StrålevernRapport 2006:21

Differences in technetium-99 accumulation and distribution between organs in male and female lobsters collected from Norwegian coastal waters

Statens strålevern

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Key words:
Technetium-99, Sellafield, concentration factor, lobster, *Homarus gammarus*

Abstract:
In this report, ^{99}Tc results for lobsters caught along the Norwegian coast between 2001 and 2005 are presented. The high accumulation of ^{99}Tc in lobsters compared to other crustecea is confirmed in this report, as well as the clear differences between males and females. Concentration factors have been estimated for each gender and also for different organs.

Referanse:
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Emneord:
Technetium-99, Sellafield, konsentrasjonsfaktor, hummer, *Homarus gammarus*

Resymé:
I denne rapporten presenteres ^{99}Tc resultater for hummer innsamlet langs norskekysten i perioden 2001 og 2005. Rapporten viser en høy akkumulering av ^{99}Tc i hummer og stor forskjell mellom hunner og hanner. Konsentrasjon faktorer er beregnet for hunner og hanner, og for forskjellige organer.

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Østerås, 2006

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1 Introduction

Between 1994 and 2004, elevated levels of technetium-99 (^{99}Tc) were discharged to the Irish Sea from the reprocessing facilities at Sellafield. ^{99}Tc is transported with ocean currents to Norwegian waters (Figure 1), and earlier studies have shown that lobsters have a high ability to accumulate this radionuclide and that female lobsters displays higher concentration in edible fraction than males (Swift, 1985 and 2001, Busby et al., 1997).

Since 2001, a more extensive sampling programme of lobsters (*Homarus gammarus)* in Norwegian Coastal areas has been carried out, in most cases with simultaneous sampling of seawater. Lobsters display considerable variation in the ^{99}Tc distribution among various organs. Previous research has shown clear differences in the organ distributions of ^{99}Tc in male and female lobsters (Smith, 1998 and Knowles et al., 1998). This report presents the results of ^{99}Tc measurements of lobsters from different locations along the Norwegian coast in 2001-2005. The activity concentration in lobster tail muscle, differences between gender and organ distribution has been investigated.

1.1 Technetium-99

The fission product ^{99}Tc, is produced by decay of Molybdenum-99 (^{99}Mo), in nuclear reactor operations or in atmospheric nuclear weapon tests. Figure 2 displays the decay sheme of ^{99}Mo. Metastable ^{99m}Tc decays by gamma emission to ^{99}Tc. ^{99}Tc is produced owing to the short half-life of ^{99m}Tc. The long half-life of ^{99}Tc (213 000 years) means that the radionuclide will persist in the environment for thousands of years.

In seawater, ^{99}Tc is present as the highly soluble pertechnetate ion TcO_4^-, allowing it to be transported over fairly long distances far away from its discharge point. ^{99}Tc is transported to Norwegian Coastal current via the North Sea.

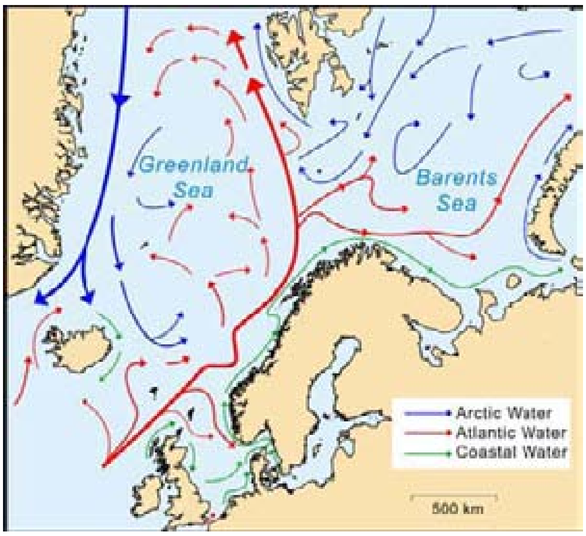


Figure 1. *Shematic of the surface circulation of the Northern Seas that are involved in the transport of ^{99}Tc to the Norwegian waters (adapted from Loeng, 1998).*

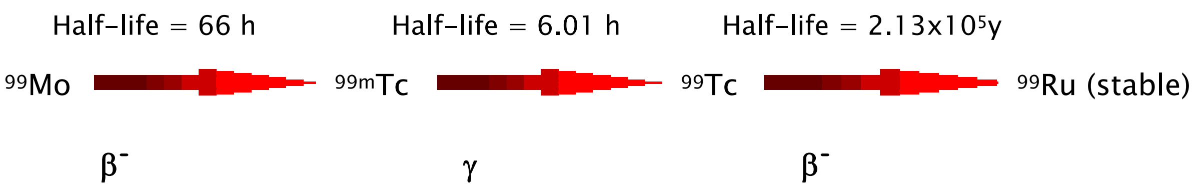


Figure 2. *Decay sheme of Molybdenum-99 (^{99}Mo).*

1.2 Sources of Technetium-99 to Norwegian waters

In the last ten years, elevated levels of ^{99}Tc have been detected in Norwegian waters. The sources are discharges from European reprocessing plants, fallout from atmospheric nuclear weapon tests and wastes from research laboratories and hospitals. However, the main source of ^{99}Tc is discharges from the reprocessing plant at Sellafield in UK.

1.2.1 Fallout from Nuclear Weapon testing

The Norwegian waters received ^{99}Tc fallout directly from the nuclear test explosions in the 1950 and 1960's. To put the relative sources of ^{99}Tc into an international context, around 85 % of the global inventory is associated with discharges from European reprocessing plants, the remaining 15 % is associated with weapons tests (Dahlgaard, 1995).

1.2.2 Discharges from European reprocessing plants

Since the 1950-1960's, ^{99}Tc has been released to the ocean through controlled discharges from the nuclear reprocessing facilities at Sellafield (UK) and Cap la Hague (France). Figure 3 shows ^{99}Tc annual discharges from Sellafield during the period 1952-2005. Throughout the 1980's and early 1990's, ^{99}Tc was discharged from Sellafield at a rate of 1.9 to 6.6 TBq/year, following an earlier peak release of 180 TBq/year in 1978. In 1994 and 1995, the discharges of ^{99}Tc from the Sellafield plant increased sharply and the higher levels have led to an increase in ^{99}Tc concentration in Norwegian coastal waters.

After 10 years with heavy international pressure, the discharge of ^{99}Tc has been reduced in 2004 and onwards. The new decontamination technique removed more than 90 % of the ^{99}Tc in the waste, using tetraphenylphosphoniumbromide (TPP) as a precipitant. The last release before new technologies came into operation was in April 2003.

The ^{99}Tc releases from Cap de la Hague are far lower than from Sellafield and only in the 1980's the releases of ^{99}Tc from La Hague was higher compared to Sellafield. The ^{99}Tc discharges from La Hague have been reduced by a factor of 100 between 1989 and 2004 peaking in the mid 1970's. Reported discharges from Sellafield and La Hague in 2004 are 14.3 TBq and 0.079 TBq, respectively. Back in the late eighties, the discharges from nuclear reprocessing in Western Europe remained the dominant source of ^{99}Tc in Norwegian waters and today the situation is still the same.

1.2.3 Chernobyl Accident

After the Chernobyl accident, maximum seawater concentration of ^{99}Tc most likely occurred in the Baltic Sea. However, Aarkrog et al., (1988) were unable to detect any additional ^{99}Tc inventory, and in the late eighties, they concluded that the Chernobyl accident did not contribute significantly to the ^{99}Tc levels in the north-eastern Atlantic Ocean. However, Illus et al., (2004) observed small amounts of ^{99}Tc in bladder wrack (*Fucus vesiculosus)* from the Finnish coastal waters in 1999 and 2003. The activity concentrations in Finish coastal waters in 1999 ranged from 1.6 Bq/kg (dry weight) to 11.6 Bq/kg (dry weight) and from 2.9 Bq/kg (dry weight) to 6.7 Bq/kg (dry weight) in 2003. In both years, the highest concentrations were found in samples collected at stations furthest to the north in the Golf of Botnia. In seawater and all other biota samples, the activity concentration of ^{99}Tc was below the detection limit and Illus *et al.* concluded that besides the global fallout in the 1950 and 1960's a small contribution from Chernobyl is possible.

1.2.4 Wastes from hospitals and research laboratories

Metastable, ^{99m}Tc, the shorter-lived isotope is primarily used as a medical diagnostic tool, and it can be found as a component of industrial and institutional wastes from hospitals and research laboratories. ^{99m}Tc with a half-life of 6.01 hours is used for diagnostic purpose in bone, kidney, heart and lung scans. ^{99m}Tc decays by gamma emission to ^{99}Tc and the radionuclide will primarily be released to the marine environment through the sewage systems. ^{99}Tc is produced owing to the short half-life of ^{99m}Tc in quantity less than 0.1 % (proportional to the ratio between half-lives, 6.01 hours to 210 000 years) and the use of ^{99m}Tc in medicine makes a relatively small contribution.

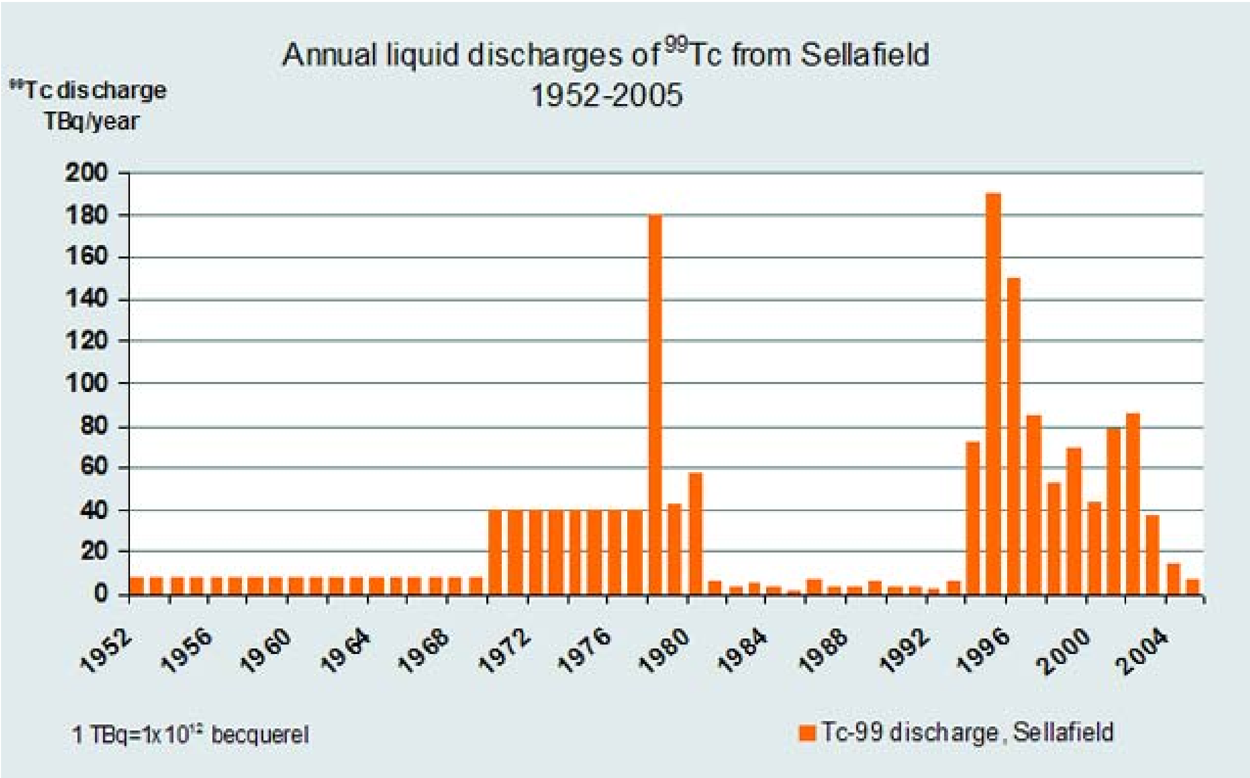


Figure 3. *Annual liquid discharges of* ^{99}*Tc (TBq) from Sellafield in the period 1952-2005.*

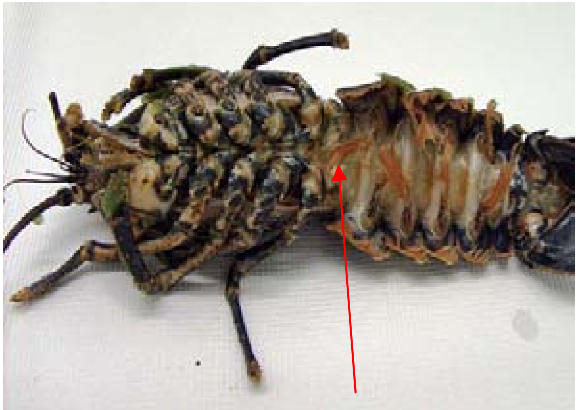
2 Materials and methods

2.1 Sampling areas

All lobster and seawater samples were obtained along the Norwegian coast in the period 2001-2005. European lobsters (*Homarus gammarus)* were collected at seven locations; Kvitsøy (2001), Stefjord (2001), Værlandet (2002), Flekkerøy (2002), Møre (2002), Tristein (2003) and Sandøya (2003, 2004, 2005). Geographic overviews of the sampling areas are shown in Figure 4. All lobsters have been analyzed for ^{99}Tc.

Figure 4. *Geographic overview of lobster sampling areas (red) and coastal stations (blue)*

Lobsters collected at Værlandet were delivered by the Institute of Marine Research, IMR. From the other sites, lobsters were bought in local shops or delivered to the NRPA by local fishermen immediately after they had been caught. The lobsters were frozen, and their gender, wet weight, carapace length and total length were recorded. The gender of each animal was determined by examining the first pair of swimmerets. The first set is hard and bony on a male, and soft and feathery on a female, see photo below.



Gender examining of a female. Photo: NRPA

2.2 Sampling of seawater

Lobster and seawater samples were collected simultaneously at Sandøya, Tristein, Kvitsøy and Værlandet. The ^{99}Tc concentration in seawater has been routinely measured around the Norwegian coastline since 1997 and the coastal sampling locations can be seen in Figure 4. The seawater concentrations were estimated at Stefjord, and Flekkerøy based on monitoring results from the regular coastal stations Hillesøy, Lista and Arendal. All seawater results refer to 50 liters of filtered water (<1µm).

2.3 Sampling of seaweed

^{99}Tc in seaweed (*Fucus vesiculosus)* has been regularly monitored at Arendal coastal station since 1999. The samples have been collected annually in the period 1999-2004 and twice a year since 2005. The station is located close to the lobster sampling site at Sandøya.

2.4 Sampling of lobsters

The European lobster is a long lived, dominant predator in Norwegian coastal waters (Figure 5). The lobsters were collected annually in October or November. A total of 103 lobsters were sampled, 43 females and 60 males (Table 1). The lobsters were frozen, and their gender, wet weight, carapace length and total length were recorded. On all occasions, all lobsters were divided by gender. The lobsters were dissected and subsamples of tail muscle tissue were prepared. The subsamples were dried to constant weight, then homogenised in a blender and analysed. In every individual, the tail muscle tissue was analysed for ^{99}Tc.

Figure 5. *European lobster (Homarus gammarus).*

Since 2003, the lobster sampling has mainly been carried out at Sandøya. Further research, has been performed on organ distribution of ^{99}Tc in lobsters from this location. 13 lobsters, six males and seven females and seawater samples were collected simultaneously. The lobsters were caught at about 10m depth. Each individual was dissected and subsamples of crusher claw muscle, cutter claw muscle, tail muscle, green gland, gills and a remainder fraction (including [1]hepatopancreas) was prepared and analysed separately The different fractions were dried to constant weight, then homogenised in a blender and finally analysed for ^{99}Tc.

[1] Lobster liver also called the tomalley, it retains contaminants, such as, nutrients and heavy metals.

Moult cycle effects on ^{99}Tc levels have been reported in lobsters (Swift, 1985) and therefore the sampling aimed to select only specimens in the intermoult stage with fully hardened shells.

Table 1. *Number of male and female lobsters sampled at different locations during the years 2001-2005.*

Location, Year	Sex (M/F)	Number of samples
Kvitsøy, 2001	M	17
	F	4
Stefjord, 2001	M	1
	F	1
Værlandet, 2002	M	5
	F	5
Flekkerøy, 2002	M	4
	F	2
Møre, 2002	M	6
	F	-
Tristein, 2003	M	3
	F	8
Sandøya, 2003-2005	M	24
	F	23
Total, 2001-2005	M	60
	F	43

2.5 Seawater and lobster analysis

Activity concentrations of ^{99}Tc in seawater and lobster samples were determined according to the method developed by Risø National Laboratory (Chen et al., 2001). To determine the activity concentration of ^{99}Tc in seawater, samples of 50 litres were filtered through a 1-micron polypropylene cartridge to remove suspended particulate matter. ^{99m}Tc was added to all samples for chemical recovery determination. Technetium is extracted by ion exchange chromatography from the water (AG 1-X4 resin, BIO-RAD 100-200 mesh). Lobster samples were dried, milled and homogenized. A 10-20 g dried sample was transferred to a specially designed bottle and ^{99m}Tc was added for chemical recovery determination. The sample was carbonised and then dissolved by adding concentrated H_2SO_4 followed by HNO_3. Technetium is separated from the matrix by ion exchange chromatography using AG 1-X4 resin (BIO-RAD 100-200 mesh). Ruthenium is eluated by 1M NaOH-0.1M EDTA-NaClO from the anion column and at the end TcO_4^- is stripped from the column by HNO_3. Further purification of ^{99}Tc by different separation techniques like precipitation of $Fe(OH)_3$ and solvent extraction by TIOA/xylene.

Finally, the samples were electrodeposited on stainless steel discs overnight with current 300 mA/2cm^2. For all samples, ^{99m}Tc is used as a yield monitor and the chemical recovery is achieved by gamma counting on a NaI well-detector. Typically, the chemical yields varied between 70 % and 85 %. After one week, the ^{99}Tc activity is measured using a low background anti-coincidence beta counter, Model Risø GM-25-5 (Bøtter-Jensen and Nielsen, 1989).

European Lobster (*Homarus gammarus*)

Widespread: The European Lobster is widespread from the Mediterranean in the south and up north to northern Norway. In Norway, the stock of European Lobster has steadily declined since 1950 and regulation has been introduced by the government. New regulation was introduced 1. May 2005.

Harvest season: between 1. October and 31. December.

Minimum legal size: 25 cm

3 Results and discussion

It has been shown that both primary pathways for the accumulation of ^{99}Tc in lobsters are directly uptake from surrounding seawater and consumption of contaminated food items at the sea floor (Smith et al., 1998 and Pentreath, 1981).

3.1 Technetium-99 in seawater

In order to assess the accumulation of ^{99}Tc in lobsters, the concentration of ^{99}Tc in the surrounding seawater has been studied. The discharges of ^{99}Tc are transported from the Irish Sea and the English Channel via the North Sea and into the Norwegian Coastal Current (NCC). NCC in eastern Skagerrak has a low salinity of about 25 ‰, due to high fraction of water originating from the Baltic Sea and river water. The NCC is mixed with Atlantic water and the salinity increases as it flows along the coast.

Observed and estimated activity concentrations of ^{99}Tc in seawater can be seen in Table 2. All seawater results refer to 50 liters of filtered surfacewater (<1µm). The sampling has been consistent in October or November. Samples of surface seawater and lobsters were simultaneously collected at Sandøya, Tristein, Kvitsøy and Værlandet. Unfortunately, seawater concentrations were not available at Stefjord, Møre and Flekkerøy. However, at these three locations seawater concentrations were estimated using results from nearby coastal stations. The ^{99}Tc concentration in seawater was considered to be 1.85Bq/m^3 at Flekkerøy, based on monitoring results from Lista (2.0 Bq/m^3) and Arendal (1.7 Bq/m^3). The ^{99}Tc concentration in seawater at Møre and Stefjord was estimated by using the same concentrations as from Værlandet and Hillesøy respectively. Hillesøy is a coastal station with monthly sampling of seawater.

The observed surface seawater concentrations of ^{99}Tc at Sandøya were 1.55 Bq/m^3 in 2003 and 1.25 Bq/m^3 in 2004. This is in good agreement with the concentrations recorded at Arendal, 12 km southwest of Sandøya, with concentrations of 1.60 Bq/m^3 and 1.15 Bq/m^3 in 2003 and 2004, respectively. The upper 30m of Skagerrak Coastal Water off Sandøya is to a large extent a mixture of water from the North Sea or Jutland Coastal water (75 %) and Kattegat Surface Waters (25 %) (Aure et al., 1998).

Table 2. *Salinity data and observed and estimated concentrations of ^{99}Tc (Bq/m^3) in one seawater sample at different locations.*

Sampling location	Year	Salinity (‰)	^{99}Tc (Bq/m^3)
Kvitsøy	2001	29.0	0.95 ± 0.11
Stefjord	2001	-	1.0[a]
Værlandet	2002	31.0	1.55 ± 0.16
Flekkerøy	2002	-	1.85[a]
Møre	2002	-	1.55[a]
Tristein	2003	28.4	1.25 ± 0.15
Sandøya	2003	30.5	1.55 ± 0.18
Sandøya	2004	27.1	1.25 ± 0.13
Sandøya	2005	32.3	1.10 ± 0.12

a = estimated value

The general observed salinity data in Skagerrak show a large variability over a year, between 25.0 ‰ and 32.0 ‰ (Sætre et al., 2003). The observed change in salinity measurements at Sandøya in 2004 and 2005 from 27.1 ‰ to 32.3 ‰ (20°C) is probably caused by a high amount water inflow of Atlantic water (Iversen et al., 2006).

Figure 6 displays the activity concentration of ^{99}Tc in seawater at the lobster sampling sites and at the regular coastal stations in southern Norway. All results between 2001 and 2005 are in the same range and the ^{99}Tc concentrations are less than 3.6 Bq/m^3. However, higher values were found at Lista compared with Tjøme and Arendal, in 2001 and 2004. The higher values at Lista could be explained by special wind conditions and long-range transportation of ^{99}Tc straight across the North Sea. There is a known pathway from the northern part of Scotland to the southern coast of Norway just north of 60 °N. The high salinity of 30 ‰ of the sample from Lista further supports this hypothesis.

It is expected that along the Norwegian coast, sufficiently distant from Sellafield, the ^{99}Tc will be well mixed in the water column and not likely to be influenced by the pulsed nature of the discharges of ^{99}Tc like in the Irish Sea. Samples collected off the Norwegian coast in 2001 showed similar to what was observed in 2004 (Figure 7a,b). This shows that the variations of seawater concentrations along the Norwegian coast in this study are moderate, in most cases between 1-2 Bq/m^3.

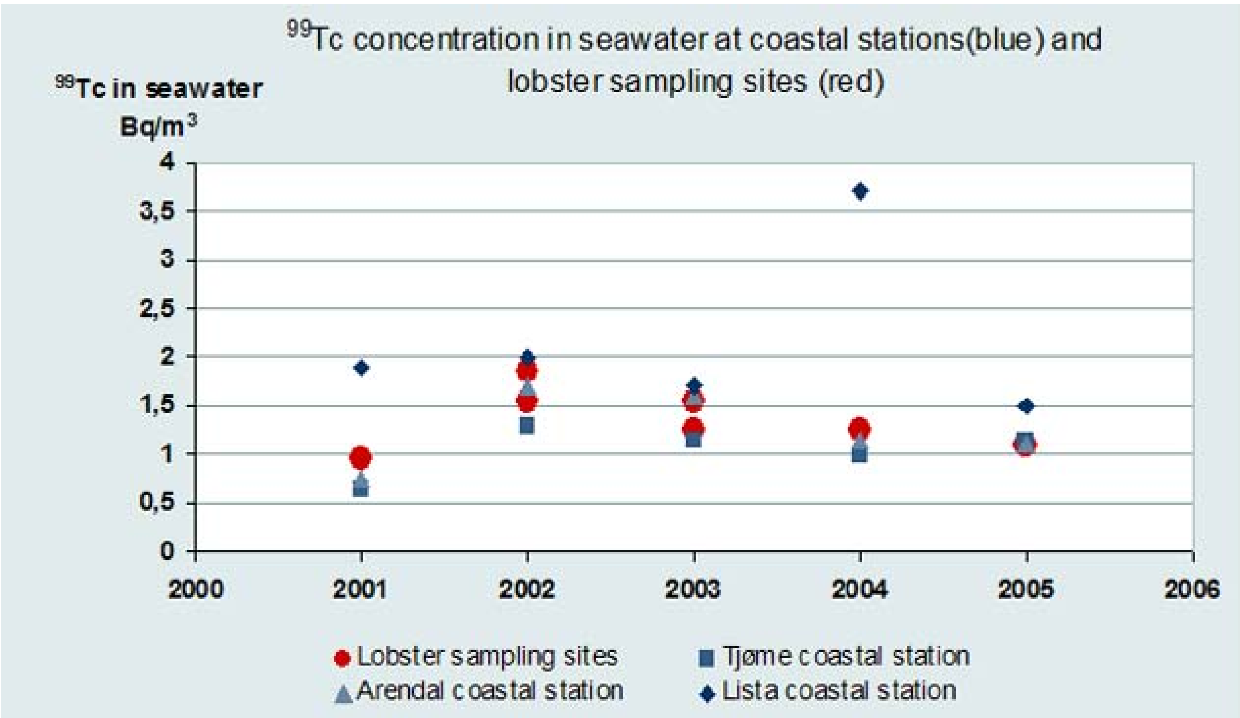


Figure 6. *Activity concentration of ^{99}Tc (Bq/m^3) in seawater samples collected at the lobster sampling sites (red dots) and regular coastal stations.*

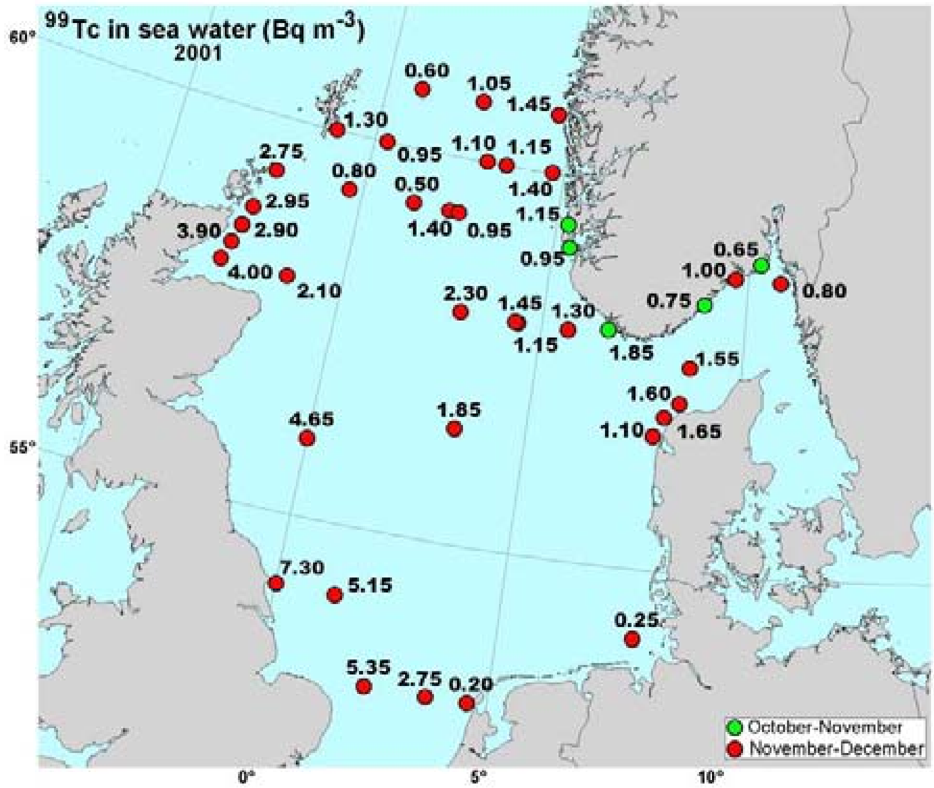


Figure 7a. *Activity concentration (Bq/m^3) of ^{99}Tc in seawater samples collected in 2001.*

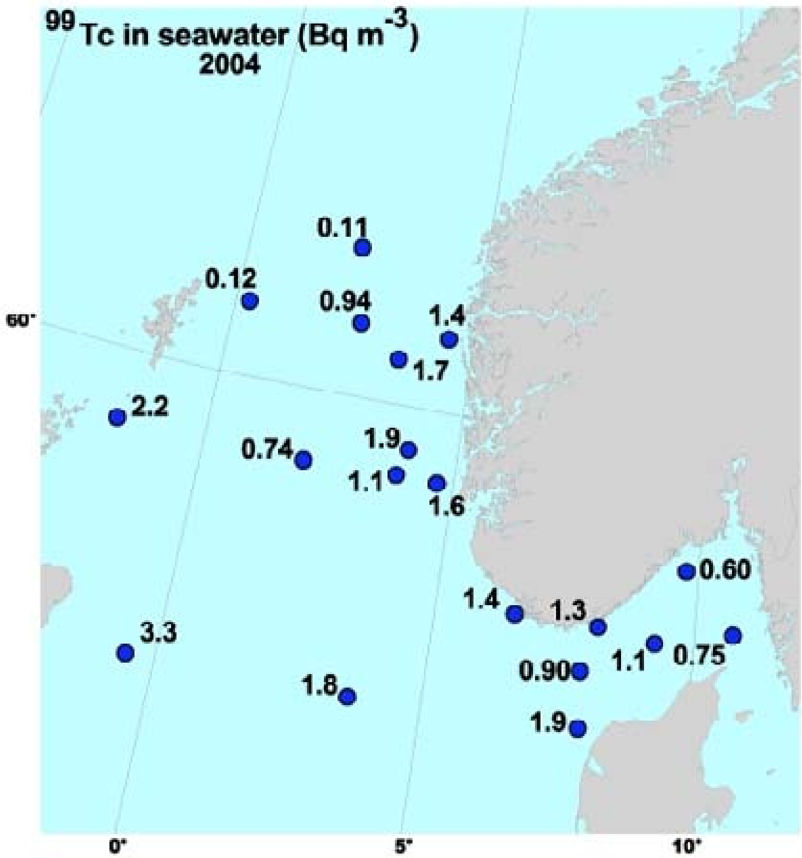


Figure 7b. *Activity concentration (Bq/m^3) of ^{99}Tc in seawater samples collected in 2004.*

3.2 Biometric data of lobsters

Of the 103 lobsters sampled, 43 were females and 60 males. The mean whole body weight for all lobsters was 736 g and ranged between 247 g and 3660 g (Table 3). Lobsters missing a claw were withdrawn from the whole live weight data set. The mean total body weights for female and male lobsters were 663 and 790 g, respectively. No significant differences between male and female lobsters were found in whole live weight (t-test: t=1.10, n=88, p=0.27), tail muscle live weight (t-test: 1.11, n=103, p=0.27), total body length (t-test: t=0.21, n=88, p=0.84) and carapace length (t-test: t=-0.3, n=103, p=0.77).

Table 3. *Lobster biometric data.*

	Whole live weight (g)	**Total body length (cm)**	**Tail muscle live weight (g)**	**Carapace length (cm)**
All				
Mean	736	29.4	99	12.7
SD	539	4.6	58	2.1
n	88	102	103	103
Min	247	24	26	8.5
Max	3660	45	342	20.3
Male				
Mean	791	29.5	105	12.7
SD	608	4.3	53	2.1
n	50	60	60	60
Min	320	24	34	8.5
Max	3660	45	333	17.2
Female				
Mean	663	29.3	92	12.8
SD	430	4.9	64	2.2
n	38	42	43	43
Min	247	24.2	26	9.7
Max	1998	45	342	20.3
t-test				
t-value	1.1	0.21	1.11	-0.3
p-value	0.27[a]	0.84[a]	0.27[a]	0.77[a]

a = not statistically significant

3.3 Technetium-99 in lobster tail muscle tissues

The ^{99}Tc concentrations in tail muscle of all lobsters are shown as a frequency distribution in Figure 8. The activity concentrations of the males seems to fall into two distinct groups 1-6 Bq/kg (wet weight) and 9-14 Bq/kg (wet weight), respectively. 85 % of all males have activity concentration less than 5.0 Bq/kg (wet weight). In comparison, all female lobsters had a ^{99}Tc concentration higher than 5.0 Bq/kg (wet weight).

Table 4 displays the ^{99}Tc concentration in tail muscle tissues obtained from different locations and years. In male lobsters, the ^{99}Tc concentration in tail muscle tissue ranged from 2.0 Bq/kg (wet weight) to 16 Bq/kg (wet weight). The ^{99}Tc concentration in female tail muscle tissue ranged from 5.1 Bq/kg (wet weight) to 70 Bq/kg (wet weight).

For male and female lobsters, the mean activity concentration of ^{99}Tc in tail muscle tissue was 5.5 Bq/kg (wet weight) and 22.5 Bq/kg (wet weight) respectively. The mean activity concentrations of ^{99}Tc in lobster tail muscle at different sampling locations for both genders are shown in Figure 9. The highest individual concentration of 70 Bq/kg (wet weight) was recorded in a female lobster caught at Sandøya in 2004 and this explains the high annual value in females this year. This female lobster was one of 3 analysed. The female was an outlier with a whole body weight of 1650 g, a whole body weight twofold higher than the mean value for both genders. The ^{99}Tc load in other organs as well, showed also maximum concentrations. However, there were no convincing reasons to remove the lobster from the data sets.

In some cases there was a large variability in ^{99}Tc activity concentrations in the tail muscle of lobster caught at the same location on the same day and of the same gender. This is consistent with the findings respectively Swift and Nicholson, found in 2001 in the Irish Sea. The reason for this is not clear. However, the most probable explanation for the large variations is related to age and moulting/reproduction cycle of the lobsters. Recently moulted specimens feed actively compared to specimens that are about to moult. Moult cycle effects on technetium levels have been reported earlier by Swift (1985).

Activity concentration of ^{99}Tc in lobster tail muscle did not differ significantly between the seven different locations, although a positive trend was found (Table 5. General linear model, Univariat analysis: SS=1011, df=6, p=0.061). Significant differences in concentration of ^{99}Tc in the tail muscle were found between male and female lobsters (General linear model, Univariat analysis: SS=3282, df=1, p=0.000). The ^{99}Tc concentrations are a factor of ~ 4 higher in females, confirming earlier observations from the Irish Sea (Swift and Nicholson, 2001). In comparison, Swift and Nicholson (2001) found a factor of 2 higher activities in female lobsters compared to males.

In female lobsters the concentration of ^{99}Tc in tail muscle increased with increasing body weight (Figure 10. Linear regression: R^2=0.51, n=37, F=38.8, p=0.000). No correlation was found for males (Linear regression: R^2=0.000, n=49, F=0.007, p=0.932). Our results are not in agreement with results reported by Swift et al. in 2001. They reported a decreasing concentration of ^{99}Tc in tail muscle with increasing body weight. However, in that study the lobsters were cooked in salted freshwater before preparation and ^{99}Tc analysis was performed.

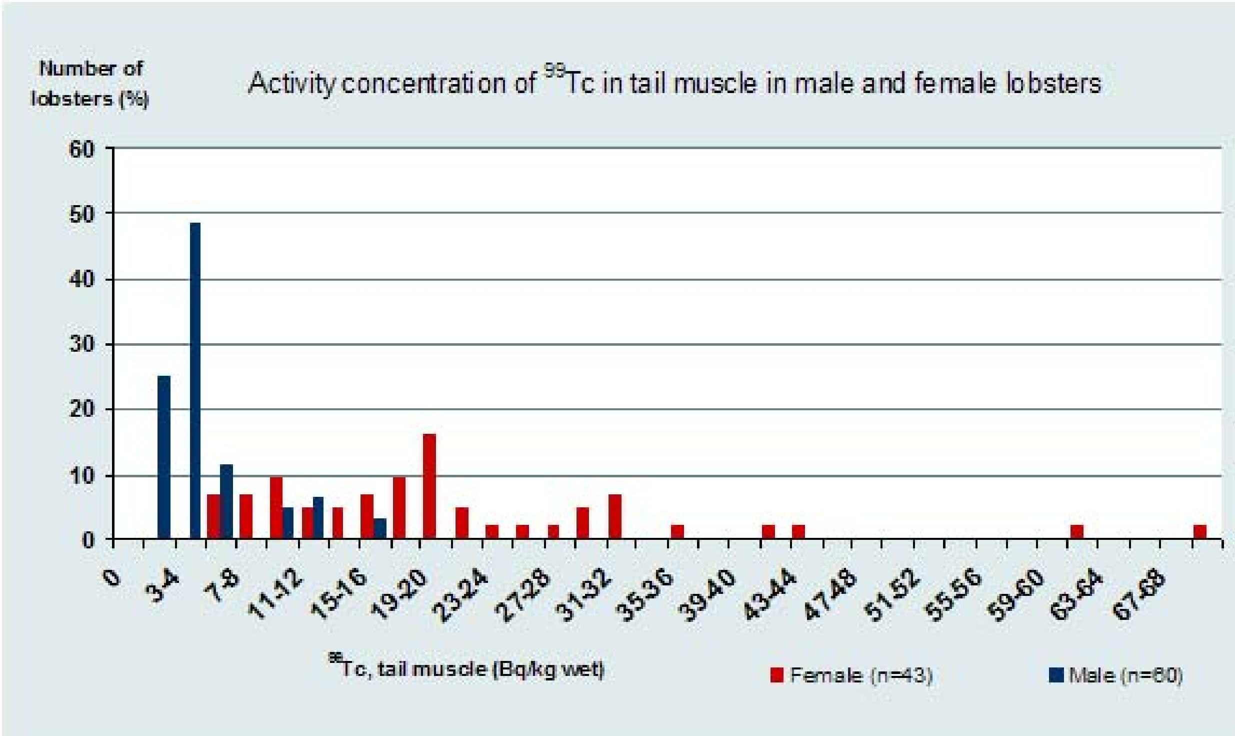


Figure 8. *Frequency distribution (%) of activity concentrations of* ^{99}Tc *in tail muscle (Bq/kg wet) in lobsters from all sampling areas.*

Table 4. *Activity concentration of* ^{99}Tc *(Bq/kg wet weight) in lobster tail muscle in males (M), females (F) and total from different locations sampled in 2001-2005.*

Location, Year	Sex (M/F)	Number of samples	Mean ^{99}Tc (Bq/kg wet)	SD	Min value	Max value
Kvitsøy, 2001	M	17	6.6	3.8	2.2	12.7
	F	4	34.2	4.9	31.1	41.4
Stefjord, 2001	M	1	2.8	-	2.8	2.8
	F	1	20.2	-	20.2	20.2
Værlandet, 2002	M	5	4.7	1.2	3.9	6.8
	F	5	27.9	19.8	14.9	62.0
Flekkerøy, 2002	M	4	6.7	6.2	3.0	15.9
	F	2	22.3		22.0	22.6
Møre, 2002	M	6	5.8	4.6	3.4	15.1
	F	-	-	-	-	-
Tristein, 2003	M	3	3.3	1.0	2.5	4.4
	F	8	19.7	8.0	9.3	36.5
Sandøya, 2003-2005	M	24	3.9	1.7	2.0	9.7
	F	23	18.4	14.5	5.1	70.0
Total, 2001-2005	M	60	5.1	3.3	2.0	15.9
	F	43	21.4	13.6	5.1	70.0

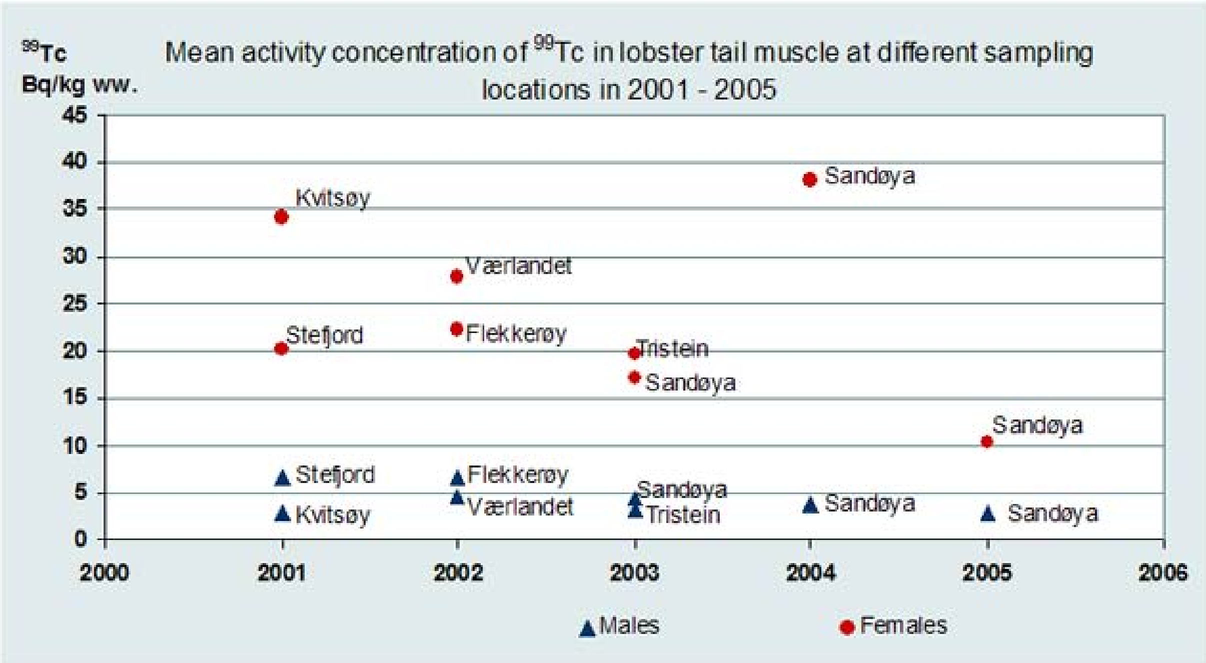


Figure 9a. *Mean activity concentration of ^{99}Tc in lobster tail muscle (Bq/kg wet weight) at different sampling locations during the years 2001-2005.*

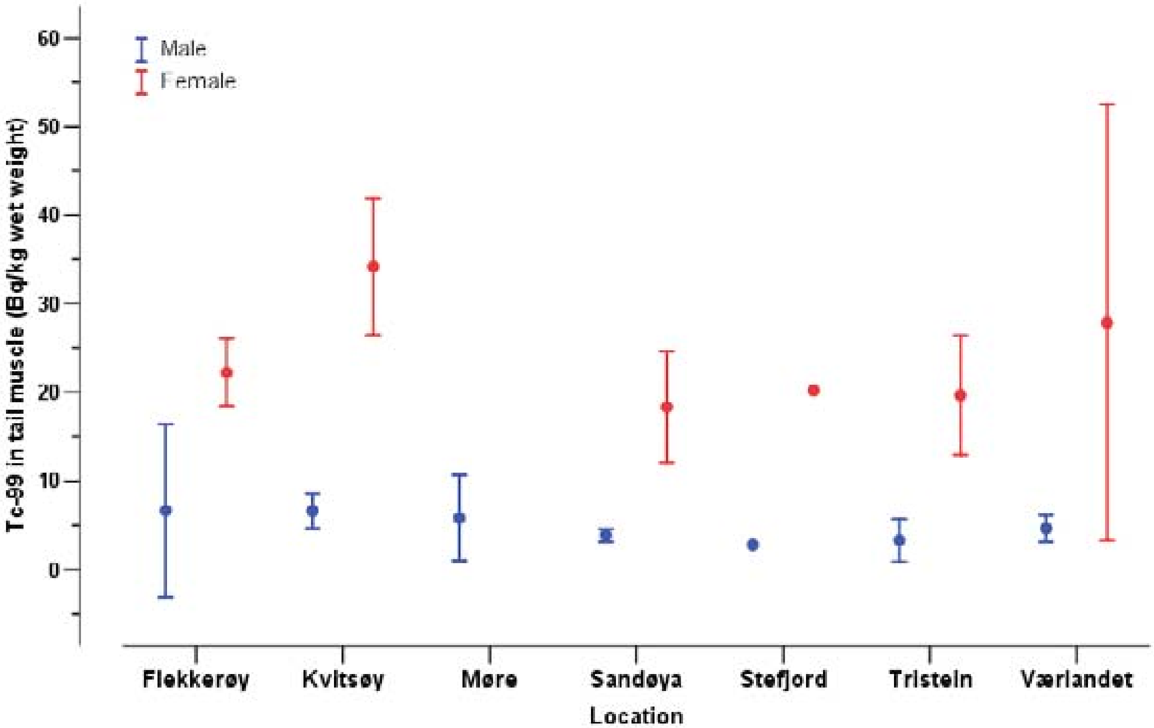


Figure 9b. *Mean activity concentration of ^{99}Tc (Bq/kg wet weight, ± standard deviation) for male and female lobsters at different locations.*

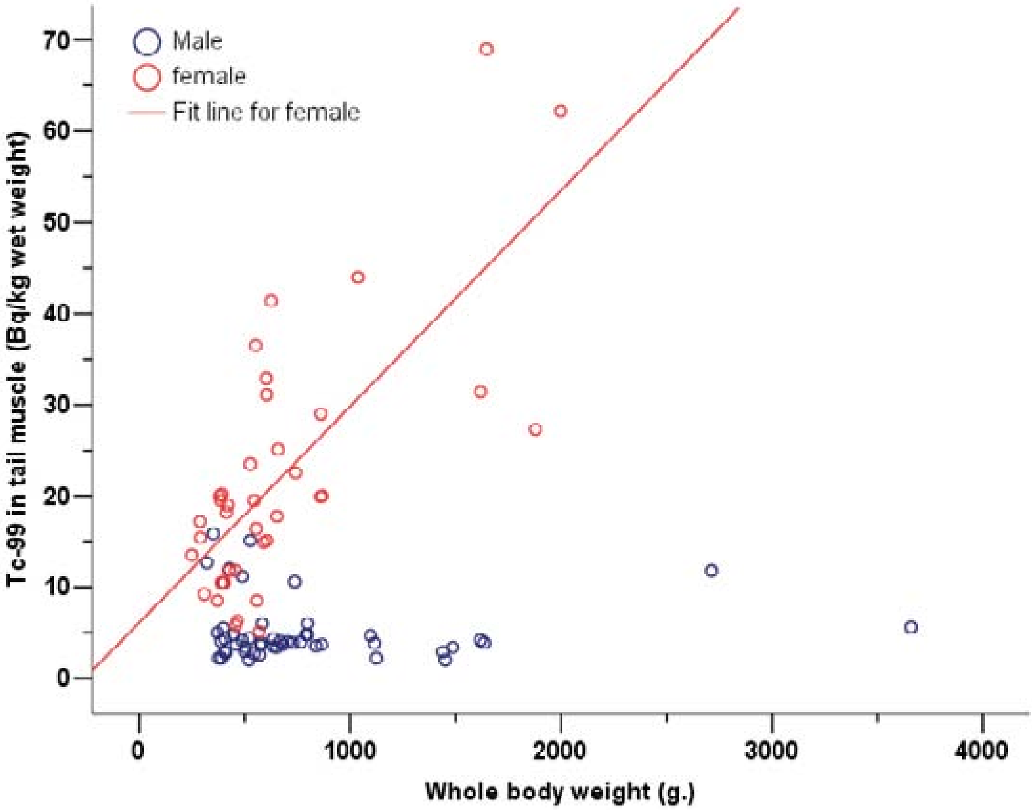


Figure 10. *^{99}Tc activity concentration (Bq/kg wet) in tail muscles for male and female lobster as a function of total body weight (g).*

Table 5. *General linear model (GLM), Univariate analysis of ^{99}Tc activity concentration in lobster tail muscle as dependent variable against different locations and sex.*

	SS	df	MS	F	p-value
Total	29703	103			
Location	1011	6	168	2.10	0.061
Sex	3283	1	3283	40.83	0.000
Error	7236	90	80.4		

3.4 Technetium-99 in lobsters from Sandøya

Lobsters have been observed annually at Sandøya since 2003 and the activity concentrations of ^{99}Tc in males and females are shown in Figure 11. Potential variations in concentration with gender and year were examined (Table 6). There was a significant difference between concentration of ^{99}Tc in tail muscle between males and females (General linear model, Univariat analysis: SS=2639, df=1, p=0.000). At Sandøya, the activity concentration of ^{99}Tc in tail muscle also varied between the years (General linear model, Univariat analysis: SS=866, df=2, p=0.007).

3.5 Technetium-99 in seaweed and lobsters

At Narestø, sampling of seaweed, *Fucus vesiculosus*, has been performed since 1999. The station is located close to the lobster sampling site at Sandøya. The activity concentrations of ^{99}Tc in *Fucus vesiculosus* show a transit time of around 3 years from Sellafield to Narestø (Figure 12). The increasing levels in 2004 and 2005 are the response to the increased discharges from Sellafield in 2001 and 2002 and onwards.

The response curve to discharges for male lobsters collected at Sandøya seems to be slower (Figure 13) and no increasing trend has been found in the last 3 years. There is no clear short-term pattern between the releases from Sellafield and uptake of ^{99}Tc to lobster in Norwegian waters, confirming earlier study in the Irsish Sea by Copplestone et al., (2004).

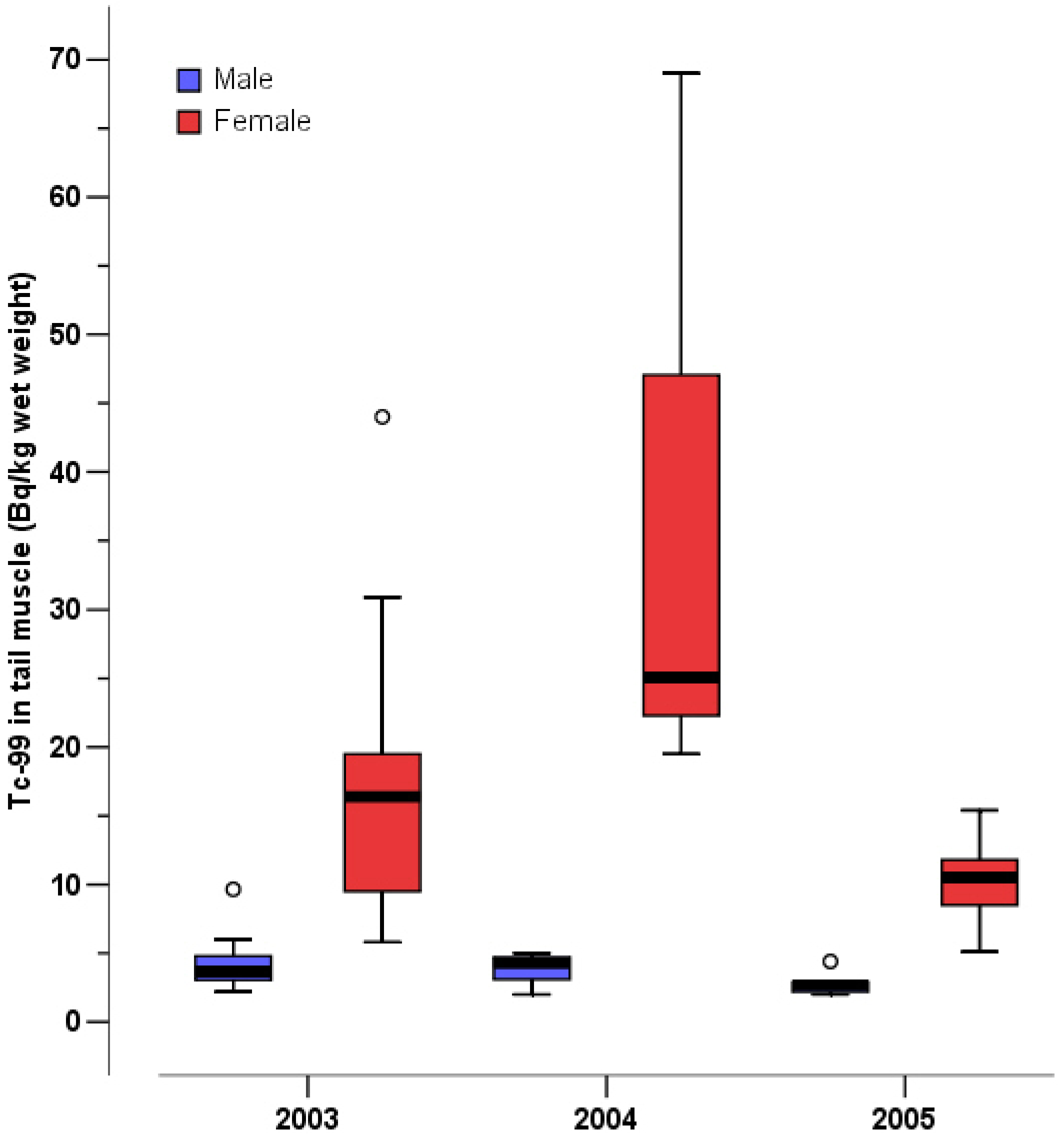


Figure 11. *Box plots of ^{99}Tc activity concentrations (Bq/kg wet) for males (blue) and females (red) collected at Sandøya with 95 % confidence interval and error bars. Horizontal lines for the median and open circles are outliers.*

Table 6. *General linear model (GLM), Univariate analyse with ^{99}Tc activity concentration in lobster tail muscle from Sandøya as dependent variable against year and sex.*

	SS	df	MS	F	p-value
Total	12796	47			
Year	866	2	432	5.59	0.007
Sex	2639	1	2639	34.05	0.000
Error					

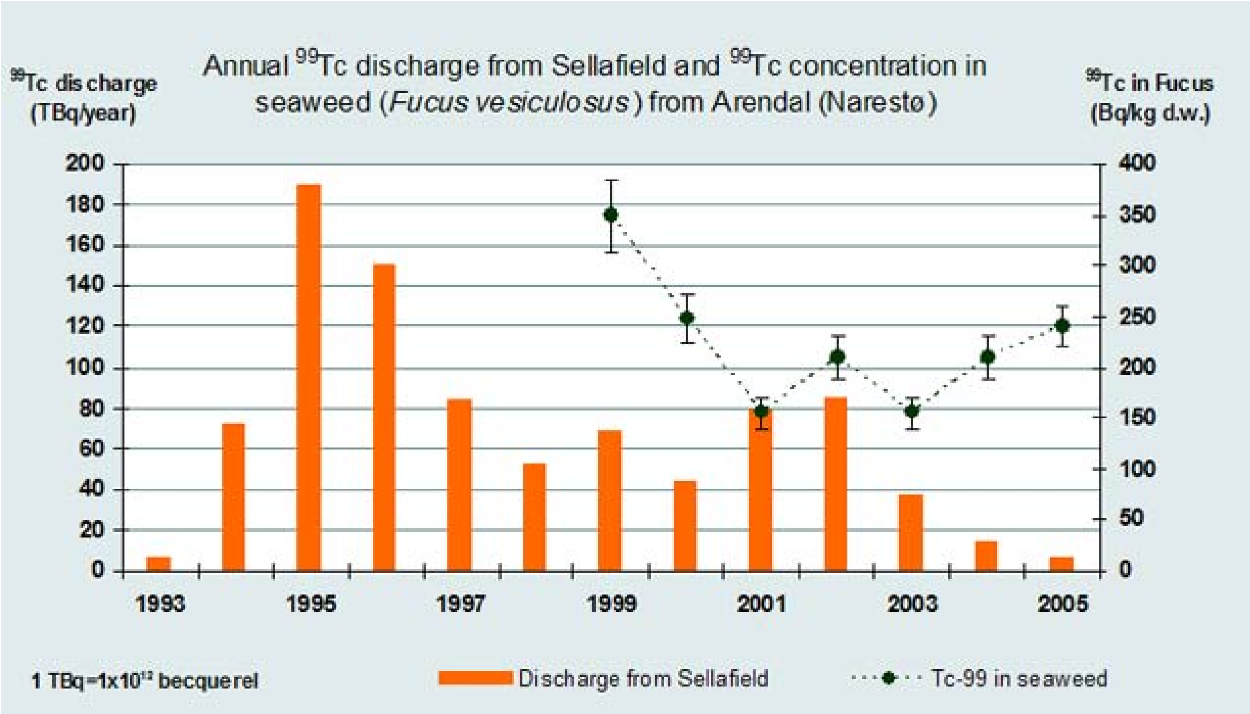


Figure 12. *Annual liquid discharge (TBq) of ^{99}Tc from Sellafield (primary axis) and annual ^{99}Tc activity concentration (Bq/kg dry weight) in Fucus vesiculosus at Narestø coastal station located at the southwest coast of Norway (± standard deviation).*

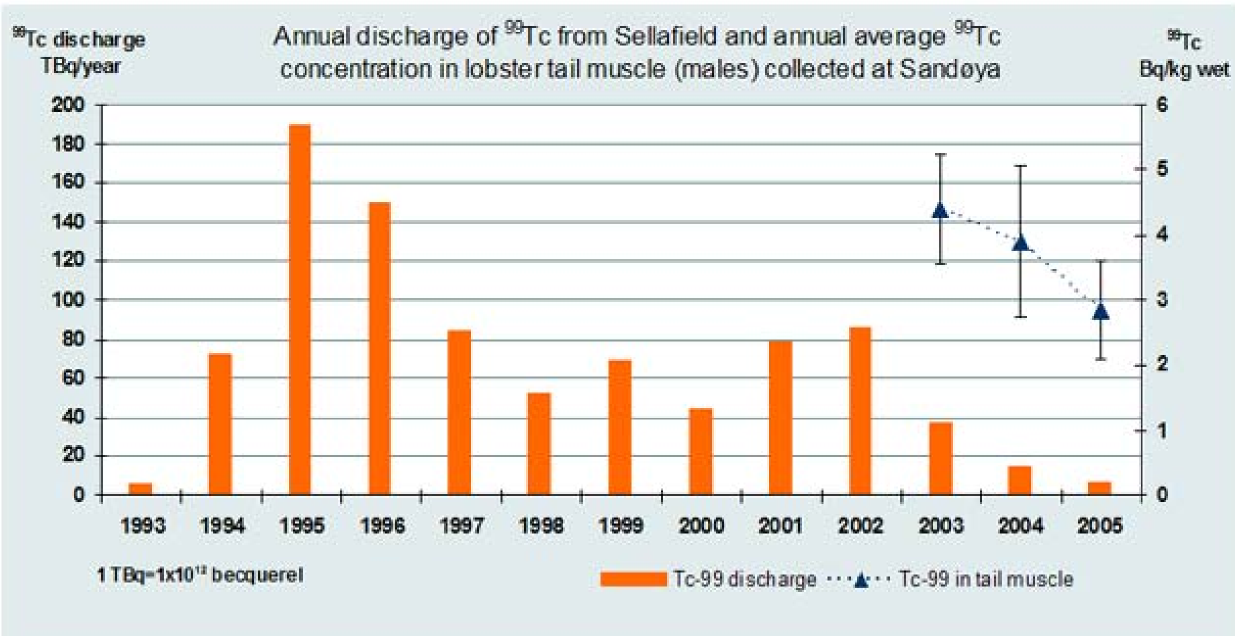


Figure 13. *Annual discharge (TBq) of ^{99}Tc from Sellafield (primary axis) and annual mean ^{99}Tc activity concentration (Bq/kg wet weight) in tail muscle tissue in male lobster collected at Sandøya (± standard deviation).*

3.6 Organ distribution

The data in chapter 3.3 has shown a clear difference of ^{99}Tc content in the tail muscle tissue between genders. The uptake by female lobsters is a factor of ~ 4 time's higher. In this chapter, uptake of ^{99}Tc in other organs is studied. Subsamples of tail muscle tissue, cutter claw, crusher claw, gills, green gland and a remainder fraction were analysed. For ^{99}Tc considerable variation in the ^{99}Tc distribution between individual organs was observed (Table 7). Like previous research has shown, the remainder fraction (including hepatopancreas) contained the largest fraction of ^{99}Tc activity in the lobsters (Smith et al., 1998, Copplestone et al., 2004). The hepatopancreas plays an important role in the metabolism and act as a storage for minerals and nutrients preduring the moult (Barker and Gibson, 1977). The highest activity concentrations of ^{99}Tc were found in organs involved in digestion of food, namely the green gland and the remainder fraction, confirming the results of a study by Busby et al., (1997). In fact, the variations in the ^{99}Tc concentrations between gender, were less smaller in the remainder fraction than in muscle tissue.

The activity concentrations in the white meat fraction (claws and tail muscle) ranged from 7.5 Bq/kg to 85 Bq/kg (wet weight) and 2.0 Bq/kg to 24 Bq/kg (wet weight) in female and male lobsters, respectively. The ^{99}Tc concentrations in the white meat fraction tended to be higher in females than in males, in good in agreement with other studies (Smith, 1998, Swift, 2001). Furthermore, a positive correlation between ^{99}Tc in crusher claw muscle and cutter claw muscle was found for females (Figure 14, Linear regression: R^2=0.924, n=11, F=121, p=0.000) but not for males (Linear regression: R^2=0.001, n=16, F=0.021, p=0.887).

Male and female lobsters differed in ^{99}Tc activity (Bq/kg wet weight) in tail muscle (t-test: t=-8.949, n=103, p=0.000), cutter claw (Mann-Whitney U-test: Z=-4.097, n=29, p=0.000) and crusher claw (Mann-Whitney U-test: Z=-3.034, n=29, p=0.002).

No differences were found between male and female lobsters in ^{99}Tc activity in green gland (Mann-Whitney U-test: Z=-1.216, n=13, p=0.224), gills (Mann-Whitney U-test: Z=-1.571, n=13, p=0.116) and remainder fraction inclusive hepatopancreas (Mann-Whitney U-test: Z=-0.857, n=13, p=0.391). This is shown in Figure 15.

Table 7. Mean activity concentration (Bq/kg wet weight) in different organs.

Organ	Sex (M/F)	n	Mean ^{99}Tc (Bq/kg w. w.)	Standard deviation (SD)	Max value	Min value
Tail muscle	M	60	5.1	3.3	15.9	2.0
	F	43	21.4	13.6	70	5.1
	All	103	11.9	12.2	70	2.0
Cutter claw muscle	M	17	4.8	1.8	9.1	2.0
	F	12	16.1	11.5	45	5.8
	All	29	9.5	9.2	45	2.0
Crusher claw muscle	M	17	10.3	5.1	24.0	5.2
	F	12	24.8	21.2	85	8.2
	All	29	16.3	15.6	85	5.2
Gills	M	6	16.9	9.5	33.0	9.0
	F	7	31.0	24.4	80	11.0
	All	13	25.0	20.0	80	9.0
Green gland	M	6	180	100	310	70
	F	7	305	190	600	85
	All	13	245	165	600	70
Spawn	F	4	11.7	4.1	17.0	8.0
Remainder fraction inclusive hepatopancreas	M	6	22.7	3.0	28.0	19.5
	F	7	29.6	13.1	55	17.0
	All	13	26.4	10.1	55	17.0

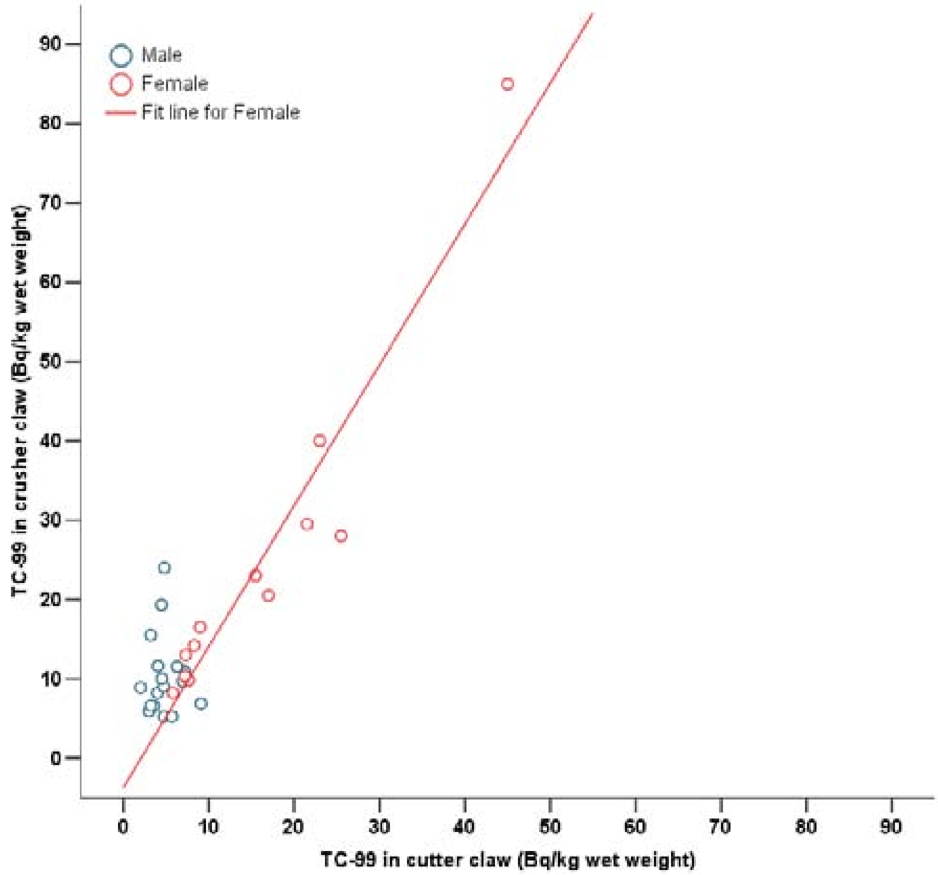


Figure 14. Activity concentrations of ^{99}Tc (Bq/kg wet weight) in cutter claw and crusher claw in male and female lobsters. Regression line is added to indicate a relation.

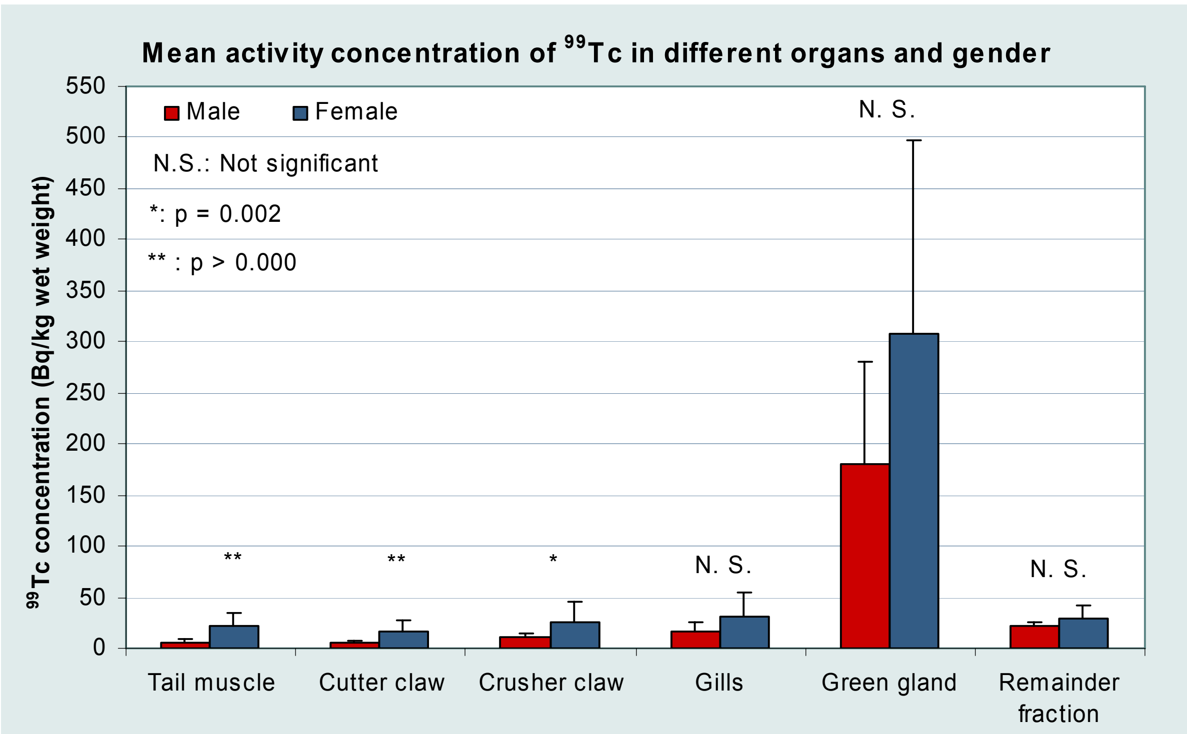


Figure 15. Activity concentrations of ^{99}Tc (Bq/kg wet weight) in different organs ($\pm$ standard deviation).

3.7 Concentration factors

To express the uptake of ^{99}Tc by lobsters from seawater, the concentration factor (CF) is used. The CF is the ratio of ^{99}Tc concentration in lobster tissue to that in the ambient seawater concentration in unit l/kg (IAEA, 2004).

The CF is based on fresh weight values. The concentration factor method assumes that the organism is in biochemical equilibrium with its surroundings. The time required for equilibrium to be attained depends on the half-life of the radionuclide and the biological half-life of the element in the organism. Even though, the ^{99}Tc will be more mixed and homogenous along the Norwegian coast compared to the Irish Sea, the results in this study are not necessarily equilibrium values and CFs calculated in this study reflects the concentration ratio at a given point in time.

$$CF = \frac{^{99}Tc\ concentration\ lobster, Bq/kg\ wet\ weight}{^{99}Tc\ concentration, seawater, Bq/l}$$

In lobster, the tail muscle is the fraction usually used for human consumption and this muscle portion is used to calculate the reported whole body CF. In most cases it's important to use the portion of the organism which is consumed by man.

Whole body CFs were determined for different locations (Figure 16). Univariate analyse showed statistically significant variations between males and females and also between locations (Table 8). Earlier reported data from the project, has shown a significant positive correlation between total weight and whole body CF for female lobsters but not for males (Rudjord et al., 2004). CFs were also determined for the different fractions dissected (Table 9). The mean reported whole body CF values for ^{99}Tc in European lobster, for males and females were 4100 l/kg and 16500 l/kg, respectively. The CFs are higher for females than for males, confirming the findings from the Irish Sea of higher accumulation in females compared to males (Swift and Nicholson, 2001).

However, large variation in CF values has been found, and the mean whole body CF ranged from 1400 l/kg to 13500 l/kg and 3700 l/kg to 55000 l/kg for males and females, respectively. Large range can be seen due to large variations which is probably related to the moulting and metabolism of organisms, or the ^{99}Tc variations in the food intake and consequently the uptake of ^{99}Tc (Rudjord et al., 2004). A lobster may eat up to 100 different kinds of animals, and occasionally eats some seaweed as well. Some brown algae like the *Ascophyllum, Fucus* have very high concentration factors for ^{99}Tc.

In this study, the CF values in lobsters collected at Sandøya in 2003 to 2005 ranged from 1400-6200 l/kg and 3700–55000 l/kg in males and females, respectively (Figure 17). The CF value in the remainder fraction was 17000 l/kg for males and 21000 l/kg for females. The green gland had the highest CF of 130000 l/kg and 220000 l/kg for male and female, respectively.

IAEA has recommended a CF of 1000 l/kg for technetium in crustaceans (IAEA, 2004) with the exception of lobster, where a higher value is recommended. The reported CFs in this study seems to be higher than earlier reported CF values from field studies (Table 10).

For 13 lobsters, six males and seven females collected at Sandøya, the various organs ^{99}Tc activities were also calculated as a percentage of each lobster's total whole body activity value (Figure 18). The remainder fraction contained the greatest proportion of ^{99}Tc activity in lobsters. However, in females the proportion of ^{99}Tc activity in the remainder fraction is lower and that in tail muscle higher, in good agreement with Smith et al., (1998).

Table 8. *General linear model (GLM), Univariate analyse with CF in lobster as dependent variable against location and sex.*

	SS	df	MS	F	p-value
Total	18365668990	101			
Location	1914631716	5	382926343	8.481	0.000
Sex	2881467523	1	2881467523	63.817	0.000
Error	4063681559	90	45152017		

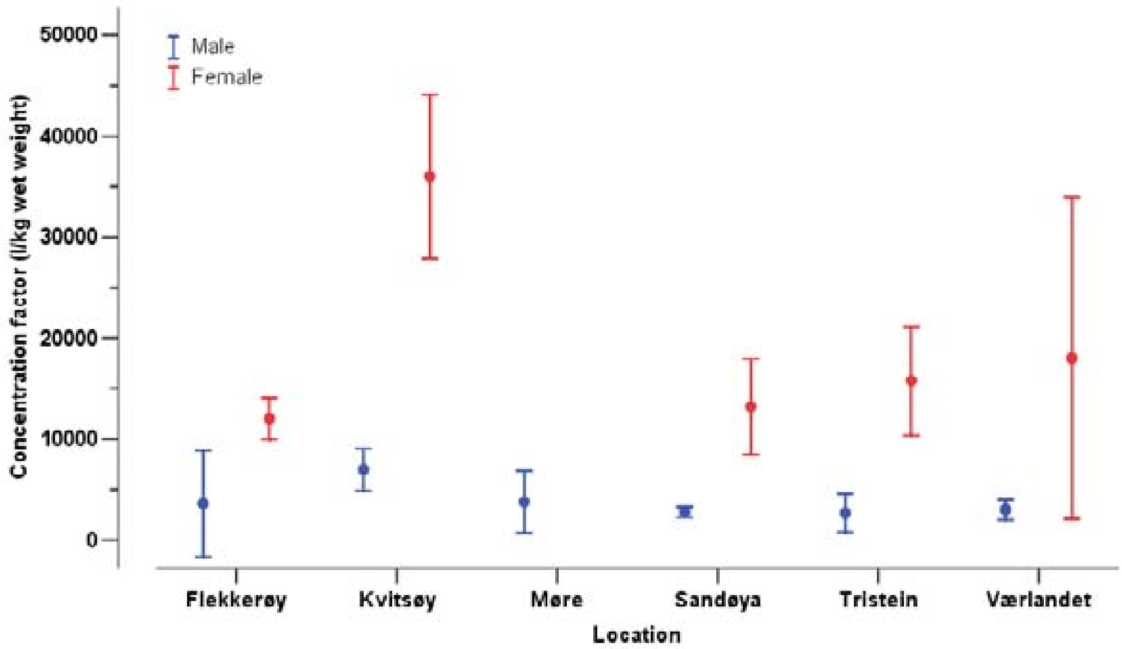


Figure 16. *Concentration factors (1/kg wet weight, (± standard deviation) for male and female lobsters at different locations.*

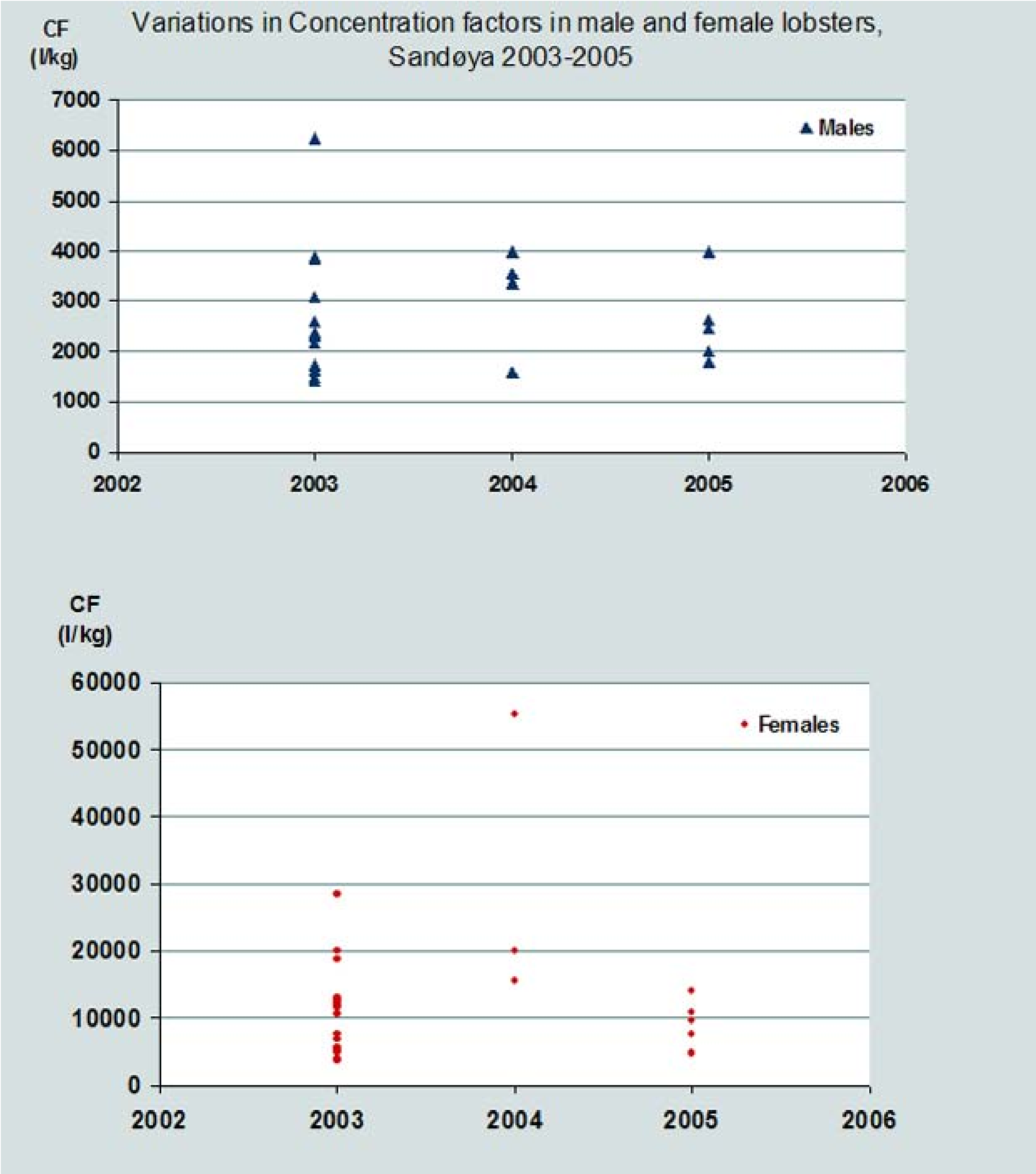


Figure 17. *CFs (l/kg) for male and female lobsters from Sandøya in the period 2003-2005.*

Table 9. *CF values (l/kg wet weight) in different organs dissected the mean ± standard deviation (SD) of the mean and max/min values.*

Organ	Sex (M/F)	n	Mean CF (L/kg) w. w.)	Standard deviation (SD)	Max value	Min value
Tail muscle	M	60	4100	3100	13500	1400
	F	43	16500	11500	55000	3700
	All	103	9300	9800	55000	1400
Cutter claw muscle	M	17	3500	1400	6600	1300
	F	12	11400	9100	36000	3700
	All	29	6800	7000	36000	1300
Crusher claw muscle	M	17	7700	4100	19000	3400
	F	12	17700	17100	68000	5300
	All	29	12000	12200	68000	3400
Gills	M	6	12500	7300	26000	7200
	F	7	22500	19500	65000	7000
	All	13	18000	15600	65000	7000
Green gland	M	6	130000	75000	250000	56000
	F	7	220000	150000	480000	55000
	All	13	180000	125000	480000	55000
Spawn	F	4	9700	3400	13000	5200
Remainder fraction	M	6	17000	3000	22500	14000
inclusive hepatopancreas	F	7	21000	11000	44000	11000
	All	13	19000	8400	44000	11000

Table 10. CF values for European lobster (*Homarus gammarus*), field studies.

Organ	**Location**	**CF (l/kg wet weight)**	**Reference**
Whole body		1000	Pentreath 1980
White meat	Irish Sea	380 - 1200	Busby et al., 1997
Tail muscle	Irish Sea	6850	Smith et al., 2001
Green gland	Irish Sea	25000-65000	Busby et al., 1997
Hepatopancreas	Irish Sea	7700-16000	Busby et al., 1997

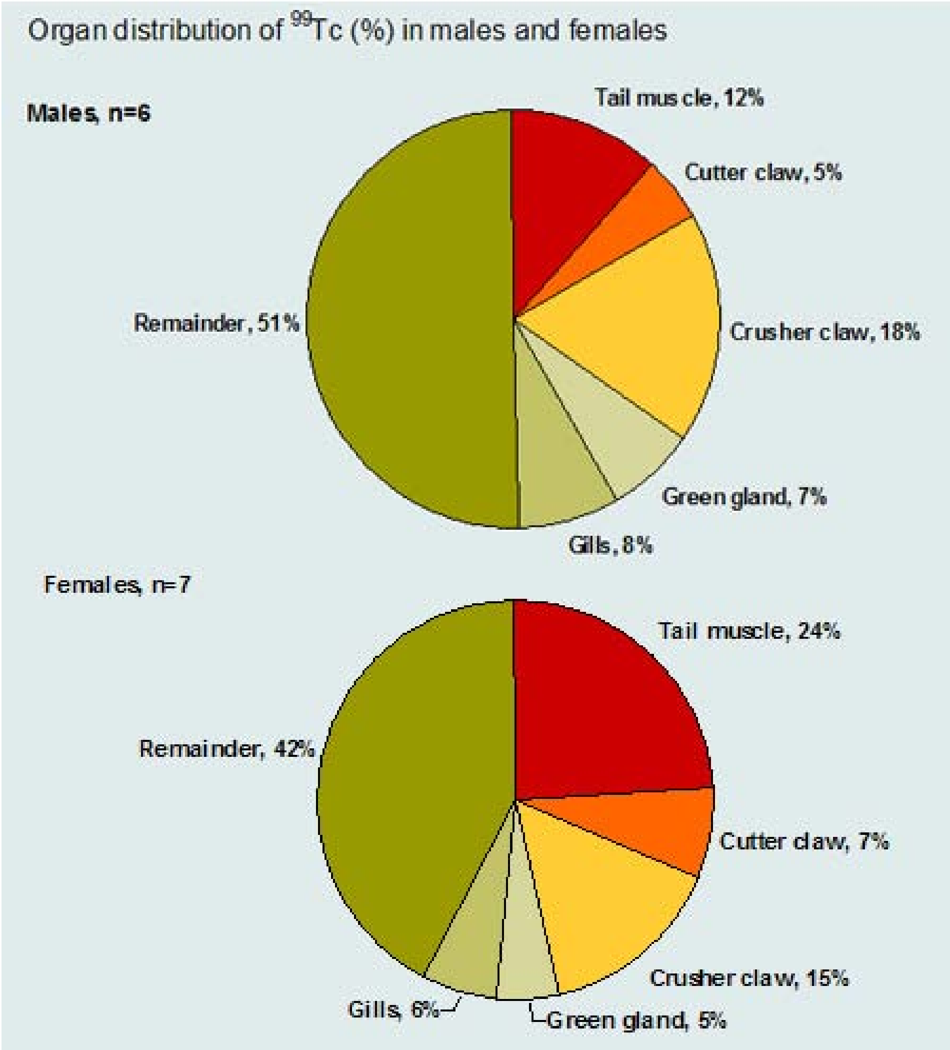


Figure 18. *Organ distribution of ^{99}Tc (%) in male and female lobsters, mean value of 6 and 7 individuals, respectively.*

4 Conclusions

The high accumulation of ^{99}Tc in lobsters compared to other crustaceans is confirmed in this report, as well as the clear differences between males and females. In all lobsters, the mean activity concentration of ^{99}Tc in tail muscle was 5.1 Bq/kg (wet weight) and 22.5 Bq/kg (wet weight) in males and females, respectively. The activity concentration of ^{99}Tc ranged from 2.0 Bq/kg (wet weight) to 16 Bq/kg (wet weight) and 5.1 Bq/kg (wet weight) to 70 Bq/kg (wet weight), in male and female lobsters, respectively. The ^{99}Tc uptake by female lobsters was significantly higher than in males ($p=0.000$, $n=103$) by a factor of ~ 4, confirming earlier observations by Swift and Nicholson (2001) from the Irish Sea. In many cases there was a large variability in ^{99}Tc activity concentrations in the tail muscle of lobster caught at the same location on the same day and of the same gender. This is consistent with the pattern Swift and Nicholson (2001) reported from the Irish Sea. The reason for that is not clear. However, the most probable explanation for the large variations is related to age and moulting/reproduction cycle of the lobsters. Recently moulted specimens feed actively compared to specimens that are about to moult. Moult cycle effects on technetium levels have been reported earlier by Swift (1985).

The reported data also suggests a sex difference for the organ distribution of ^{99}Tc. The lobsters displayed considerable variation in the ^{99}Tc distribution among subsamples of tail muscle tissue, cutter claw, crusher claw, gills, green gland and a remainder fraction (including hepatopancreas). The lowest ^{99}Tc activity concentration was found in white meat (tail and claws) in good agreement with Busby et al., (1997). The activity concentrations in the white meat ranged from 7.5 Bq/kg (wet weight) to 85 Bq/kg (wet weight) and 2.0 Bq/kg (wet weight) to 24 Bq/kg (wet weight) in female and male lobster, respectively. Like previous research has shown, the remainder fraction contained the greatest proportion of ^{99}Tc activity in the lobsters (Smith et al., 1998, and Copplestone, 2004). Furthermore, a statistically significant difference between male and female lobsters in ^{99}Tc activity (Bq/kg wet weight) in tail muscle tissue, crusher claw and cutter claw was found (Figure 15).

In the period 2003-2005, lobsters have been sampled annually at Sandøya and a significant difference has been found in the activity concentrations of ^{99}Tc and sampling years ($p=<0.007$) but the load of activity show no general decline over the period.

The mean reported whole body CF values for ^{99}Tc in European lobster, for males and females were 4100 l/kg and 16500 l/kg, respectively. However, in the remainder fraction the difference between the genders is less marked with CF of 17000 l/kg and 21000 l/kg for males and females, respectively. In the future, individual CF for male and female lobsters should be reported separately. The CFs in this study seems to be higher than earlier reported CF values. The green gland had the highest CF with 130000 l/kg for males and 220000 l/kg for females.

Organ ^{99}Tc activities were also calculated as a percentage of each lobster's whole body activity value, (Figure 18). The proportion of ^{99}Tc activity in the remainder fraction is lower and that in tail muscle greater in females, in good agreement with Smith et al., (1998).

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