P, Børretzen P, Jølle T, eds. International conference on radioactivity in the environment, Monaco 2002. Extended abstracts. Österås: Norwegian Radiation Protection Authority, 2002: 516-520.

Environment Agency *et al.*, 2010 Environment Agency *et al.*, 2010. Radioactivity in Food and the Environment, 2010. RIFE 15. <u>http://www.cefas.co.uk/publications/rife/rife15</u> .<u>pdf</u> (28.03.11)

Evans, 1991

Evans S. Impacts of the Chernobyl fallout in the Baltic Sea ecosystem. In: Moberg L, ed. The Chernobyl fallout in Sweden. Stockholm: Swedish Radiation Protection Institute, 1991: 109-127.

Gäfvert et al. 2003

Gäfvert T, Føyn L, Brungot AL, Kolstad AK, Lind B, Christensen GC, Strålberg E, Drefvelin J, Rudjord AL.. Radioactivity in the marine environment 2000 and 2001. Results from the Norwegian National Montoring Programme (RAME). StrålevernRapport 2003:8. Østerås: Norwegian Radiation Protection Authority, 2003.

Harvey et al. (1992)

Harvey BR, Williams KJ, Lovett MB, Ibbett RD. Determination of technetium-99 in environmental material with rhenium as a yield monitor. Journal of Radioanalytical and Nuclear Chemistry 1992; 158: 417-436.

Holm et al. 1984

Holm E, Rioseco J, Garcia-Leon, M. Determination of ⁹⁹Tc in environmental samples. Nuclear Instruments and Methods in Physics Research 1984; 223: 204-207.

Holm et al. 1986

Holm E et al. Technetium-99 in algae from temperate and arctic waters of the North Atlantic. In: Desmet G, Myttenaere C. Technetium in the environment. EUR 10102. London: Elsevier Applied Science 1986: 53-59.

IAEA, 1990

IAEA (International Atomic Energy Agency). The Environmental Behaviour of Radium. Vol. 1. Technical Reports Series No. 310. ISBN 92-0-125090-9. Vienna, 1990

IFE, 2006a

Institutt for energiteknikk, IFE. Måling av radioaktive stoffer i utslipp til luft, i avløpsvann og i prøveobjekter fra IFE Kjellers omegn og Nitelva. Rapport for 2005. IFE Rapport nr: IFE/I-2006/012. Kjeller: IFE, 2006.

IFE, 2007a

Institutt for energiteknikk, IFE. Måling av radioaktive stoffer i utslipp til luft, i avløpsvann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen - Rapport for 2006. IFE Rapport nr: IFE/I-2007/007. Kjeller: IFE, 2007.

IFE, 2008a

Institutt for energiteknikk, IFE. Kontroll av radioaktive stoffer i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2007. IFE Rapport nr: IFE/I-2008/005. Kjeller: IFE, 2008.

IFE, 2009a

Institutt for energiteknikk, IFE. Kontroll av radionuklider i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2008. IFE Rapport nr: IFE/I-2009/010. Kjeller: IFE, 2009.

IFE, 2010a

Institutt for energiteknikk, IFE. Kontroll av radionuklider i utslipp til luft og vann og i prøveobjekter fra IFE Kjellers omegn, Nitelva og Himdalen. Rapport for 2009. IFE Rapport nr: IFE/I-2010/011. Kjeller: IFE, 2010.

IFE, 2006b

Institutt for energiteknikk, IFE. Utslipp fra drift av Haldenreaktoren i 2005. Sv-rapport 666. Halden: IFE, 2006.

IFE, 2007b

Institutt for energiteknikk, IFE. Utslipp fra drift av Haldenreaktoren i 2006. Sv-rapport 693. Halden: IFE, 2007.

IFE, 2008b

Institutt for energiteknikk, IFE. Utslipp fra drift av Haldenreaktoren i 2007. Sv-rapport 726. Halden: IFE, 2008.

IFE, 2009b

Institutt for energiteknikk, IFE. Utslipp fra drift av Haldenreaktoren i 2008. Sv-rapport 784. Halden: IFE, 2009.

IFE, 2010b

Institutt for energiteknikk, IFE. Utslipp fra drift av Haldenreaktoren i 2009. Sv-rapport 818. Halden: IFE, 2010.

Ikäheimonen et al., 2009

Ikäheimonen TK *et al.* Radioactivity in the Baltic Sea : inventories and temporal trends of ¹³⁷Cs and ⁹⁰Sr in water and sediments. Journal of Radioanalytical and Nuclear Chemistry. Vol. 282, No. 2, pp 419-425.

Kolstad, 1995

Kolstad AK. Expeditions to «Komsomolets» in 1993 and 1994. StrålevernRapport 1995:7. Østerås: Norwegian Radiation Protection Authority. (In Norwegian)

Leonard et al., 1999

Leonard KS, McCubbin D, Blowers P, Taylor BR. Dissolved plutonium and americium in surface waters of the Irish Sea, 1973-1996. Journal of Environmental Radioactivity 1999; 44: 129-158.

NRPA, 2003.

NRPA. Radioactivity in the marine environment 2000 and 2001. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2003:08. Østerås: Norwegian Radiation Protection Authority, 2003.

NRPA, 2004.

NRPA. Radioactivity in the marine environment 2002. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2004:10. Østerås: Norwegian Radiation Protection Authority, 2004.

NRPA, 2005

NRPA. Radioactivity in the marine environment 2003. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2005:20. Østerås: Norwegian Radiation Protection Authority, 2005.

NRPA, 2006.

NRPA. Radioactivity in the marine environment 2004. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2006:14. Østerås: Norwegian Radiation Protection Authority, 2006.

NRPA, 2007.

NRPA. Radioactivity in the marine environment 2005. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2007:10. Østerås: Norwegian Radiation Protection Authority, 2007.

NRPA, 2008.

NRPA. Radioactivity in the marine environment 2006. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2008:14. Østerås: Norwegian Radiation Protection Authority, 2008.

NRPA, 2009.

NRPA. Radioactivity in the marine environment 2007. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2009:15. Østerås: Norwegian Radiation Protection Authority, 2008.

OLF, 2009

Oljeindustriens Landsforening, OLF. Miljørapport 2009. Olje- og gassindustriens miljøarbeid. Stavanger: OLF, 2009. Utslippsdata fra de enkelte innterningene på norsk sokkel: <u>http://www.olf.no/PageFiles/6508/OLF%20Mi</u> <u>lj%c3%b8rapport%202009%20webversjon_sis</u> <u>te.pdf</u> (17.01.11)

OLF, 2010

Oljeindustriens Landsforening, OLF. Miljørapport 2010. Olje- og gassindustriens miljøarbeid. Stavanger: OLF, 2010. Utslippsdata fra de enkelte innterningene på norsk sokkel:

http://www.olf.no/PageFiles/6508/OLF%20Mi lj%c3%b8rapport%202009%20webversjon_sis te.pdf (17.01.11)

OSPAR, 2010a

OSPAR Commission, Discharges of radioactive substances from the non-nuclear sectors in 2008. Radioactive substances series, Publication number 516/2010. London: OSPAR, 2010.

OSPAR, 2010b

OSPAR Commission. Liquid discharges from nuclear installations in 2008. Radioactive substances series, Publication number 515/2010. London: OSPAR, 2010.

Roos et al., 1994

Roos P, Holm E, Persson RBR. Comparison of AMP precipitate method and impregnated $Cu_2[Fe(CN)_6]$ filters for the determination of radiocesium concentrations in natural waters. Nuclear Instruments and Methods in Physics Research A 1994; 339: 282-286.

Rudjord et al., 2001

Rudjord AL, Føyn L, Brungot AL, Kolstad AK, Heldal HE, Brown J, Iosjpe M, Christensen G. Radioactivity in the marine environment 1999. StrålevernRapport 2001:9. Østerås: Norwegian Radiation Protection Authority, 2001.

Strand, 1998

Strand P, Balonov M, Aarkrog A, Bewers MJ, Howard B, Salo A, Tsaturov YS. Radioactivity. In: AMAP assessment report: Arctic pollution issues, Chapter 8. Oslo: Arctic Monitoring and Assessment Programme, AMAP 1998: 525-620.

Swift, 1985

Swift DJ. The accumulation of ^{95m}Tc from sea water by juvenile lobsters (*Homarus gammarus*). Journal of Environmental Radioactivity 1985; 2: 229-243.

UNSCEAR (2000a)

UNSCEAR. United Nations Scientific Committee on the Effects on Atomic Radiation. Sources and effects of ionizing radiation: United Nations report to the general assembly, with scientific annexes. Volume I: Sources. New York: United Nations, 2000. <u>http://www.unscear.org/unscear/en/publication</u> <u>s/2000_1.html</u> (17.01.11)

UNSCEAR (2000b) UNSCEAR. United Nations Scientific Committee on the Effects on Atomic Radiation, Sources and effects of ionizing radiation: United Nations report to the general assembly, with scientific annexes. Volume II: Effects. New York: United Nations, 2000. <u>http://www.unscear.org/unscear/en/publication</u> <u>s/2000_2.html</u> (17.01.11)

Vakulovsky et al., 1995 Vakulovsky SM, Kryshsev II, Nikitin AI. Radioactive contamination of the Yenisei River. Journal of Environmental Radioactivity 1995; 29: 225-236.

Varskog et al., 1997

Varskog P, Bjerk TO, Ruud AB. Ph-control EDTA titration as an alternative method for the determination of the chemical yield of yttrium in ⁹⁰Sr analysis. In: Rapid radioactivity measurements in emergency and routine situations. Teddington: National Physical Laboratory, NPL, 1997: 237-241.

Zalewska and Saniewski, 2009 Zalewska T, Saniewski M.¹³⁷Cs and ⁹⁰Sr activities in sea water in 2008 the southern Baltic region. Gdynia: Institute of Meteorology and Water Management, Maritime Branch, 2009.

http://meeting.helcom.fi/c/document_library/ge t_file?p_1_id=19548&folderId=457982&name =DLFE-37745.pdf (17.01.11)

Appendix

Analytical methods

During recent years, laboratories at the Norwegian Radiation Protection Authority, and the Institute of Marine Research have both been accredited according to the requirements of NS-EN ISO/IEC 17025. Laboratories and organisations that are not named here have not been accredited according to the requirements of NS-EN ISO/IEC 17025. The scope and conditions governing the accreditation of the institutes mentioned above are quite different, but they have all been accredited for gamma spectrometric measurements. For example, the NRPA is accredited for gamma spectrometric measurements in the energy interval 100-1800 (except 226 Ra), while the other keV organisations are accredited for gamma spectrometric measurements of ¹³⁷Cs. The analytical techniques employed at each institution are described below. The results in this report are not claimed to be accredited results as not all results are accredited.

The Norwegian Radiation Protection Authority (NRPA)

NRPA analyses alpha and gamma emitters, using the procedures described in this Appendix.

Detection of gamma emitters

At NRPA analyses of gamma-emitting nuclides are performed with HPGe detectors. All the detectors are situated in a low-background laboratory to ensure low detection limits. The HPGe detectors have relative efficiencies in the range of 23 % to 40 %. The resolution of the detectors, the full width at half maximum (FWHM) at 1332 keV, was less than 1.9 keV. Three of the detectors cover the energy interval 50-2000 keV, and two cover the interval 20-2000 keV.

Caesium-absorbing filters (sometimes also the prefilter) from the filtering system for sea water samples were dried separately at 105°C and ashed at 450°C before the activity was determined with an HPGe detector. The counting time varied from 1 to 4 days.

Samples of seaweed were dried at 105°C and homogenized, and placed in containers prior to gamma counting. Sediment samples were freeze-dried and placed in containers prior to gamma counting. Samples were counted for a minimum of 2 days. At NRPA, the detection limits for ¹³⁷Cs and ¹³⁴Cs in the sediment samples ranged from approximately 0.3-2 Bq kg⁻¹ (d.w.) Uncertainties in the analysis include uncertainties from counting statistics, calibration and sample preparation and are normally in the range 5% to 20%.

Determination of ²³⁹⁺²⁴⁰Pu and ²⁴¹Am

The concentrations of ²³⁹⁺²⁴⁰Pu and ²⁴¹Am were measured in samples of 200 litres of sea water or 10-20 g of sediment. ²⁴²Pu and ²⁴³Am were added for chemical yield deter-mination. Precipitation from the water samples was achieved using the analytical procedure described by Chen et al., (1991). Different radiochemical separation techniques were applied to separate plutonium and americium from other nuclides using solvent extraction with 10 % TIOA/xylene solution and ion-exchange chromatography with a BIO-RAD AG1-X4 (100-200 mesh) column. Purified americium and plutonium fractions were electrodeposited on stainless steel discs and the activity measured in semiconductor silicon detectors. alpha spectrometry is not able to As distinguish between ²³⁹Pu and ²⁴⁰Pu, because the energies of their emitted alpha particles are too close to be resolved, these isotopes are measured and reported as the sum $^{239+240}$ Pu. Relative efficiencies of the detectors were in the range 25 to 30 %. The resolution of the detectors, the full width at half maximum (FWHM), was approximately 20 keV at 5486 keV (²⁴¹Am). Chemical yields obtained from the ²⁴²Pu and ²⁴³Am yield monitor were in the range 40 % to 80 %.

Determination of 226Ra

Radium was coprecipitated with MnO₂ from 5-10 l of sea water, after ¹³³Ba had been added as a yield determinant. The MnO₂ precipitate was then dissolved in hydrochloric acid and hydrogen peroxide, and after the addition of sulphuric acid, K_2SO_4 and $Pb(NO_3)_2$, radium was coprecipitated with $PbSO_4$. The precipitate was dissolved in EDTA at pH = 10, and transferred to a liquid scintillation vial. After measuring the radiochemical yield with gamma spectrometry, a liquid scintillation cocktail was added (OptiFluor O). The sample was then stored for about four weeks after which the activity of ²²⁶Ra was measured through alpha peaks from short-lived radon daughters with a low-background, liquid scintillation counter (Quantulus).

The Institute of Marine Research (IMR)

Detection of gamma emitters

Sediment samples were transported deepfrozen to IMR, where they were subsequently ground, freeze-dried, homogenized and transferred to polyethylene containers of appropriate size prior to analysis. The gamma counting for ¹³⁷Cs detection was performed in IMR's low-background laboratory using a HPGe detector with 30 % relative efficiency and an HPGe detector with 60 % relative efficiency. Both detectors have electric cryostat cooling systems, and 10 cm lead shielding.

Determination of 99Tc

Before the activity concentration of ⁹⁹Tc can be determined, a radiochemical separation is necessary. Briefly, rhenium is added as a yield monitor in the form of KReO₄, and a preliminary extraction of ⁹⁹Tc (and Re) based on anion-exchange separation is performed. After iron hydroxide scavenging, ⁹⁹Tc and Re are further extracted by a second anionexchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts are isolated. ⁹⁹Tc is betacounted using а low-background anticoincidence beta counter (Model Risø GM-25-5), and the yield of the rhenium tetraphenyl arsonium salt is determined gravimetrically. See Harvey et al. (1992) for a detailed description of the method.

Institute for Energy Technology (IFE)

Determination of 99Tc

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to ⁹⁹Tc content. The pretreatment was performed at Kjeller and the analysis at the Department of Radiation Physics, at Lund University. The

following analytical procedure was used at the Department of Radiation Physics at Lund University, Sweden. Technetium was extracted into tri-butyl-phosphate (TBP) from sulphuric acid-hydrogen fluoride solution. Technetium was then back-extracted from the organic phase with a sodium hydroxide solution from which the technetium was electrodeposited onto stainless steel discs, and ^{99m}Tc was used as a radiochemical yield determinant. After decay of the yield determinant, ⁹⁹Tc was measured with an anti-coincidence-shielded GM counter (Holm *et al.*, 1984).

Determination of 90Sr

For ⁹⁰Sr, the standard method using fuming nitric acid was used. The recovery of ⁹⁰Sr in the analytical process was monitored by adding ⁸⁵Sr as a yield determinant, and the recovery of the daughter nuclide ⁹⁰Y was determined by titration with EDTA (Varskog *et al.*, 1997). Finally, ⁹⁰Y was measured with an anticoincidence-shielded GM counter.

Detection of gamma emitters

At IFE analyses of gamma-emitting nuclides are performed with low-background HPGe detectors. Samples of seaweed were dried at 105°C and then homogenized, and placed in suitable containers prior to gamma counting.

Determination of 239+240Pu

Samples of dried and homogenized *Fucus vesiculosus* were ashed and treated with *aqua regia* before separation of plutonium isotopes.²⁴²Pu was used as a tracer for radiochemical yield determination. The separation process involved solvent extraction with 10 % TIOA/Xylene followed by anion exchange on AG1-X4 columns. The plutonium fraction was then electrodeposited on a stainless steel disc and counted using alpha spectrometry.



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StrålevernRapport 2011:2 Måling av naturlig ultrafiolett stråling i Norge

StrålevernRapport 2011:3 Radioecological consequences after a hypothetical accident with release into the marine environment involving a Russian nuclear submarine in the Barents Sea

StrålevernRapport 2011:4 Radioactivity in the Marine Environment 2008 and 2009