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## Natural Radioactivity in Nordic Fish and Shellfish – Summary report 2018

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## Abstract

Studies in several countries have identified fish and shellfish as the food group causing the largest contribution to the ingestion dose, due to relatively high levels of naturally occurring radionuclides. However, levels have been shown to vary drastically between species. The objective of the NANOD project is to fill knowledge gaps related to levels of naturally occurring radionuclides in the fish and shellfish species commonly consumed in the Nordic region, in order to enable more accurate dose assessments for seafood and the total diet in the Nordic countries.

Species-specific consumption data was collected from each of the Nordic countries. The mean total fish and shellfish consumption for adults varied from 37 to 57 g/d. There was also substantial variation in species composition among the countries.

Samples of commonly consumed fish and shellfish species from each of the Nordic countries were collected for analysis of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$ , as these radionuclides previously have been shown to be the main contributors to the ingestion dose. The results from analyses completed so far show  $^{210}\text{Po}$  concentrations in wild fish ranging from 0.079 to 1.9 Bq/kg, and from 0.94 to 77 Bq/kg in shellfish. Overall, preliminary results indicate higher  $^{210}\text{Po}$  levels in species with a  $^{210}\text{Po}$ -rich diet. This is in line with previous studies showing that  $^{210}\text{Po}$  enters the organisms primarily via ingestion. Moreover, farmed trout showed the lowest  $^{210}\text{Po}$  levels, likely due to a diet of plant-based feed with low  $^{210}\text{Po}$  content. Concentrations also varied significantly between samples of the same species. This may partly be related to geographic differences, although the direct cause of this is not apparent. Results of this and other work indicate that influences on  $^{210}\text{Po}$  concentrations in fish and shellfish are complex, and several factors may play a role.

Results of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  analyses performed so far are mainly below the detection limits, but are sufficiently low to provide valuable information. Preliminary findings indicate highest level in blue mussels, with 1.1 and 1.5 Bq/kg, respectively.  $^{210}\text{Pb}$  and further results of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$  are expected in 2019. A better picture of variation and possible patterns among fish and shellfish may be possible remaining analyses are complete. The complete data set, further discussions and dose estimates will be presented in the 2019 Final Report, provided continued funding of the NANOD project.

Data on naturally occurring radionuclides in fish and shellfish is important not only due to the food group's role in the Nordic diet, but also because the Nordic countries are important in the worldwide fish trade.

## **Key words**

Gamma spectrometry, radioactive sources, activity determination, shielded sources, nuclear security

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Final Report from the NKS-B NANOD activity 2018 (Contract: AFT/B(18)8)

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## 1. Introduction

### 1.1. Background

The vast majority of the ingestion dose received by the general population is caused by naturally occurring radionuclides (UNSCEAR 2000; O'Connor et al. 2014; Komperød et al. 2015). Nonetheless, natural radioactivity in food receives far less attention than anthropogenic radionuclides. The reasons for this is likely their natural origins, and that there is no legislation regulating their concentration in food products. In addition, some of the most important natural radionuclides are relatively costly and time-consuming to analyse.

In several countries, previous studies have found seafood to be the single food group that causes the largest contribution to the mean ingestion dose, due to the relatively high content of natural radionuclides in fish and shellfish (Komperød & Skuterud 2018; Ota et al. 2009; Renaud et al. 2015; Watson et al. 2005). Concentrations of natural radioactivity has also been shown to vary dramatically between species (Carvalho et al. 2011; Díaz-Francés et al. 2013; HELCOM 2018; Yamamoto et al. 1994). In order to make relevant dose assessments for fish and shellfish, it is therefore important to use measurement data for the species actually consumed in the respective country or region.

### 1.2. Scope and objectives

Due to the importance of seafood to the dose from the diet and its relatively high consumption in the Nordic countries, the NANOD project focuses on the knowledge gap associated with naturally occurring radionuclides in the fish and shellfish species commonly consumed in this region. The aim of the project is to enable more accurate dose assessments for seafood and the total diet in the Nordic countries.

Based on the radionuclides identified as most important to ingestion doses in earlier work, the naturally occurring radionuclides analysed in this study is limited to  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ <sup>1</sup>, with  $^{210}\text{Po}$  expected to be the single largest contributor (Renaud et al. 2015; Komperød Skuterud 2018; Ota et al. 2009).  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ , and  $^{226}\text{Ra}$  are all products in the  $^{238}\text{U}$  decay chain, while  $^{228}\text{Ra}$  is a progeny of  $^{232}\text{Th}$ . These radionuclides are ubiquitous in the environment and in our food in varying concentrations.

This report covers the work completed in 2018 in the NANOD project, summarising available information on Nordic consumption and previously collected data on natural radioactivity in fish and shellfish, overview of the samples collected for this project, as well as results from completed measurements. Some of the measurement procedures include a waiting period of up to six months ( $^{210}\text{Pb}$ ). When also considering the fact that some of the different species are typically harvested at particular times of the year (e.g. mackerel and shrimp in the summer), it was difficult to complete all of the analyses and assessments within the same year. The project has therefore been planned to continue in 2019. The final report covering the complete

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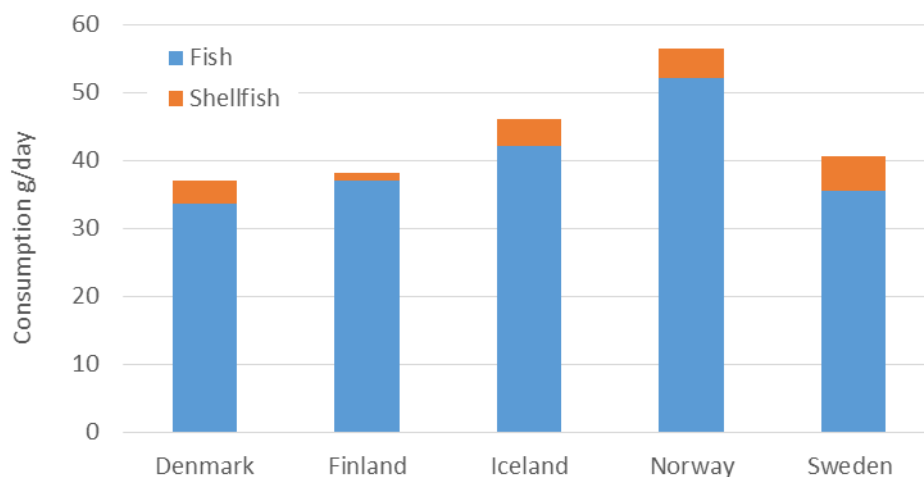
<sup>1</sup>  $^{40}\text{K}$  is one of the largest contributor to dose from the diet, equal to or exceeding that of  $^{210}\text{Po}$ , but since the amount of K (incl.  $^{40}\text{K}$ ) is strictly regulated by the body, the dose is constant regardless of dietary intake. This means that the  $^{40}\text{K}$  content in food is not relevant for dose estimates.

NANOD project on fish and shellfish, including all results and dose assessments, is therefore expected in 2019, provided continued funding of the project.

## 2. Nordic consumption

### 2.1. Total fish and shellfish consumption

Although fish and shellfish are an important part of the diet all Nordic countries, the consumption level varies considerably at the national level. The most recent consumption data available for adults in each country is presented in Figure 2.1-1. It should be noted that differences between national data might also arise due to different methods of registering dietary data and time since the most recent survey, as statistics show there have been both increases and decreases in national fish consumption during the last decade.



**Figure 2.1-1.** Mean consumption (g/day) of fish and shellfish (edible parts) for adults in each Nordic country<sup>2</sup>.

According to the dietary data obtained, Norwegian mean fish consumption is the highest among the Nordic countries, with an average of 52 g/d in the adult population, while Iceland has the second highest, with an estimated 42 g/d. Denmark, Finland, and Sweden have similar intake of fish around 35 g/d. The consumption of shellfish (molluscs and crustaceans) in Iceland, Norway, and Sweden varies around 4-5 g/d on average, and is lowest in Finland with an estimated 1.1 g/d<sup>2</sup>.

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<sup>2</sup>References for consumption data:

Denmark: The Danish National Survey of Diet and Physical Activity 2011-2013. Data from Pedersen et al. (2015) and Jeppe Matthiessen at DTU Food (personal communication regarding shellfish consumption).

Finland: Natural Resources Institute Finland, Statistics service: Fish consumption 2017 (Natural Resources Institute Finland 2018)

Iceland: Estimated based on the dietary survey "Hvað borða Íslendingar?" 2010-11 (Þorgeirsdóttir et al. 2013).

Norway: Data collected through the National dietary survey for adults 2010-2011, Norkost 3 (Totland et al. 2012). Estimates of consumption from the same source in terms of pure fish provided in VKM (2014).

Sweden: Swedish National Dietary Survey - Riksmaten 2010-11, via EFSA database (EFSA 2018).

Certain groups consume far more than the average population. Consequently, some parts of the population will receive ingestion doses from seafood that are several times higher (or lower) than the mean population data presented in Figure 2.1-1. For example, the 95<sup>th</sup> percentile for combined fish and shellfish consumption in Norway is 201 g/d (VKM 2014).

## 2.2. *Estimated consumption by species*

It has proved somewhat difficult to obtain reliable species-specific consumption data for fish and shellfish, and considerable work was put into making the best possible estimates from different sources of information and new inquiries. An overview of the resulting estimated consumption by species in each of the Nordic countries is provided in Table 2.2-1. A list of all Latin species names relevant for this report is given in Appendix C.

While some non-Nordic seafood species such as *Pangasius* and Alaska pollock have become increasingly significant in the Nordic diet, especially in combination products, species native to the Nordic region still make up the major share of Nordic consumption according to the collected data. Overall, cod, salmon, rainbow trout, herring, mackerel, plaice and haddock are the main consumed species, although there are clear differences between countries in their relative consumption. Canned tuna is also a significant food item, especially in Finland and Denmark. Shrimp is the main type of shellfish consumed, mainly the deep-water species northern prawn, which is caught in large volumes both by Denmark, Iceland, and Norway.

Due to its brackish environment, different species tend to inhabit the Baltic Sea, especially inner areas like the Gulf of Bothnia, than the open seas. This naturally affects the choices of species consumed and partly explains the significant differences in preferred species, for example between Finland and Iceland. Likewise, there is also a significant difference between the share of saltwater vs. freshwater species consumed, with the Finnish population consuming larger amounts of freshwater fish. It should be noted that some of these species also inhabit the brackish environments in the inner parts of the Baltic Sea, sometimes making it difficult to draw a clear distinction between freshwater organisms and “seafood” based on information on species consumption. In comparison, in Norway for example, freshwater fish is estimated at only approx. 5% or less of total fish and shellfish consumption (Komperød et al. 2015).

Import makes up an important part of some Nordic fish markets; however, fish imported from other Nordic countries appear to make up the major fraction of that import. For example, in Sweden, almost 75% of fish and shellfish consumed is imported; however, Norway is the main country of origin for the 10 most consumed species of that import, followed by Denmark (Ziegler & Bergman 2017). Similarly, in Finland import accounts for approximately 80% of fish sold, of which around 50% is imported from Norway. Therefore, it appears that the fish and shellfish consumed within the Nordic countries mainly originate from within the Nordic region.

Data from Norway, Finland and Sweden show that the salmon and rainbow trout consumed is usually farmed. Farming could have significant impact on the concentration of certain radionuclides in the fish muscle, due to different diet than its wild relatives.



**Table 2.2-1.** Consumption of various species in the different Nordic countries. See footnotes for references.

Species	Mean consumption (g/d)				
	Denmark <sup>3</sup>	Finland <sup>4</sup>	Iceland <sup>5</sup>	Norway <sup>6</sup>	Sweden <sup>7</sup>
Alaska pollock <sup>a</sup>	1.7			3.4	0.5
Atlantic salmon <sup>b*</sup>	3.6	11	3	12.5	4.9
Arctic char <sup>c</sup>			3		
Atlantic cod <sup>b</sup>	2.05		11	14	8.6
Atlantic mackerel <sup>b</sup>	5.5			4	5.6
Tuna, canned <sup>b</sup>	6.2	4.1	1	2	1
Cod roe <sup>b</sup>				1	1.2
Crab <sup>b</sup>	0.07			0.4	
European perch <sup>d</sup>		1.1			
European plaice <sup>b</sup>	3.5		2	0.5	4.1
European whitefish <sup>d</sup>		0.8			
Greater argentine <sup>b</sup>				3.4	
Haddock <sup>b</sup>			15	3.9	0.78
Halibut <sup>b</sup>			2	1	
Herring <sup>b</sup>	7.4	2.2	2	1	4
Mussels <sup>b</sup>			0.1	0.3	
Northern pike <sup>d</sup>		1.1			
Norway lobster <sup>b</sup>			1		
Pangasius <sup>a*</sup>					0.3
Pike-perch <sup>d</sup>		1.1			4.3
Rainbow trout <sup>c*</sup>		5.5		1	
Redfish <sup>b</sup>			1	1	
Saithe <sup>b</sup>		1.1	1	3	
Scallops <sup>b</sup>			1	0.1	
Shrimp <sup>b</sup>	3.3	1.1	2	3	4
Vendace <sup>a</sup>		1.6			
Wolffish <sup>b</sup>			1	0.5	

<sup>a</sup> Mainly freshwater origin

<sup>b</sup> Mainly seawater origin (some species extend into brackish sea)

<sup>c</sup> Both freshwater and seawater/brackish water origins are common (e.g. in Norway, rainbow trout is farmed in sea; in Finland, it is farmed both in freshwater and in brackish water)

<sup>d</sup> Freshwater/brackish water origin

\*Mainly farmed

<sup>3</sup> Based on total consumption from The Danish National Survey of Diet and Physical Activity 2011-2013. Data from Pedersen et al. (2015) and Jeppe Matthiessen at DTU Food (personal communication regarding shellfish consumption). Relative species-specific consumption estimated based on SEAFOODplus (2016) for fish and data from the Danish dietary survey 2005-2008 available in the EFSA database (EFSA 2018).

<sup>4</sup> Natural Resources Institute of Finland, Statistics service. Fish consumption 2017. (Natural Resources Institute 2018)

<sup>5</sup> Estimated based on the dietary survey “Hvað borða Íslendingar?” 2010-11 (Þorgeirsdóttir et al. 2013) as well as information from fish mongers.

<sup>6</sup> Data collected through the National dietary survey Norkost 3 2010-2011 (Totland et al. 2012), with species-specific estimates based on VKM (2014). Based on NRK (2018), species composition in fish products was adjusted from VKM assumption of 100% cod to 25% each of cod, haddock, Alaska pollock and greater argentine.

<sup>7</sup> Total consumption from Riksmaten 2010-2011 (Amcoff et al. 2012). Share of most consumed species estimated from Swedish Market Basket 2015, Annex 1 (Darnerud et al. 2017).

### 3. Existing data

#### 3.1. *Previous studies on natural radioactivity in seafood in Nordic areas*

Literature searches were performed for previous studies of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in seafood in the Nordic region. Each country also looked for relevant published or unpublished data at their institution. A limited number of reports and scientific publications were obtained, as well as some unpublished data. The largest data source in terms of analyses performed was the HELCOM database, which contains data on various sample types from the Baltic Sea (HELCOM 2018).

The overall observation is that data for  $^{210}\text{Po}$  was available for several species, although with few analyses in most cases. There was quite a bit of data for  $^{210}\text{Po}$  in cod and herring, although the results were highly variable. Relatively little data was available for  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ , and especially  $^{228}\text{Ra}$ . In addition, results for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were often below the detection limit. Radium levels in muscle and edible parts are usually very low, but due to a higher dose per Bq emitted, this does not necessarily mean that doses are insignificant – depending on the detection limit value. For these reasons, efforts were made to try to get detection limits as low as possible for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in this project.

For several of the commonly consumed species, no data whatsoever on naturally occurring radionuclides was found from the Nordic countries. A recent survey on farmed salmon from Norway, the main producer in the region, was recently performed, and this data was found to be sufficient to cover this important product (Heldal et al. 2017).

More detailed information on the relevant previous work is provided in Appendix A.

**Table 3.1-1** Summary of radionuclide concentrations (Bq/kg fw) in previous studies on commonly consumed species in the Nordic region. In this table, *n* reflects the number of samples analysed, regardless of the number of individuals included in each sample. See Appendix A for more details and references. Just over half of these studies are from the Baltic Sea. Other species commonly consumed, but not of Nordic origin, include tuna, *Pangasius*, and Alaska pollock.

Species	<sup>210</sup> Po			<sup>210</sup> Pb			<sup>226</sup> Ra			<sup>228</sup> Ra		
	<i>n</i>	Mean	Min-max	<i>n</i>	Mean	Min-max	<i>n</i>	Mean	Min-max	<i>n</i>	Mean	Min-max
Arctic char	0			0			0			0		
Atlantic cod	82	1.34	0.043 - 4	6	0.064		79	0.19	0.042 - 4.9	0		
Atlantic mackerel	23	1.9		1	0.08		0			0		
Atlantic salmon, farmed	7		0.003 - 0.23	7		0.03 - 0.07	100		<0.04 - <0.18	100		<0.006 - <0.39
Baltic clam	0			0			3	2.5	0.64 - 3.98	0		
Blue mussels	N/A	2	7.5 - 37	N/A	2.1		11	1.4	0.029 - 12	3	1.5	0.35-3.4
Capelin	1	5.3		0			0			0		
Cockle	0			0			1	0.71		0		
Cod roe	0			0			0			0		
Crab	0			0			0			0		
Flounder	15	6.7	2.7-16	0			71	0.046	0.026 - 0.075	0		
Haddock	4	1.4		0			1	0.188		0		
Halibut	0			0			0			0		
Herring	55	2.86	0.19 - 23	6	0.19	0.076 - 0.45	1	0.028	0.02 - 0.055	0		
Norway lobster	0			0			0			0		
Perch	16	0.19	0.038 - 0.37	14	0.05	0.010 - <0.15	3		<0.95-<3.2	3	0.54	<0.54-<1.3
Pike	3	1.9	0.94 - 3.8	1	0.092		0			0		
Pike-perch	0			0			0			0		
Plaice	47	4,7	0.26 - 12	4	0.1	0.055 - 0.15	0			0		
Rainbow trout	N/A		0.039 - <0.26	N/A		0.013 - <0.26	1	<0.73		1	<0.25	
Redfish	1	0.16		0			0			0		
Saithe	2	0.92		0			0			0		
Scallop	0			0			0			0		
Shrimp	0			0			0			0		
Sprat	0			0			75	0.073	0.05 - 0.11	0		
Vendace	3	1.3	0.79 - 1.6	3		<0.38 - <0.47	3		<2.2 - <4	3		<0.64 - <1.2
Whitefish	N/A	3.2	<0.23 - 13	N/A	0.02	0.018 - <0.25	4		<0.43 - <0.96	4		<0.16 - <0.37
Wolffish	0			0			0			0		

### 3.2. *Previous studies on the effects of cooking*

It is well documented that some radionuclides may be lost during the process of food preparation. For example, a significant portion of the anthropogenic radionuclide  $^{137}\text{Cs}$  is removed through dilution when food is boiled (IAEA 2010). However, all such effects will vary according to the physical and chemical properties of the element.

$^{210}\text{Po}$  volatilizes at high temperatures and it has been hypothesized that some  $^{210}\text{Po}$  also may be lost due to volatilization when food is grilled or baked at high temperatures. Due to the importance of  $^{210}\text{Po}$  to ingestion doses from seafood, the available scientific literature was examined to see whether the effect of cooking on  $^{210}\text{Po}$  levels should be taken into account. A very limited number of studies was found, and are summarised in Table 3.2-1. All represent muscle/edible parts of the fish and shellfish. The organisms have received slightly different treatments, but the main cooking methods and net gain or loss in  $^{210}\text{Po}$  concentration are summarised.

The data varies dramatically and suggest that both increases and decreases in  $^{210}\text{Po}$  concentrations may occur during food preparation. Increases likely represent a loss of water from the tissue, thereby increasing the concentration of remaining substances, whereas decreases could represent either loss via fluids, loss due to volatilisation, or both.

Due to the ambiguous results of the existing data, it was concluded that no adjustment in  $^{210}\text{Po}$  concentrations due to cooking could be made in the dose assessments from fish and shellfish (to be presented in the 2019 Final Report). Due to limited resources and the apparent complexity of the subject, it was not possible to make such studies within the scope of this project since producing data on the main consumed species in the Nordic countries was the main priority. However, studies on the effects of cooking may be the scope of later work.

**Table 3.2-1.** Calculated changes in <sup>210</sup>Po concentrations in seafood based on levels observed in previous studies.

Species	Cooking method	Change in <sup>210</sup> Po concentration (%)
Mackerel ( <i>Scomber scombrus</i> ) <sup>a</sup>	Grilled in pan	+ 5.6
Salmon ( <i>Salmo salar</i> ) <sup>a</sup>	Grilled in pan	> - 80
Sardine ( <i>Sardine pichardus</i> ) <sup>a</sup>	Grilled in pan	- 31
Blue whiting ( <i>Micromesistius poutassou</i> ) <sup>a</sup>	Grilled in pan	- 426
Red mullet ( <i>Mullus barbatus</i> ) <sup>a</sup>	Grilled in pan	- 237
Sword fish ( <i>Xiphias gladius</i> ) <sup>a</sup>	Baked in oven	- 60
Cod ( <i>Gadus morhua</i> ) <sup>a</sup>	Grilled in pan	+ 11
Anchovy ( <i>Engraulis encrasicolus</i> ) <sup>a</sup>	Grilled in pan	- 17
Sole ( <i>Solea solea</i> ) <sup>a</sup>	Grilled in pan	- 25
Clam ( <i>Camelea gallina</i> ) <sup>a</sup>	Steamed	- 62
Blue mussel ( <i>Mytilus edilus</i> ) <sup>a</sup>	Steamed	- 105
European anchovy ( <i>Engraulis encrasicolus</i> ) <sup>b</sup>	Boiled in water	+ 4.3
Mediterranean mussel ( <i>Mytilus galloprovincialis</i> ) <sup>b</sup>	Boiled in water	+ 49
Cross-cut carpet shell ( <i>Venerupis decussata</i> ) <sup>b</sup>	Boiled in water	+ 37
Mediterranean shore crab ( <i>Carcinus aestuarii</i> ) <sup>b</sup>	Boiled in water	- 90
Spot-tail mantis shrimp ( <i>Squilla mantis</i> ) <sup>b</sup>	Boiled in water	+ 1.1
Mediterranean mussel ( <i>Mytilus galloprovincialis</i> ) <sup>c</sup>	Boiled in hot oil	+ 3.1
Mediterranean mussel ( <i>Mytilus galloprovincialis</i> ) <sup>c, d</sup>	Boiled in hot oil	+ 2.3

a. Díaz-Francés et al. 2017

b. Roselli et al 2017

d. Kristan et al. 2015

c. Assumed 15% dry matter when converting dry weight to wet weight concentrations.

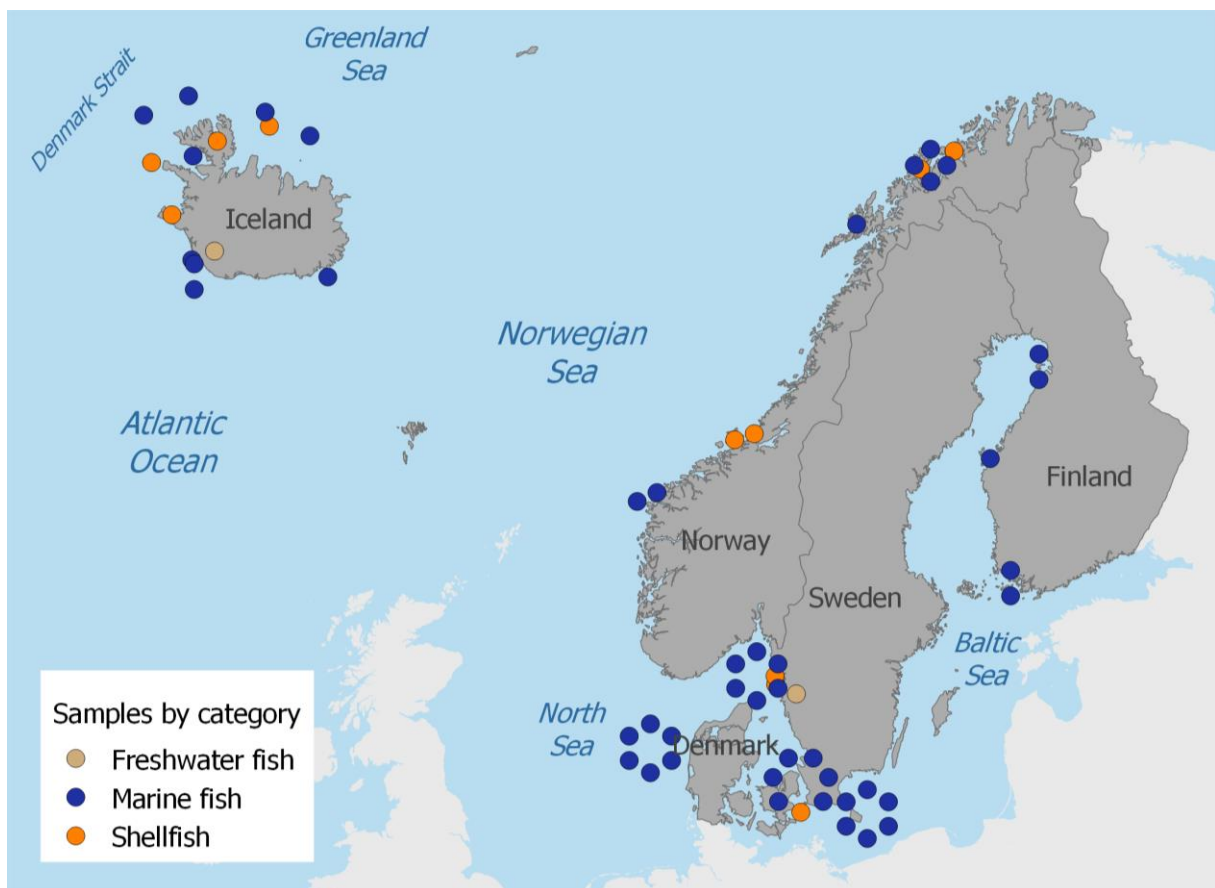
## 4. Methods

### 4.1. Identification of species prioritised for sampling

Since the main objective of this study is to collect and produce the data needed to make more appropriate dose estimates for seafood in the Nordic countries, the following factors were prioritised when selecting species to be sampled:

- Species with high consumption in the Nordic countries (Chapter 2.2)
- Species with no or insufficient existing data from prior studies, or exhibiting highly variable levels
- Species with suspected high levels of  $^{210}\text{Po}$ , typically shellfish and plankton-eating fish (see chapter 6)

Some species were also sampled at several locations, in order to examine whether there were substantial regional differences in natural radioactivity levels. Practical considerations were also sometimes limiting in determining what species were possible to sample, such as seasonal changes in availability. An overview of all samples collected in this study is shown in Figure 4.1-1.



**Figure 4.1-1.** Map showing location of the samples collected in this study. Locations in the North Sea, Skagerrak/Kattegat and the Bornholm area of the Baltic Sea are approximate. Rings of points represent several samples from the same location reference (in these cases, exact location is unknown).

## 4.2. *Sampling and analyses*

The different countries chose different ways of sampling species to be analysed in the project. A summary of the samples collected is available in Table 5-1, while more details regarding each sample is provided in Appendix B.

According to the project plan, all samples are to be analysed for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , and half of them for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . Due to an ingrowth time of several months, the plan was for all of the  $^{210}\text{Pb}$  analyses to be performed in 2019. The methods of sampling and analyses for each country is described below.

### *Denmark*

Samples of fish and other marine biota were sampled through personal contacts with professional fishermen, but some of the sampling is also done by DTU employed staff. Edible parts of the various foodstuffs were prepared for further analysis by freeze-drying. A suitable fraction of the freeze-dried material was taken for  $^{210}\text{Pb}/^{210}\text{Po}$  analysis (10-30g). Following ashing at 450 °C, samples were measured by gamma spectrometry for 2-3 days. A fraction of the ash was taken for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  analysis, unless determined by gamma spectrometry. Following dissolution of the ash, half of the sample was used for  $^{226}\text{Ra}$  which is co-precipitated onto lead sulphate.  $^{226}\text{Ra}$  was determined by liquid scintillation counting of radon of the EDTA-dissolved precipitate.  $^{133}\text{Ba}$  was used as yield determinant.  $^{228}\text{Ra}$  was determined through  $^{228}\text{Th}$  ingrowth. The dissolved ash was evaporated, dissolved in 8M  $\text{HNO}_3$ , and passed through an anion exchanger to remove  $^{228}\text{Th}$ . The sample was spiked with a known amount of  $^{229}\text{Th}$ , allowed to stand for 3-6 months and thorium isolated from the sample by anion exchange chromatography, electroplated onto stainless steel discs and counted by solid state alpha spectrometry for about a week.

$^{210}\text{Po}$  is determined by alpha spectrometry of polonium spontaneously plated onto silver discs following sample dissolution and addition of  $^{209}\text{Po}$  and stable lead (10 mg) as yield determinants. Following plating the supernate is passed through a short anion exchange resin to adsorb residual Po, transferred to a bottle for storage during 3-6 months to allow for  $^{210}\text{Po}$  ingrowth. Following ingrowth, new  $^{209}\text{Po}$  is added and the plating on a silver disc repeated.

### *Finland*

Samples were collected as a part of the Surveillance Programme of Environmental Radiation and Monitoring Programme of Radioactive Substances in the Baltic Sea (HELCOM-MORS).

STUK has several HPGe gamma-ray spectrometers for determining gamma-ray emitting radionuclides in environmental samples. All spectrometers have digital multichannel analysers for data acquisition. The edible parts of fish samples were dried overnight at 105 °C and minced before the measurement. The samples were directly transferred to a standard plastic container. STUK uses three main measurement geometries of which two simple cylindrical were used for activity measurements of fish samples (diameters 42 mm and 74 mm, heights 0–26 mm and volumes 0–30 mL and 0–100 mL, respectively). All samples were measured on top of the detector end-cap. In the case of simple cylindrical samples, the efficiency

calibration is determined for the sample thickness of 0 mm. Analysis software (UniSampo-Shaman) corrects this for real sample thickness and density. The measuring time of fish samples varied between 6 to 14 hours. Some of the samples were vacuum-packaged to obtain secular equilibrium between radon and its daughters in order to reliably determine the  $^{226}\text{Ra}$  activity concentration.

$^{210}\text{Pb}/^{210}\text{Po}$  analyses will be completed in 2019, and these analytical methods will be described in next year's report.

### *Iceland*

Samples of cod, haddock, ling, saithe, and black halibut were collected during experimental trawling trips organised by the Marine and Freshwater Research Institute in February and March 2018. Each fish sample consists of a pool of at least ten individuals of a specific length distribution. Standardized sample preparation was in the hands of Matís (Icelandic Food and Biotech R&D). The skinless fish fillets from the individuals were pooled, homogenised and freeze-dried for further analysis.

Samples of Atlantic halibut, plaice, farmed arctic charr, and Atlantic herring were bought fresh by IRSA from a trusted source at a fish market. The edible part from about 5 kg (fw) of each species was pooled and dried at 40 °C in a slow-airflow drying cabinet, then ground in a food processor.

Three samples of northern prawn were obtained directly from two fisheries in West-Iceland that were able to provide 5-kg samples of freshly caught shrimp with full sample information. The sampling of blue mussels was carried out by specialists of the University of Iceland's Institute of Research Centre in Suðurnes, for IRSA near Reykjanes. All soft tissue of 100 individuals (length ~50mm, common for human consumption) was removed from the shells and divided into three 2-litre beaker glasses for drying at 50 °C for 4 days.

### *Norway*

Samples collected in Norway were purchased directly from producer or at fish markets that had knowledge of when and where the fish and shellfish were caught. A minimum of 10 individuals were obtained for each species.

Equal amounts of muscle/edible parts were removed from each organism in order to make a representative bulk sample for each species. Care was taken to remove any detectable pieces of bones and shell as not to affect the  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  analyses, since this material generally contains several times higher concentrations of radium than soft tissues. Samples were dried at 80 °C for a minimum of 48 hours and homogenised before further treatment.

Determination of  $^{210}\text{Po}$  was carried out according to a slight modified version of the method described by Chen et al. (2001).  $^{209}\text{Po}$  tracer was added to dried samples. After treating sample several times using *aqua regia*,  $\text{NaNO}_3$ ,  $\text{H}_2\text{O}_2$ ,  $\text{HCl}$ ,  $\text{H}_2\text{O}$  and  $\text{NH}_2\text{-HCl}$ , the sample was deposited onto silver discs before measurement with Canberra Alpha Analyst.  $^{210}\text{Po}$  results presented in this report are preliminary, pending final determination after  $^{210}\text{Pb}$  analysis. The



sample solution will be used to determine the  $^{210}\text{Pb}$  activity. Adding  $^{209}\text{Po}$  tracer once more, the sample will be stored for 6 months before a new spontaneous deposition and measured with Canberra Alpha Analyst.

Samples of sufficient size (rainbow trout, plaice, northern prawn, blue mussels, saithe, haddock and cod) were ashed at  $550\text{ }^{\circ}\text{C}$  in order to achieve detection limits as low as possible for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . The samples were prepared in hard plastic cylindrical beakers. To prevent radon leakage, the beakers were placed in aluminium-lined bags, and evacuated and sealed using a commercial vacuum packing machine. The samples were stored for a minimum of three weeks to ensure equilibrium and analysed by using HPGe detectors. The  $^{226}\text{Ra}$  activity was determined by using a weighted mean of the background-corrected signals from the 295 keV and 352 keV peaks of  $^{214}\text{Pb}$  and the 609 keV peak of  $^{214}\text{Bi}$ . The  $^{228}\text{Ra}$  activity was determined by a weighted mean of 338 keV, 911 keV and 969 keV peaks of  $^{228}\text{Ac}$ .

### *Sweden*

Samples collected in Sweden were purchased in fish shops where Swedes usually buy seafood. Approximately 2-4 kg of a representative amount of individuals for each species were collected during the sampling campaigns.

As standard pre-treatment, the samples were washed and non-edible parts (skin, bones, etc.) were removed. Then, each sample was ground and mixed before dried at  $80\text{ }^{\circ}\text{C}$  to constant weight. After the drying process, the samples were again milled, sieved and mixed to ensure the total homogenization of the sample before radiochemical determinations.

Determination of  $^{210}\text{Po}$  was carried out according to the radiochemical procedure described by Díaz-Francés (2016).  $^{209}\text{Po}$  was added as tracer to check the yield recovery. For the radiochemical determination of  $^{210}\text{Po}$ , 2-4 g of dried sample were acid digested by Microwave Digestion System (Milestone Ethos Easy) using 65% Nitric Acid and 35% oxygen peroxide as reagent. Then, polonium was separated by liquid-liquid solvent extraction method using Tributyl Phosphate (TBP) and  $\text{HNO}_3$  (8M). For the source preparation, Po was deposited on a copper disk in HCl (2M) at  $80\text{ }^{\circ}\text{C}$ , shaking continuously the sample during 5h. Finally,  $^{210}\text{Po}$  was measured by high-resolution alpha spectrometry in order to determine the activity concentration.

For the determination of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , dried samples were reduced to ashes at  $450\text{ }^{\circ}\text{C}$  in order to remove organic matter and improve the detection limits. Samples were milled, sieved and homogenized to prepare optimal gamma measurement beaker. Finally, the measurement containers were sealed, using a commercial vacuum packing machine, to avoid any loss of radon in order to determine  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  by secular equilibrium. The samples were stored for a minimum of three weeks to ensure secular equilibrium before the measurement by HPGe gamma spectrometry detectors. The  $^{226}\text{Ra}$  activity was determined by using a weighted mean of the background-corrected signals from the 352 keV peak of  $^{214}\text{Pb}$  and the 609 keV peak of  $^{214}\text{Bi}$ . The  $^{228}\text{Ra}$  activity was determined by a weighted mean of the 237 keV peak of  $^{212}\text{Pb}$  and 911 keV peak of  $^{228}\text{Ac}$ .

## 5. Results

The results from the analyses completed in 2018 are provided in Table 5-1. According to the project plan, all samples are to be analysed for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , while only half of the samples for radium analyses. Fewer analyses of  $^{210}\text{Po}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$  than expected have been completed within 2018, mainly due to unforeseen problems and partial shutdown at DTU laboratories, affecting the scheduling of analyses for Danish and Icelandic samples. As mentioned above, all  $^{210}\text{Pb}$  analyses were planned for analysis in 2019 due to a required ingrowth time of several months, and are therefore not presented in this report. All remaining analyses will be completed in 2019.

All results are presented in fresh weight (fw) concentrations. More details regarding each sample is provided in Appendix C.

**Table 5-1.** Summary of results (Bq/kg fresh weight) from the present study. More details on sampling location and time is provided in Appendix C.

Species	Country	Sample origin	$^{210}\text{Po}$	$^{226}\text{Ra}$	$^{228}\text{Ra}$
<b>Fish</b>					
Arctic char <sup>a</sup>	I	Inland, Southern Iceland (farmed)			
Atlantic cod	D	Baltic Sea	0.16		
	D	Baltic Sea	0.18		
	D	Kattegat	0.16		
	D	Kattegat	0.13		
	D	North Sea	1.1		
	D	North Sea	1		
	N	Norwegian Sea	0.079	<0.021	<0.059
	S	Kattegat/Skagerrak	1.4	<0.057	<0.090
Atlantic halibut	I	Atlantic Ocean			
Atlantic herring	D	Kattegat	1.4		
	D	Kattegat	0.68		
	D	North Sea	1.1		
	D	North Sea	0.78		
	I	Atlantic Ocean			
	S	Kattegat/Skagerrak	0.92	<0.084	<0.138
Atlantic mackerel	N	North/Norwegian Sea	0.79	<0.40	<0.86
	S	Kattegat/Skagerrak	1.9	<0.050	<0.081
Atlantic salmon	D	Baltic Sea (wild)	0.18		
	D	Baltic Sea (wild)	0.16		
Baltic herring	F	Baltic Sea		<0.439	<0.179
	F	Baltic Sea		<0.962	<0.264
Black halibut	I	Greenland Sea			
Common ling	I	Atlantic Ocean			
	I	Atlantic Ocean			
European plaice	D	Baltic Sea	1.1		
	D	Baltic Sea	1.3		
	D	Kattegat	0.83		
	D	Kattegat	1.9		
	D	North Sea	0.98		
	D	North Sea	1.6		
	N	Norwegian Sea	0.29	<0.038	<0.093
	S	Kattegat/Skagerrak	1.3	<0.049	<0.076
Haddock	I	Atlantic Ocean			

	N	Norwegian Sea	0.32		
Hake	S	Kattegat/Skagerrak	1.2		
Perch	F	Baltic Sea			
Pike	F	Baltic Sea		<1.66	<0.371
	F	Baltic Sea		<0.309	<0.122
	F	Baltic Sea		<0.603	<0.226
Pike-perch <sup>a</sup>	S	Inland, Sweden	0.24		
Rainbow trout	F	Baltic Sea (farmed)			
	N	Norwegian Sea (farmed)	0.024	0.038	<0.061
Saithe	I	Greenland Sea			
	N	Norwegian Sea	0.40	<0.033	<0.088
	S	Kattegat/Skagerrak	0.56	0.080	<0.075
Skipjack tuna, canned	N	Seas near Thailand/Vietnam	N/A <sup>b</sup>	<0.11	<0.22
Whitefish	F	Baltic Sea			
<b>Shellfish</b>					
Blue mussel	I	Atlantic Ocean			
Blue mussel	N	Norwegian Sea (farmed)	77	1.1	1.5
Blue mussel	S	Kattegat/Skagerrak (farmed)	37		
Brown crab	N	Norwegian Sea	5.3	<0.06	<0.12
Great scallop	N	Norwegian Sea	0.94	<0.98	<0.45
Northern prawn	I	Kattegat/Skagerrak			
Northern prawn	N	Norwegian Sea	2.1	<0.022	<0.050
Northern prawn	S	Kattegat/Skagerrak	30	<0.064	<0.099
Norway lobster	D	Kattegat			

<sup>a</sup> Freshwater origin

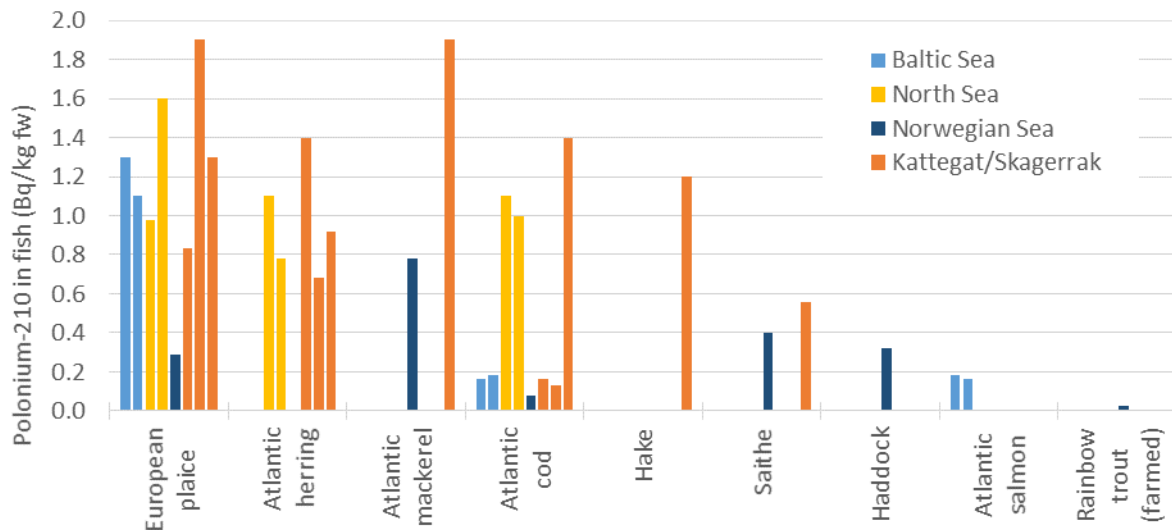
<sup>b</sup> Mix of six common brands. Expiration dates and production dates indicate that the fish was caught around one year prior to purchase. Due to the long delay, <sup>210</sup>Pb analyses must be performed in order to adequately estimate <sup>210</sup>Po concentrations.

### <sup>210</sup>Po concentrations

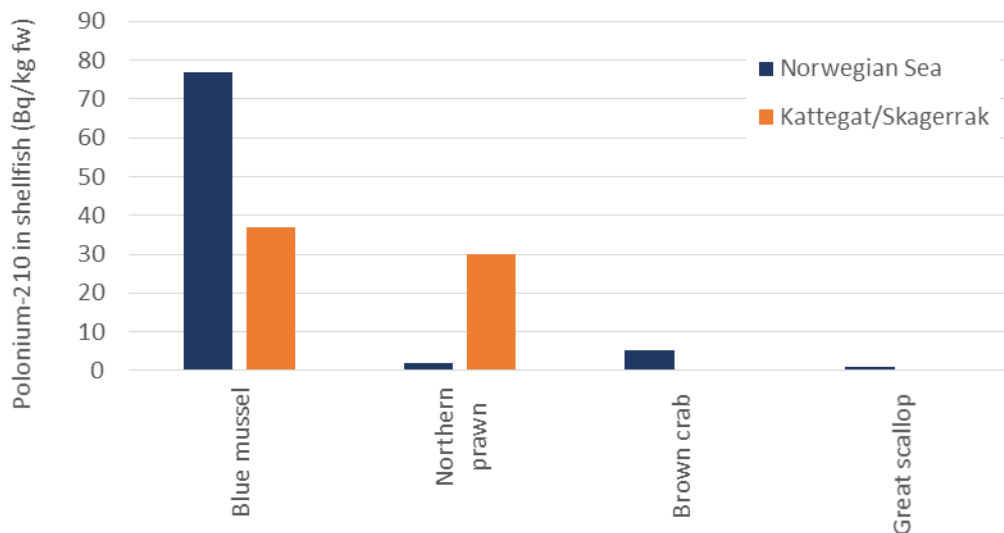
Concentrations of <sup>210</sup>Po in *wild* fish analysed so far ranged from 0.079 to 1.9 Bq/kg, with the lowest levels found in cod from the Norwegian Sea and the highest level found in samples of mackerel and plaice from the Kattegat/Skagerrak region. Overall, the highest levels generally found in samples of mackerel, plaice, and herring, but also samples of cod and hake were among those containing more than 1 Bq/kg (Table 5-1 and Figure 5-1). The lowest <sup>210</sup>Po concentration of all samples was found in farmed rainbow trout from Northern Norway, containing 0.024 Bq/kg.

Shellfish generally contained more <sup>210</sup>Po than fish, ranging from 0.94 to 77 Bq/kg. The highest concentrations were found in blue mussels on the Swedish and Norwegian coasts, containing 37 and 77 Bq/kg, respectively. Completed analyses of northern prawn varied from 2.1 to 30 Bq/kg. The sample of scallops analysed included only pure muscle, which likely explain the much lower concentrations than in blue mussels, in which the digestive gland was included in analysis.

An illustration of differences in <sup>210</sup>Po concentration between both species and geographical regions in this work is presented in Figures 5-1 and 5-2. Results also show significant variations in <sup>210</sup>Po concentrations exist within different samples of the same species. Potential reasons for this are discussed in chapter 6.



**Fig. 5-1.** <sup>210</sup>Po concentrations (Bq/kg fw) in completed analyses of fish from the sea, showing preliminary results from different species and regions..



**Figure 5-2.** <sup>210</sup>Po concentrations (Bq/kg fw) in completed analyses of shellfish, showing preliminary results from different species and regions.

### <sup>226</sup>Ra and <sup>228</sup>Ra concentrations

Completed radium analyses show values mainly below detection limits. This is not surprising, as <sup>226</sup>Ra and <sup>228</sup>Ra concentrations in muscle are generally low. Radium acts similar to calcium, and accumulates mainly in bones. However, high dose conversion factors mean that doses are not necessarily insignificant even though results are below detection levels<sup>8</sup>. Results below the limit also provide valuable information – and the lower the detection limit, the more valuable the data. Efforts were made to perform analyses with detection limits as low as possible. By ashing the samples before performing gamma spectrometry, as was procedure for Swedish

<sup>8</sup> <sup>226</sup>Ra, <sup>228</sup>Ra, and <sup>210</sup>Po ingestion dose coefficients from the ICRP are about two order of magnitude higher than e.g. for <sup>137</sup>Cs for children and adults. Respective coefficients for adults is  $2.8 \cdot 10^{-8}$ ,  $6.9 \cdot 10^{-7}$ , and  $1.2 \cdot 10^{-6}$ , while for 1-year-olds, they are  $9.6 \cdot 10^{-7}$ ,  $5.7 \cdot 10^{-6}$ , and  $8.8 \cdot 10^{-6}$  (ICRP 2012).

samples and the majority of Norwegian samples, detection limits of 0.02-0.08 Bq/kg for  $^{226}\text{Ra}$  and 0.05-0.14 Bq/kg for  $^{228}\text{Ra}$  were obtained.

Some of the completed analyses are also above detection limit. The highest  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  concentrations in samples analysed so far have been found in blue mussels from Northern Norway, containing 1.1 and 1.5 Bq/kg, respectively – the same sample that contained the highest  $^{210}\text{Po}$  concentration.

## 6. Discussion

Since radium results are partially incomplete and most of the data for completed analyses are below detection levels, it is difficult to make many comparisons between species, regions and other potential influencing factors. Further discussions on  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  will be more appropriate when all analyses are complete in 2019. Therefore,  $^{210}\text{Po}$  results will be the main topic of this discussion.

### *Results compared to previous work*

The preliminary  $^{210}\text{Po}$  results from the NANOD project are generally well within the range of activity concentrations reported in previously performed studies in the Nordic region (Table 3.1-1). One exception was the  $^{210}\text{Po}$  concentration of 77 Bq/kg in blue mussels on the Norwegian coast, which was higher than in the previous Nordic studies available, showing 7.5 Bq/kg in mussels from the Baltic Sea and 37 Bq/kg from the coast of Denmark. However, similar or higher concentrations have been reported from other regions, for example by Bustamante et al. (2002) from the French Atlantic coast or by Ryan et al. (1999) from the Irish coast. Therefore, the concentration still appear to be within the normal range.

As for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  results, there is still several analyses yet to be completed in the NANOD project, and there is little Nordic data available for comparison.  $^{226}\text{Ra}$  analyses of Atlantic cod in the Norwegian Sea and Kattegat/Skagerrak (<0.021 and <0.057 Bq/kg) so far indicate slightly lower values than the mean value reported in the HELCOM database for measurements in the Baltic Sea (HELCOM 2018)<sup>9</sup>. In cases when detection limits are sufficiently low, this information is much more useful for use in dose assessments than the alternative references values available. For example, UNSCEAR (2000) provides a reference value of 0.1 Bq/kg for  $^{226}\text{Ra}$  in fish products, and Brown et al. (2004) suggested a reference value of  $^{226}\text{Ra}$  of 0.2 Bq/kg in fish and 0.7 Bq/kg in shellfish in the European region. None of these found sufficient material to provide a reference level for  $^{228}\text{Ra}$ . Hosseini et al. (2010) used a reference value of 1.8 Bq/kg for  $^{228}\text{Ra}$  for use with ICRPs Reference Animals and Plants, although this was based on data from an area with enhanced levels of natural radioactivity. Representative data on  $^{228}\text{Ra}$  in fish and shellfish are therefore in demand. Most values below detection limits in the present work suggest that  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  values are below these reference values.

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<sup>9</sup> The mean  $^{226}\text{Ra}$  value of 0.19 Bq/kg in the HELCOM database appears to be strongly affected by two reported values from the 1980s that are so high that they appear to be erroneously registered. The median value registered in the database 0.08 Bq/kg, which seems more reasonable, although still somewhat higher than preliminary NANOD results. The HELCOM database only include data from the Baltic Sea.

### *Variation between species*

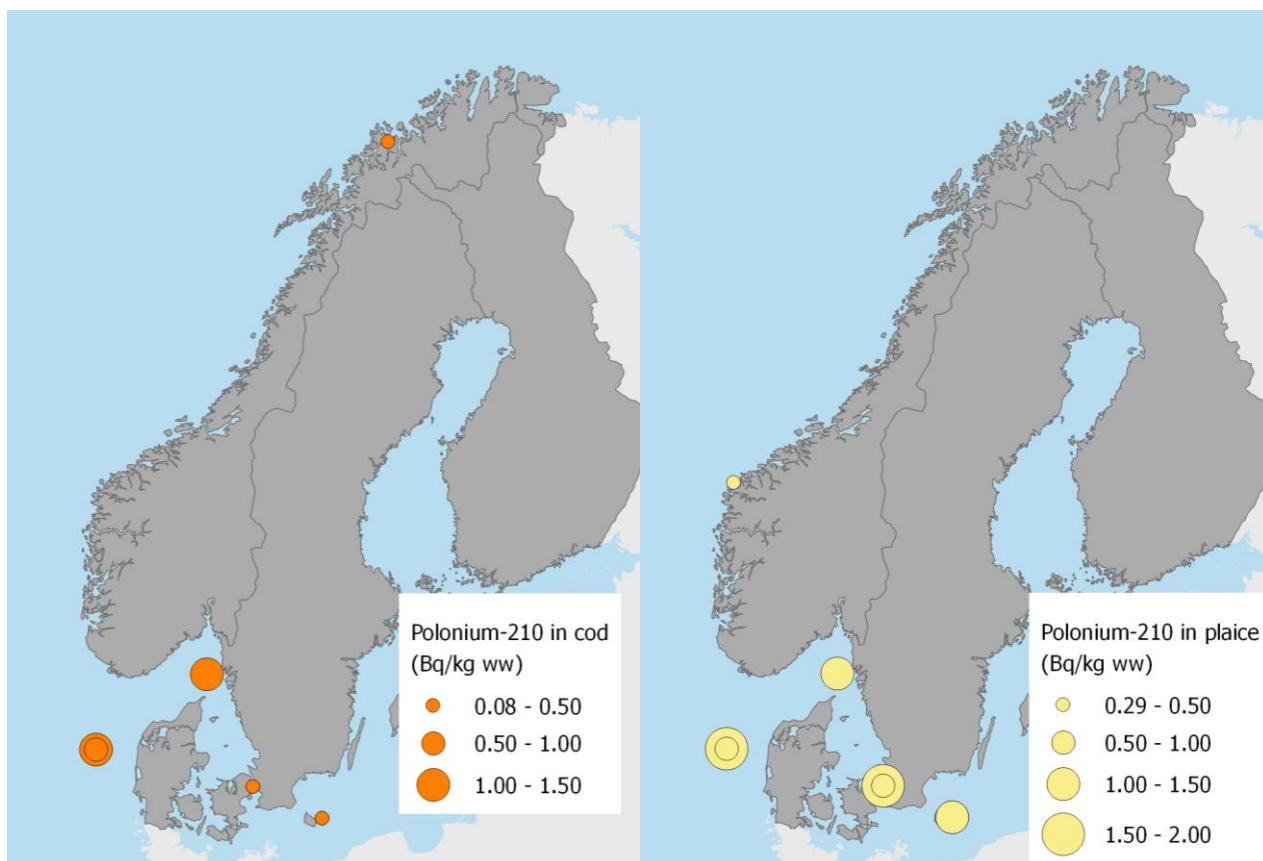
Differences in radionuclide concentrations between different species is believed to mainly be due to their different diets, as  $^{210}\text{Po}$  enters the body primarily via ingestion (Carvalho & Fowler 1994, Carvalho 2011). Polonium is particle reactive, adhering to surfaces in the marine environment, and thus the vast majority of  $^{210}\text{Po}$  in seawater is associated with suspended particles, including plankton (Wildgust et al. 1999; Carvalho et al. 2011; Ryan et al. 1999; Skwarzec & Bojanowski 1988). It has been shown that species like mackerel and herring, whose diet mainly consists of plankton or other small plankton consumers, generally have higher  $^{210}\text{Po}$  concentrations than larger predators like cod and salmon (Carvalho 2011, Fernando & Carvalho 2011). Similarly,  $^{210}\text{Po}$  is expected to be particularly high in filter feeders feeding directly on particles, such as blue mussels, and also elevated in consumers of filter feeders/bottom feeders, such as plaice. This is in line with the results from this study, finding the highest activity concentrations among the fish species in plaice, herring, and mackerel. However, cod shows extremely variable results, ranging from 0.079 to 1.4 Bq/kg.

### *Geographic variations*

In addition to the differences between species, there are sometimes large differences between samples of the same species caught in different regions. Dahlgaard (1995) examined geographical effect on  $^{210}\text{Po}$  concentrations in cod, plaice, and herring caught in the Baltic Sea, Kattegat, and the North Sea, but no significant geographical differences were found. The concentrations in the various species were in the same range as in this work. Carvalho (2011) compared organisms of different ocean depths, and found no apparent difference in radionuclide levels between organisms inhabiting the various depths. Instead, it was observed that the species inhabiting the same ecological niches tended to have comparable  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  levels, despite different habitats. Neither did Pearson et al. (2016) find any significant differences between various species in the coastal regions of New Zealand.

Any variation in geographic region that may exist, could also be related to diet, as different foods may be available for the same species in different regions. One variant of this is that populations occupying a coastal niche may have a very different diet than populations of the same species inhabiting the larger oceans, as is the case with e.g. cod inhabiting coastal areas vs. open seas.

Based on the data gathered so far in this work, it would appear that fish from the Norwegian Sea, and perhaps the Baltic Sea, contains somewhat lower  $^{210}\text{Po}$  concentrations than the North Sea and Kattegat/Skagerrak (Figures 5-1 and 5-2). Looking at the geographical variation of  $^{210}\text{Po}$  in plaice and cod (the species with most data points) in maps of the region, as presented in Figure 6-1, it is easy to draw such a conclusion. However, one must consider that the apparent differences between regions could also be affected by other factors. For example, some studies have indicated seasonal variations in  $^{210}\text{Po}$  concentrations. Since sampling could not be carried out at the same time by all countries, this and other potential influences should be considered as well. In addition, of the blue mussels analysed so far, the geographical effect was opposite of that seen the other species, with higher levels in Northern Norway than in Kattegat/Skagerrak. Once the final  $^{210}\text{Po}$  results are also complete, there should be a better basis for determining whether there are substantial geographical variations in this study.



**Figure 6-1.** Maps showing  $^{210}\text{Po}$  concentrations in the two species with the most available results: cod and plaice. Locations are approximate, especially in the Kattegat/Skagerrak area and the North Sea.

### *Temporal effects*

Several studies have looked into seasonal variations in molluscs; however, with mixed outcomes. Carvalho et al. (2011) examined monthly variations in  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations in the Mediterranean mussel on the coast of Portugal, also in relation to the physiological condition of the mussels. Despite no clear changes in seawater concentrations, the authors found an apparent seasonal fluctuation throughout the year. However, this was believed to be caused by changes in body weight due to storage of lipids.  $^{210}\text{Po}$  generally binds to protein and amino acids, not fat, meaning the activity per mussel can remain fairly constant and that only variations in body mass affect the activity concentration. Similarly, Wildgust et al. (1999) found an increase in  $^{210}\text{Po}$  levels in the common periwinkle on the Welsh coast in the summer, likely due to a drop in body weight due to spawning. Ryan et al. (1999) found significant temporal variability in blue mussels in several sites on the Irish coast, but no clear patterns. Germain et al. (1995), on the other hand, found no distinct seasonal changes in  $^{210}\text{Po}$  concentrations in edible parts of blue mussels on the coast of France. The temporal effect was examined for  $^{210}\text{Po}$  concentrations in the present study, but no apparent seasonal patterns were observed.

Ryan et al. (1999) also found strong correlation between  $^{210}\text{Po}$  concentrations of suspended material in seawater and the turbidity of the seawater at the given site. Since this factor changes according to both place and time, it could be one possible explanation for both geographic and temporal variations that might be observed in some species.

### *Variation between individuals or populations of same species*

Bulk samples were analysed in this work, as it was not within the scope of this study to analyse differences between individuals. However, some of the factors that affect individuals can also apply to whole populations and are therefore relevant to discuss. One such factor is that the exact ecological niche and individual feeding habits may affect differences in radionuclide concentrations within the same species (Cherry & Heyraud 1991; Carvalho et al. 2011). Individual body size or condition can also have an effect on radionuclide concentrations. Dahlgaard (1995) observed significantly higher  $^{210}\text{Po}$  levels in blue mussel soft parts with low condition index<sup>10</sup>. Ryan et al. (1999) found no clear correlation with condition index, but found a strong linear dependency between  $^{210}\text{Po}$  concentration and dry matter content of blue mussel soft tissues, with higher concentrations in smaller individuals. As a side note, studies of cod have also found that different populations of cod are genetically different, including having different growth rates (IMR 2018). It is possible that e.g. growth rates could also potentially influence radionuclide accumulation.

Large variability between individuals sampled at the same place and time has also been observed. For example, Dahlgaard (1995) found standard deviation values ranging from 70-100% in plaice, herring, and cod from the same catches. The significant variations between individuals emphasize the importance of analysing a large number of individuals, or bulk samples consisting of many individuals, in order to obtain representative values.

### *Farmed vs. wild fish*

The sample of Norwegian farmed rainbow trout analysed in this work contain a lower  $^{210}\text{Po}$  concentration (0.024 Bq/kg) than what was generally found in the wild fish samples, and the levels are in line with what Heldal et al. (2017) found in farmed salmon along the Norwegian coast (0.003-0.023 Bq/kg). Rainbow trout and salmon are large predator fish that would be expected to contain relatively low  $^{210}\text{Po}$  concentrations compared to fish lower on the food chain in a wild setting. However, the  $^{210}\text{Po}$  concentrations in wild salmon from the Bornholm area analysed in this work (0.16-0.18 Bq/kg), are nonetheless about one order of magnitude higher than that of the farmed salmon. The observation of lower concentrations observed in farmed fish vs. wild fish is likely due to the different diets, as the fish feed used in farming consists of feed produced mainly from plant-based ingredients.

The share of farmed fish in the Nordic countries is relatively high. In Sweden, for example, an estimated 40% of seafood consumed is farmed (Ziegler & Bergman 2017). Therefore, it is important to also include farmed fish in studies of naturally occurring radionuclides.

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<sup>10</sup> There are several methods of determining the ‘condition index’ (CI), although most rely on the relationship between weight of the mussel’s soft parts (dry weight) vs. length, volume or weight of the shell. Dahlgaard (1995) defined CI in relation to length,  $\text{CI} = \text{g dry soft parts} \cdot 10^6 \text{ mm}^{-3}$ . Ryan et al. (1999) used  $\text{CI} = \text{dry flesh weight} / \text{dry shell weight}$ . It’s possible that different methods used for determining CI influenced the contrasting outcomes.



### *Nordic fish and shellfish in the global market*

Nordic seafood makes up over 10% of worldwide exports in terms of trade value (FAO 2018). Norway is the biggest Nordic exporter, second only to China in the global market. This means that Nordic seafood is not only important to Nordic consumers, it is also an important part of global consumption, making it all the more relevant to have knowledge of radionuclide levels. Documentation of concentrations of anthropogenic radionuclides for exported species are already frequently requested by exporters. Requests for documentation of natural radioactivity is still relatively scarce; however, the IAEA, FAO and WHO currently have a joint project to examine the need for guidelines for naturally occurring radionuclides in food as well (IAEA 2017), including fisheries products. Such guidelines may increase the need for data on natural radioactivity in seafood.

## 7. Conclusions

All Nordic countries have a substantial fish and shellfish consumption, which varied from around 37 to 57 g/d for an average adult. There is also a large variation in species composition among the countries.

The results from analyses completed so far show that concentrations of  $^{210}\text{Po}$  in wild fish ranged from 0.079 to 1.9 Bq/kg in muscle tissue, while the activity concentration in shellfish were in the range of 0.94 to 77 Bq/kg. Overall, higher  $^{210}\text{Po}$  levels were observed in species with a  $^{210}\text{Po}$ -rich diet, such as plankton. This is in line with previous studies showing that  $^{210}\text{Po}$  enters the organisms primarily via ingestion. Moreover, the sample of farmed trout showed lower  $^{210}\text{Po}$  levels than wild fish, likely due to a diet of feed based mainly on plant material low in  $^{210}\text{Po}$ . However,  $^{210}\text{Po}$  levels although varied considerably within the same species. Some of this variability may be related to geographic differences, although it is not clear what the direct cause of this is. Results of the present and previous studies indicate that influences on  $^{210}\text{Po}$  concentrations in fish and shellfish are complex, and several factors may play a role.

Results of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  analyses performed so far are mainly below the detection limits, but still provide valuable information. Preliminary findings indicate highest level in blue mussels, with 1.1 and 1.5 Bq/kg, respectively.

Data on naturally occurring radionuclides in fish and shellfish is important not only due to the food group's role in the Nordic diet, but also because the Nordic countries play an important role in the worldwide fishing industry. A better picture of variation and possible patterns among fish and shellfish will be discussed when remaining analyses are complete. The complete data set, including  $^{210}\text{Pb}$  results, and associated discussions and dose estimates are expected in the 2019 final report, provided continued funding of the project in 2019.

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### Disclaimer

The views expressed in this document remain the responsibility of the author(s) and do not necessarily reflect those of NKS. In particular, neither NKS nor any other organisation or body supporting NKS activities can be held responsible for the material presented in this report.

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## Appendix A – Previously collected data

Detailed overview of data from previous studies in the Nordic region.

Species	Region	Mean levels (min-max) in Bq/kg fw, and associated number of batches (and/or individuals in parentheses) analysed in the study								References
		<sup>210</sup> Po		<sup>210</sup> Pb		<sup>226</sup> Ra		<sup>228</sup> Ra		
		Bq/kg	n	Bq/kg	n	Bq/kg	n	Bq/kg	n	
Farmed salmon	Coast of Norway	0.013 (0.003-0.023)	7	0.03-0.07	7	<DL 0.04 - <0.18	100	<0.006- <0.39	100	Heldal et al. 2017
Cod	Baltic Sea (Southern Baltic and Bay of Gdansk)	2.3 (0.9-3.3)	4 (9)							Cited in Holm 1994
	Coast of Norway	2.1 (0.9-4)	3 (75)							Holm 1994
	Coast of Sweden	3.5 (3.0-3.9)	2 (12)							Holm 1994
	Coast of Iceland	0.9	1 (10)							Holm 1994
	Baltic Sea	0.38 (0.043-1.5)	41	0.069 (0.062-0.082)	4	0.19 (0.042-4.9)	79			HELCOM 2018
	Barents Sea	0.518	2	0.058	2					NRPA monitoring data
	Coast of Norway	0.26	23							Heldal et al. 2015
	North Sea	0.83	5							Heldal et al. 2015
	North Sea and Baltic Sea	0.35	1 (13)							Dahlgard 1995
Haddock	Barents Sea					0.188	1			NRPA monitoring data
	Coast of Norway	1.35	2							Heldal et al. 2015
	North Sea	1.45	2							Heldal et al. 2015
Mackerel	Barents Sea	1.29	1	0.068						NRPA monitoring data

Species	Region	Mean levels (min-max) in Bq/kg fw, and associated number of batches (and/or individuals in parentheses) analysed in the study								References
		<sup>210</sup> Po		<sup>210</sup> Pb		<sup>226</sup> Ra		<sup>228</sup> Ra		
		Bq/kg	n	Bq/kg	n	Bq/kg	n	Bq/kg	n	
	North Sea	2.5	22							Heldal et al. 2015
Saithe	Coast of Norway	0.92	2							Heldal et al. 2015
Herring	Baltic Sea	8.3 (1.9-23)	4 (23)							Cited in Holm 1994
	Coast of Norway	2.8 (0.5-5)	2 (50)							Holm 1994
	Coast of Sweden	4.0 (1.6-9.6)	7 (105)							Holm 1994
	Baltic Sea	1.36 (0.19-8.5)	41	0.17 (0.076-0.3)	5	0.028 (0.02-0.055)				HELCOM 2018
	Barents Sea	2.91	1	0.108	1					NRPA monitoring data
	North Sea	2.88	14							Heldal et al. 2015
	Baltic Sea	3.247	1	0.183						STUK monitoring data
	Baltic Sea	1.944	1	0.247						STUK monitoring data
	Baltic Sea	3.958	1	0.454						STUK monitoring data
	Baltic Sea	0.586	1	0.041						STUK monitoring data
	Baltic Sea	1.656	1	0.110						STUK monitoring data
	North and Baltic Sea	0.65	1 (14)							Dahlgaard 1995
Redfish	Coast of Norway	0.16	1							NRPA monitoring data
Plaice	Coast of Norway	10.3 (8-12)	3 (75)							Holm 1994
	Coast of Iceland	6.4	1 (15)							Holm 1994



Species	Region	Mean levels (min-max) in Bq/kg fw, and associated number of batches (and/or individuals in parentheses) analysed in the study								References
		<sup>210</sup> Po		<sup>210</sup> Pb		<sup>226</sup> Ra		<sup>228</sup> Ra		
		Bq/kg	n	Bq/kg	n	Bq/kg	n	Bq/kg	n	
	Baltic Sea	1.1 (0.26-3.4)	42	0.1 (0.055-0.15)	4					HELCOM 2018
	North and Baltic Sea	0.96	1 (14)							Dahlgaard 1995
Pike	Coast of Finland	2.8 (1.7-3.8)	2							Holm 1994
	Baltic Sea	0.94	1	0.092	1					HELCOM 2018
Perch	Coast of Finland	0.2 (0.2-0.2)	2							Holm 1994
	Bothnian Sea	0.327 (0.28-0.37)	8	0.088	8					Gjelsvik et al. 2009
	Bothnian Sea	0.042 (0.038-0.048)	3	0.013 (0.010-0.018)	3					Gjelsvik et al. 2009
	Baltic Sea	<0.14- <0.15	2	<0.14- <0.15	2	<0.95- <1.8	2	<0.54- 0.54	2	Vesterbacka 2018
	Unknown (Finland)	<0.15	1	<0.15	1	<3.2	1	<1.3		Vesterbacka 2018
Vendace	Unknown (Finland)	1.29 (0.79-1.64)	3	<0.38- <0.47	3	<2.2-<4	3	<0.64- <1.2		Vesterbacka 2018
Whitefish	Coast of Finland	1.9 (0.8-2.9)	2							Holm 1994
	Baltic Sea	0.244		0.018						STUK monitoring data
	Baltic Sea	0.380		0.022						STUK monitoring data
	Baltic Sea	13	1	<0.25	1	<0.96	1	<0.37	1	Vesterbacka 2018
	Unknown (Finland)	0.40 (<0.23-0.82)	3	<0.16- <0.23	3	<0.43- <0.79	3	<0.16- <0.28	3	Vesterbacka 2018

Species	Region	Mean levels (min-max) in Bq/kg fw, and associated number of batches (and/or individuals in parentheses) analysed in the study								References
		<sup>210</sup> Po		<sup>210</sup> Pb		<sup>226</sup> Ra		<sup>228</sup> Ra		
		Bq/kg	n	Bq/kg	n	Bq/kg	n	Bq/kg	n	
Capelin	Coast of Iceland	5.3	1							Holm 1994
Sprat (brising)	Baltic Sea					0.073 (0.05-0.11)	75			HELCOM 2018
Flounder	Baltic Sea	6.7 (2.7-16)	15							Cited in Holm 1994
	Baltic Sea					0.046 (0.026-0.075)	71			HELCOM 2018
Rainbow trout	Baltic Sea	0.039		0.013						STUK monitoring data
	Unknown (Finland)	<0.26	1	<0.26	1	<0.73	1	<0.25	1	Vesterbacka 2018
Blue mussel	Baltic Sea					1.4 (0.029-12)	11	1.5 (0.35-3.4)	3	HELCOM 2018
	Baltic Sea			2.186						STUK monitoring data
	Baltic Sea			2.821						STUK monitoring data
	Baltic Sea	7.541		1.172						STUK monitoring data
	Coast of Denmark (incl. East and West coasts)	37 (est. from 149 d.w.)	1 (72)							Dahlggaard (1995)
Cockle	Baltic Sea		1			0.71				HELCOM 2018
Baltic clam	Baltic Sea					2.5 (0.64-3.98)	3			HELCOM 2018

## Appendix B - Sample overview

Details on collected samples. All samples consist of fish muscle or edible parts of shellfish.

Country	Species	Catch date	Origin	Comment
Denmark	Atlantic cod	19.02.2018	Kattegat FAO 23	
	Atlantic cod	19.02.2018	Kattegat FAO 23	
	European plaice	19.02.2018	Kattegat FAO 23	
	European plaice	19.02.2018	Kattegat FAO 23	
	Atlantic herring	19.02.2018	Kattegat FAO 23	
	Atlantic herring	19.02.2018	Kattegat FAO 23	
	Atlantic cod	06.03.2018	North Sea FAO 27-A	
	Atlantic cod	06.03.2018	North Sea FAO 27-A	
	European plaice	06.03.2018	North Sea FAO 27-B	
	European plaice	06.03.2018	North Sea FAO 27-B	
	Atlantic herring	06.03.2018	North Sea FAO 27-A	
	Atlantic herring	06.03.2018	North Sea FAO 27-A	
	Atlantic cod		Bornholm FAO 25	
	Atlantic cod		Bornholm FAO 25	
	European plaice		Bornholm FAO 25	
	European plaice		Bornholm FAO 25	
	Salmon		Bornholm FAO 25	
	Norway lobster		Kattegat FAO 23	
Finland	Baltic Herring	24.11.2017	Bothnian Bay, Hailuoto	
	Baltic Herring	21.10.2017	Bothnian Sea, Seili	
	Pike	12.11.2017	Bothnian Bay, Hailuoto	
	Pike	04.05.2017	Bothnian Sea, Seili	
	Pike	20.05.2017	Bothnian Sea, Vaasa	
	Perch		Baltic Sea	
	Rainbow trout		Baltic Sea	Farmed
	Whitefish		Baltic Sea	
Iceland	Common ling	01.03.2018	Denmark Strait (West of Iceland)	
	Common ling	28.02.2018	South of Iceland	
	Atlantic cod	08.03.2018	Denmark Strait (West of Iceland)	
	Haddock	28.02.2018	South of Iceland	
	Saithe	12.03.2018	North of Iceland	
	Black halibut	02.03.2018	North of Iceland	
	Atlantic halibut	31.07.2018	South of Iceland	
	European plaice	31.07.2018	Breiðafjörður (West coast)	
	Arctic char	31.07.2018	Southern Iceland (inland)	Farmed, freshwater
	Atlantic herring	31.07.2018	South-East of Iceland	Salted
	Northern prawn	16-21.05.2018	West of Iceland	
	Northern prawn	07-11.04.2018	North of Iceland	
	Northern prawn	13.04.2018	Isafjarðardjúp (inner part of fjord, North-West coast)	
Blue mussels	02.05.2018	South-Western coast of Iceland		
Norway	Atlantic mackerel	24.06.2018	Bremanger, Sogn og Fjordane	
	Great scallop <sup>a</sup>	08.08.2018	Hitra, Trøndelag	
	Brown crab	08.08.2018	Fosen, Trøndelag	
	Rainbow trout	08.08.2018	Stokmarknes, Nordland	Farmed

Country	Species	Catch date	Origin	Comment
	European plaice	08.08.2018	Near Stadt, Sogn og Fjordane	
	Skipjack tuna	01.09.2017	Thailand/Vietnam	Canned
	Atlantic cod	17.08.2018	Troms	
	Haddock	17.08.2018	Troms	
	Saithe	17.08.2018	Troms	
	Northern prawn	05.09.2018	Troms	
	Blue mussel	20.08.2018	Troms	Farmed
Sweden	Atlantic mackerel	03.09.2018	Kattegat/Skagerrak	
	European plaice	06.09.2018	Kattegat/Skagerrak	
	Atlantic cod	18.08.2018	Kattegat/Skagerrak	
	Atlantic herring	30.08.2018	Kattegat/Skagerrak	
	Saithe	18.08.2018	Kattegat/Skagerrak	
	Hake	18.08.2018	Kattegat/Skagerrak	
	Pike-perch	30.08.2018	West coast region	Freshwater
	Blue mussel	08.06.2018	Mollösund	Farmed
Northern prawn	30.08.2018	Lysekyl		

a. Muscle only

## Appendix C - List of English and Latin species names

English	Latin
Alaska pollock	<i>Gadus chalcogrammus</i>
Atlantic salmon	<i>Salmo salar</i>
Arctic char	<i>Salvelinus alpinus</i>
Atlantic cod	<i>Gadus morhua</i>
Atlantic halibut	<i>Hippoglossus hippoglossus</i>
Atlantic herring	<i>Clupea harengus</i>
Atlantic mackerel	<i>Scomber scombrus</i>
Baltic Herring	<i>Clupea harengus membras</i>
Black halibut	<i>Reinhardtius hippoglossoides</i>
Blue mussel	<i>Mytilus edulis</i>
Brown crab	<i>Cancer pagurus</i>
Brown trout	<i>Salmo trutta</i>
Common ling	<i>Molva molva</i>
Common periwinkle	<i>Littorina littorea</i>
European hake	<i>Merluccius merluccius</i>
European perch	<i>Perca fluviatilis</i>
European plaice	<i>Pleuronectes platessa</i>
European whitefish	<i>Coregonus lavaretus</i>
Greater argentine	<i>Argentina silus</i>
Great scallop	<i>Pecten maximus</i>
Haddock	<i>Melanogrammus aeglefinus</i>
Mediterranean mussel	<i>Mytilus galloprovincialis</i>
Northern prawn	<i>Pandalus borealis</i>
Norway lobster	<i>Nephrops norvegicus</i>
Pangasius	<i>Pangasius sp.</i>
Pike	<i>Esox lucius</i>
Pike-perch	<i>Sander lucioperca</i>
Rainbow trout	<i>Oncorhynchus mykiss</i>
Redfish	<i>Sebastes sp.</i>
Saithe	<i>Pollachius virens</i>
Skipjack tuna	<i>Katsuwonus pelamis</i>
Vendace	<i>Coregonus albula</i>
Wolffish	<i>Anarhichas lupus</i>