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Long-time variations of radionuclides and metals in the marine environment of the Swedish west-coast studied using brown algae (*Fucus serratus* and *Fucus vesiculosus*)

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Swedish Radiation Safety Authority

Research

Long-time variations of radionuclides and metals in the marine environment of the Swedish west-coast studied using brown algae

(*Fucus serratus* and *Fucus vesiculosus*)

2022:13

Authors: Sören Mattsson ¹⁾, Kristina Eriksson Stenström ²⁾, Guillaume Pedehontaa-Hiaa ³⁾

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SSM perspective

Background

The marine environment receives discharges from nuclear facilities around the world as well as in Sweden. The brown seaweeds, *Fucus serratus* and *Fucus vesiculosus*, are important organisms in the marine ecosystem both due to their ecological importance but also due to its well known ability to accumulate contaminants and thereby usefulness in environmental monitoring as a bio indicator. Bio indicators provide information on levels and variations of elements, like for example radionuclides, in the marine environment. By studying the elemental and radionuclide composition in brown algae over a time period it is possible to gain an increased understanding on processes affecting transfer and accumulation in the marine environment. In this study SSM supported additional analyses of radionuclides and stable metals in brown algae collected every second month at the same site on the Swedish west coast since 1967, which has resulted in a unique dataset covering over fifty years. The data set is useful in order to understand the fate of radionuclide discharges in the marine environment, in risk assessments and in case of emergency situations to provide background levels of relevant radionuclides. The project also contributes to the development of analysis techniques in environmental samples.

Results

This project has resulted in the completion of a unique half-century-long time series of concentrations of radionuclides and metals in brown sea weed on the Swedish west coast. Sea weed samples for the whole time period is also saved in a sample bank. For some radionuclides the whole time period is covered, while for others and for the stable elements parts of the time series are covered. Peaks and variations in the time series are described, compared with discharges and explained, which gives valuable information on how currents transfer radionuclides in the marine environment and on the time-scale for such processes. Analysis capacity for Pu and Am isotopes has been developed. Long-term changes as well as seasonal variations in concentrations is described and discussed. This has provided better information about long-term processes and seasonal variations for different radionuclides in both *Fucus serratus* and *Fucus vesiculosus*. The study also shows that the brown algae could be important for the monitoring of emissions of naturally occurring radioactive materials (NORM) from the offshore oil and gas industry by systematically studying long-term and seasonal variations of the content of ^{228}Ra and in the future also ^{226}Ra and ^{210}Pb .

The method development part of the project includes a modernisation of the methods used for the extraction and separation of actinides and the preparation of alpha spectroscopy sources. In addition, a method for the analysis of Pu isotopes in seaweed has been fully developed.

The project has also demonstrated the value of access to long time series of regularly collected samples of algae for other analyses than of radionuclides. Measurements on *Fucus* samples has also made it

possible to describe time trends for a number of heavy metals like lead and cadmium. It has demonstrated a decrease of mercury levels, probably an effect of the gradual phasing out of mercury in Sweden and other countries and an increase of the concentrations of gadolinium, which most likely is a consequence of the increased use of gadolinium-based contrast agents for MR imaging in our hospitals. The metal analyses contributes to assessment of transport routes for radionuclides generated in new radiation sources, such as gadolinium isotopes from the European Spallation Source (ESS).

The project has given new knowledge about the function and capacity of *Fucus* as a bioindicator especially regarding the degree of seasonal variations for various radionuclides and stable elements.

Relevance

The project's relevance for SSM is high as it is connected to several areas. Among other things, SSM participates in the OSPAR convention as a representative for Sweden, where, among other things, ^{238}Pu is included among the key nuclides that the member states report. Sweden has chosen not to report ^{238}Pu . For the Swedish part, the current project provide knowledge about how the levels of ^{238}Pu have developed in the marine environment over the past 50 years and thereby increase the ability to make a correct decision regarding monitoring and reporting according to the OSPAR convention for radioactive substances. Furthermore, the project has increased knowledge of how radioactive substances are spread and accumulated in the marine environment and thus become a tool for improving transport and dose models and thereby risk assessments for humans and the environment. The results will contribute to knowledge that the authority needs in order to make well-founded decisions and to maintain and develop national and international competence.

Need for further research

The sample bank can be used for further studies of radionuclides, stable elements, long-term transfer and accumulation in the marine environment. By completing the time series for important radionuclides and study potential correlations and connections in present dataset further understanding of the influence from nuclear facilities and from the offshore oil and gas industry (NORM) on the marine environment could be gained.

Maintaining the sample bank as a historical archive and continuing the analyses and collection of samples is valuable for the possibilities of further studies in general of marine transfer and accumulation processes on a long-term time scale.

Project information

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SSM2020-797 and SSM2018-905 / 226749, 4530335



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This report concerns a study which has been conducted for the Swedish Radiation Safety Authority, SSM. The conclusions and viewpoints presented in the report are those of the author/authors and do not necessarily coincide with those of the SSM.

Summary

The support from SSM has made it possible to continue collecting brown algae from the place on the Swedish west coast where regular collections began as early as 1967. The support has also enabled more extensive analyses of previously collected material than was possible before. This applies to both supplementary analyses of previously reported radionuclides and analysis of new ones. The project has also enabled a start of more comprehensive analyses of the overall results. This has provided better information about long-term processes and seasonal variations for different radionuclides in both toothed wrack (*Fucus serratus*) and bladderwrack (*Fucus vesiculosus*). What is new is that the sample material has now also been used to analyse metals, both those with known toxic effects such as lead, cadmium and mercury, as well as those that are of interest in assessing transport routes for radionuclides generated in new radiation sources, such as gadolinium isotopes from the European Spallation Source (ESS).

Regarding the long-term development of various radionuclides in the marine environment on the Swedish west coast, the studies show rapidly increasing levels of iodine-129, decreasing levels of cesium-137, technetium-99 and plutonium-239+240. The carbon-14 analyses suggest a continued inflow of this radionuclide from the North Sea. The study also shows that the brown algae could be important for the monitoring of emissions of naturally occurring radioactive materials (NORM) from the offshore oil and gas industry by systematically studying long-term and seasonal variations of the content of radium-228 and in the future also radium-226 and lead-210. Interesting and hitherto unexplained variations in the beryllium-7 content in *Fucus* have been made. The tritium content in *Fucus* and in seawater has been analysed in a limited number of samples from 2020. No levels beyond the expected normal ambient level for tritium have been found.

During the 10-year period 2011-2020, there was a doubling of the levels of lead and nickel and a 50% increase in the levels of cadmium and cobalt in *Fucus*. For mercury a 10-fold decrease is registered between 2011 and 2016 and then a doubling of the concentration between 2016 and 2020. The decreasing trend for mercury probably indicates an effect of the gradual phasing out of mercury in Sweden and other countries. However, some businesses and companies have an exemption for continued use. For gadolinium there is an increase with a factor of around 5 from 2011 to 2020, most likely explained by the increased use of gadolinium-containing contrast agents in magnetic resonance imaging in healthcare.

Sammanfattning

Stödet från SSM har gjort det möjligt att fortsätta insamlingar av brunalger från den plats på svenska västkusten där regelbundna insamlingar påbörjades redan 1967. Stödet har också möjliggjort mer omfattande analyser av tidigare insamlat material än vad som hittills varit möjligt. Detta gäller såväl kompletterande analyser av tidigare rapporterade radionuklider som analys av nya sådana. Projektet har också möjliggjort mer omfattande analyser av de sammantagna resultaten. Detta har gett bättre information om långtidsförlopp och säsongsvariationer för olika radionuklider i såväl sågtång som blåstång. Nytt är att provmaterialet också utnyttjats för att analysera metaller, såväl sådana med kända toxiska effekter som bly, kadmium och kvicksilver som sådana som är av intresse när det gäller att bedöma transportvägar för radionuklider som genereras i nya strålkällor, som gadolinium-isotoper från den Europeiska Spallationskällan (ESS).

Beträffande långtidsutvecklingen för olika radionuklider i den marina miljön vid svenska västkusten visar studierna på snabbt ökande nivåer av jod-129, minskande nivåer av cesium-137, teknetium-99 och plutonium-239+240. Kol-14 analyserna antyder fortsatt inflöde av denna radionuklid från Nordsjön. Skillnaderna i säsongsvariationer för olika radionuklider och dess skillnader mellan de två *Fucus*-arterna tror vi kan bidra till att lättare avslöja utsläppskällor. Studien visar också att brunalgerna skulle kunna ha betydelse för monitoreringen av utsläpp av naturligt förekommande radioaktiva material (NORM) från den havsbaserade olje- och gas-industrin genom att systematiskt studera långtids- och säsongsvariationer av innehållet av radium-228 och i framtiden även radium-226 och bly-210. Intressanta och hittills icke förklarade variationer i beryllium-7 innehållet i *Fucus* har gjorts. Tritiumhalten i *Fucus* och i havsvatten har analyserats i ett begränsat antal prover från 2020. Inga nivåer utöver den förväntade normala omgivningsnivån har konstaterats.

Under 10-årsperioden 2011-2020 skedde en fördubbling av halterna av bly och nickel och en 50-procentig ökning av halterna av kadmium och kobolt i *Fucus*. För kvicksilver registrerades en 10-faldig minskning mellan 2011 och 2016 och sedan en mindre uppgång av koncentrationen mellan 2016 och 2020, troligtvis som en effekt av den stegvisa utfasningen av kvicksilveranvändningen i Sverige och andra länder. Vissa verksamheter och företag har dock dispens för fortsatt användning. För gadolinium ses en kraftig ökning med en faktor på omkring 5 från 2011 till 2020, sannolikt förklarad av den ökade användningen av gadoliniuminnehållande kontrastmedel vid magnetresonansdiagnostik inom sjukvården.

Abbreviations and notations

AMS	Accelerator Mass Spectrometry
BCR	European Community Bureau of Reference
d.wt.	dry weight
EA-IRMS	Elemental Analyser - Isotope Ratio Mass Spectrometry
HELCOM	Baltic Marine Environment Protection Commission (<u>Helsinki Commission</u>)
IAEA	International Atomic Energy Agency
ICP-AES	Inductively Couple Plasma Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
LA-ICP-MS	Laser Ablation Inductively Coupled Plasma Mass Spectrometry
LOD	Limit of detection
OSPAR	Convention for the Protection of the Marine Environment of the North-East Atlantic (<u>Oslo-Paris Convention</u>).
SSAMS	Single Stage Accelerator Mass Spectrometry
SSM	Swedish Radiation Safety Authority
w.wt.	wet weight

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1. Introduction

1.1. Background

Algae can photosynthesize and produce their own nutrition by using light energy from the sun and carbon dioxide to generate carbohydrates and oxygen. They take their nutrients from the water and from there they also accumulate heavy metals, hydrocarbons, pesticides, PCBs, radionuclides and many other pollutants from the water (Barsanti and Gualtieri, 2014). Accumulation of pollutants makes marine macroalgae good bioindicators and has received significant attention because algae attached to rocks at the seabed reflect environmental conditions for long periods. They are also relatively easy to collect. Accumulation, turnover and release from the algae depend on the chemical properties of the substances, the concentration in the water and many other parameters such as algae uptake mechanisms, growth rate, light intensity, temperature, and salinity in the water. Knowledge of current concentrations is of great importance in assessing values measured in connection with future planned releases and nuclear and non-nuclear accidents and emergencies.

Other reasons to study the content of various substances in algae are that they are base for food additives in the form of alginate. Since long, algae have been used as fertilizers, soil conditioners, in small-scale sustainable agriculture and gardening as they are rich in potassium, nitrogen and phosphorus. Also in this way, elements and compounds in algae can be transferred to humans.

Anthropogenic radionuclides are introduced into the marine environment from several sources, as well as a number of natural radionuclides and other natural and anthropogenic substances, chemicals and elements. Anthropogenic radionuclides include global fallout from atmospheric nuclear weapons tests, discharges of radionuclides from nuclear installations, fallout from the Chernobyl disaster (1986), and from Fukushima Daiichi (2011), applications of radionuclides in medicine and in industry, past dumping of radioactive wastes, nuclear submarine accidents, loss of radioactive sources, contributions from nuclear test sites, and the burn-up of satellites using radionuclides as their power source (Povinec et al., 2003; Bailly du Bois et al., 2020). There is for example a recent indication of leakage from a previously unrecognized (or unreported) additional radioactive source in the Baltic Sea, e.g., disposed nuclear waste in the seabed (Qiao et al., 2021). Other industrial activities, such as mining and offshore oil and gas installations and phosphate fertilizer production, may change the distribution of naturally occurring radionuclides in the environment. Estuarine and marine sediments that have accumulated radionuclides over long periods can be an additional source after discharges from the point sources have ceased (OSPAR, 2010).

The water and thus also the levels of radioactive substances along the Swedish west coast are affected both by the influx from the North Sea and the outflow from the Baltic Sea. In addition, there are run-off from land and supply via sewers and sewage treatment plants. One way to follow changes in the marine environment is to use algae as bioindicators.

This project is mainly based on samples from one station (Särdal; 56.76N, 12.63E) located between Halmstad and Falkenberg (Figures 1, 2 and 3). Here, some of the first analyses of radionuclides in algae from Nordic coastal waters were made in 1967 (Mattsson 1984; Mattsson and Erlandsson 1991; Bernhardsson et al., 2008) and today there is a very long time series (55 years) of continuous sampling of both *Fucus serratus* (toothed wrack; *Sw. sågtång*) and *Fucus vesiculosus* (bladderwrack; *Sw. blåstång*) taken simultaneously. Samples have since the middle of the 1970s, i.e. during most of the time, been taken every second month and the collections continue in the same way.

1.2. International interest and cooperation

Due to the prevailing ocean currents, a large part of discharged, dumped or deposited radioactive wastes in northern European marine waters as well as from the Baltic will reach the Swedish west-coast waters. Important sources that have affected the levels of anthropogenic radionuclides during the last decades are global fallout from nuclear weapons testing in the 1950s and 1960s, liquid discharges from the nuclear fuel reprocessing plants at Sellafield (UK) and Cap de la Hague (France), and the Chernobyl disaster in 1986.

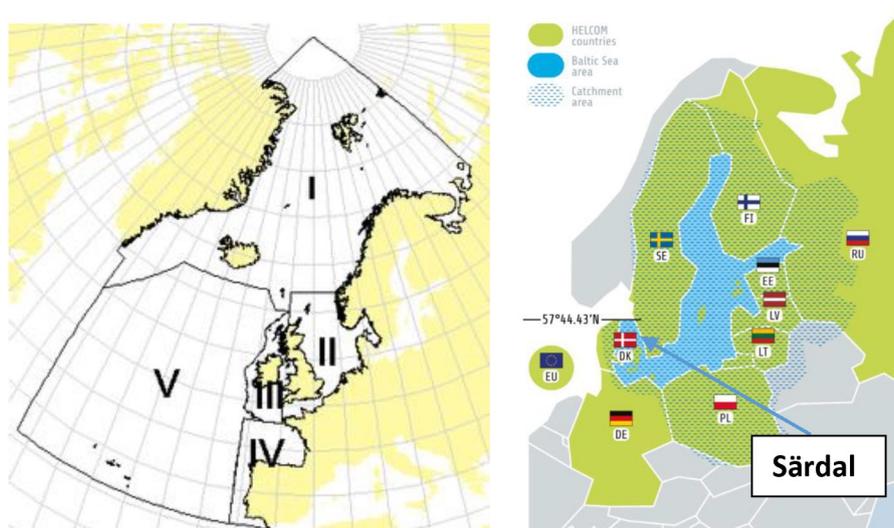


Figure 1: Map over the OSPAR area (left) and HELCOM area (right). OSPAR: Region I – Arctic waters, Region II – Greater North Sea, Region III – Celtic Seas, Region IV – Bay of Biscay and Iberian Coast, Region V – Wider Atlantic. HELCOM: “Baltic Sea Area” is defined as the Baltic Sea and the entrance to the Baltic Sea bounded by the parallel of the Skaw (Skagen) in the Skagerrak at 57.72N, 10.58E. It also includes the internal waters. The sampling place Särdal is also marked on the map.

The OSPAR Convention from 1992 (OSPAR, 2010) is the legal instrument guiding international cooperation for the protection of the marine environment of the North-East Atlantic (Figure 1). Work under the Convention is managed by the OSPAR Commission, made up of representatives of the Governments of 15 Contracting Parties and the European Commission. OSPAR is taking forward work related to the implementation of the Ecosystem Approach and five thematic strategies (on eutrophication, hazardous substances, radioactive substances, offshore oil and gas industry, biodiversity and ecosystems). There are almost 100 nuclear installations in the OSPAR catchment in the form of

nuclear power plants, nuclear fuel fabrication and enrichment plants, nuclear fuel reprocessing plants and research and development facilities. A parallel to OSPAR is the HELCOM (Helsinki Commission) organization (see Figure 1). HELCOM has 10 contracting parties: Denmark, Estonia, the European Union, Finland, Germany, Latvia, Lithuania, Poland, Russia and Sweden. For the purposes of the Helsinki Convention the “Baltic Sea Area” is defined as the Baltic Sea and the entrance to the Baltic Sea bounded by the parallel of the Skaw (*Sw. Skagen*) in the Skagerrak at 57.72N, 10.58E.

Nuclear fuel reprocessing plants, and nuclear fuel fabrication and enrichment plants stand for 98% of discharges of radionuclides from the nuclear sector (OSPAR, 2016, 2018). The input of radionuclides to the sea is associated with liquid discharges and to a lesser extent with solid wastes and emissions to air.

Much effort has been devoted to investigating the dispersion of anthropogenic radionuclides in the Northern European Seas from the nuclear fuel reprocessing plant in Sellafield (UK), which has been discharging into the Irish Sea since 1952, and La Hague (France), which has been discharging into the English Channel since 1966. The authorized release of radioactive waste has resulted in the widespread contamination of NE Atlantic waters with anthropogenic radionuclides such as ^3H , ^{14}C , ^{90}Sr , ^{99}Tc , ^{125}Sb , ^{134}Cs , ^{137}Cs , $^{239,240}\text{Pu}$ and others. These radionuclides have been used in numerous studies to investigate the movements of water masses over the NW European shelf (Povinec et al., 2003). The discharges of most radionuclides from the Sellafield and La Hague plants have been reduced by more than two orders of magnitude, compared with peak levels in the 1970s and 1980s (OSPAR, 2018). However, the resulting radionuclide concentrations in seawater are still easily measurable, partly as a result of remobilization from contaminated sediments (Mitchell et al., 1999). A third reprocessing plant at Dounreay, on the north coast of the Scottish mainland, also discharges directly into the sea. In general, contamination from this facility has been local and has tended to have been masked by the Sellafield contribution. However, contamination by ‘hot particles’ has been significant in a restricted area (Povinec et al., 2003). Inputs from nuclear power stations usually have been detectable only close to the discharge points. The Chernobyl accident in 1986 contributed significantly mainly to ^{134}Cs and ^{137}Cs inventories in seawater of the Baltic and North Seas, resulting in the Baltic Sea being the most highly contaminated by ^{137}Cs , more contaminated than e.g. the Black Sea (Povinec et al., 2003).

The main source of radioactive discharges from the medical sector is ^{131}I from treatment of thyroid cancer and thyrotoxicosis. However, its short half-life and delayed discharge via sewers means that only low activities of ^{131}I reach the marine environment.

The naturally occurring radionuclides in water include radium-226 and -228 (^{226}Ra and ^{228}Ra), lead-210 (^{210}Pb), and polonium-210 (^{210}Po). These radionuclides are also released during offshore oil and gas extraction.

2. Previously published results

Analyses have been made continuously for different gamma emitters where the artificial ones are dominated by ^{137}Cs (and ^{134}Cs after large releases from Sellafield and after the Chernobyl disaster) (Mattsson, 1984; Erlandsson and Mattsson, 1991; Bernhardsson et al., 2008). Analyses of radioactive cesium have provided new information on sources of pollution and large-scale transport modes and transport times from the source to the Swedish west coast (e.g. about 4 years from the Irish Sea to the Swedish west coast), information that is of interest also for other environmental pollutants.

During the summer of 1976, ^{60}Co began show up in the *Fucus* samples from Särödal, followed by other neutron activation products such as ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, ^{54}Mn , and others (Mattsson et al., 1980a). Analyses of samples from other places along the coast between Malmö and Gothenburg clearly pointed out the Barsebäck nuclear power plant as the dominant source for these activation products. Barsebäck's first boiling water reactor was put into use in 1975 the second one in 1977. They were taken out of service in 1999 and 2005, respectively.

Studies of the above-mentioned activation products from the Barsebäck nuclear power plant have provided new information on flow conditions, and transport times in coastal waters and uptake mechanisms in algae as well as in crustaceans living among the algae (Mattsson et al., 1980a, 1980b; Nilsson and Mattsson 1980; Nilsson et al., 1981).

In a recent publication (Eriksson Stenström and Mattsson, 2022), spatial and temporal variations of ^{14}C in *Fucus* spp. were reported. The study included ^{14}C analyses in brown algae collected at 45 sites along the Swedish coast in 2020 (project SSM 2019-5225, P.I. Kristina Eriksson Stenström). The ^{14}C levels in *Fucus* spp. were significantly higher on the west coast than on the east coast of Sweden. Additionally, the ^{14}C specific activity in *Fucus* increased towards the north of the Swedish west coast, indicating contributions from foreign nuclear facilities through inflow of contaminated water from the North Sea. *Fucus* collected close to the Ringhals nuclear power plant (NPP) on the west coast displayed the highest ^{14}C specific activity in the study, demonstrating a clear influence from ^{14}C discharged from this NPP. Additional gamma spectrometric measurements and ICP-MS analysis of the samples are reported in Eriksson Stenström and Mattsson (2021). The paper by Eriksson Stenström and Mattsson (2022) also reports on ^{14}C analyses from the Särödal series: these results are summarized below.

3. Objectives

The aim of the project was:

- 1) To continue sampling and analysis of the brown algae *Fucus serratus* and *F. vesiculosus* at the sampling place on the Swedish west coast where sampling started already in 1967.
- 2) To analyse new radionuclides in the new as well as in the historical material (biobank).
- 3) To build up capacity for analysis of $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am at the radiochemistry lab at Medical Radiation Physics in Malmö, Lund University.
- 4) To intensify analysis of ^{14}C .
- 5) To investigate the temporal variation of ^{129}I .
- 6) To investigate other radionuclides of interest.
- 7) To describe the long-time changes as well as seasonal variations of the activity concentration of different radionuclides in *Fucus serratus* and *F. vesiculosus*.
- 8) To use the new and historical material for studies of the long-term variation of stable elements, like heavy metals.
- 9) To preserve the material in the biobank for future studies.

4. Material and methods

4.1. Sample collection and preparation

Samples of *Fucus serratus* and *F. vesiculosus* have been collected at the same place and by the same person (SM) since 1967 (Figures 2 and 3). The samples were taken from a water depth of 0.3-0.9 m and were all stuck on rocks or bottom stones. The salinity of the water varied from 15 to 23 ‰ due to varying mixing between saline-poor Baltic Sea water and saline-rich North Sea water. The salinity was highest in January-March. At least 10 individual plants were collected, usually many more. Individuals without superficial growth of other algae were selected. After run off and drying in air, the samples were dried in an oven at 70 °C, ground into a fine grain and homogenized. The dry-weight to wet-weight (at sampling) ratio was normally between 24 and 27%.

Starting in 2020, additional samples of *Fucus* as well as of sea water were taken for ³H analysis at Särö and other places along the Swedish west-coast (see section 4.2.2).

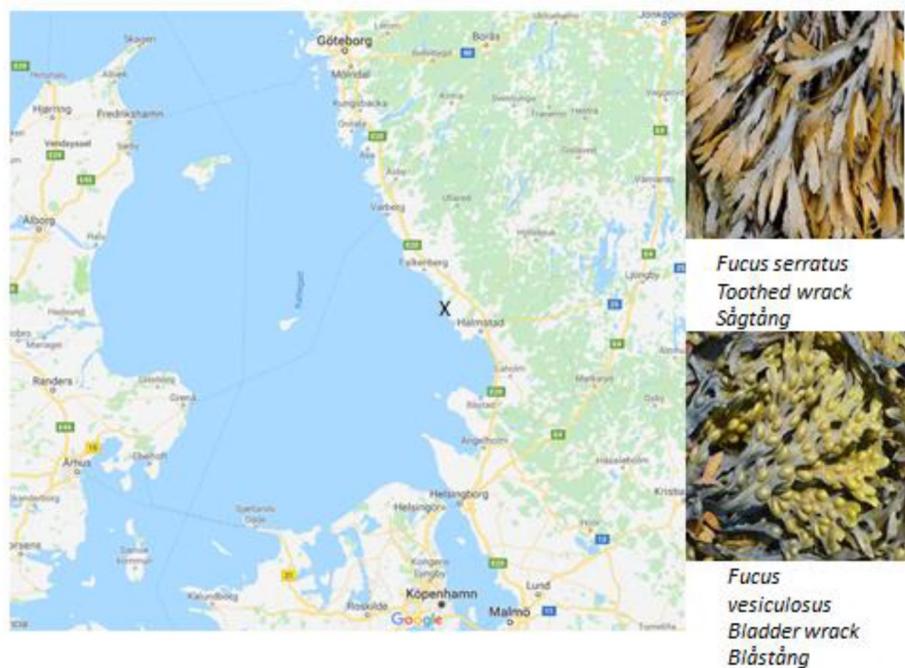


Figure 2: Map indicating the sampling place (X) at Särö (56.76N, 12.63E) and pictures of the two bioindicators used with their Latin, English and Swedish names.

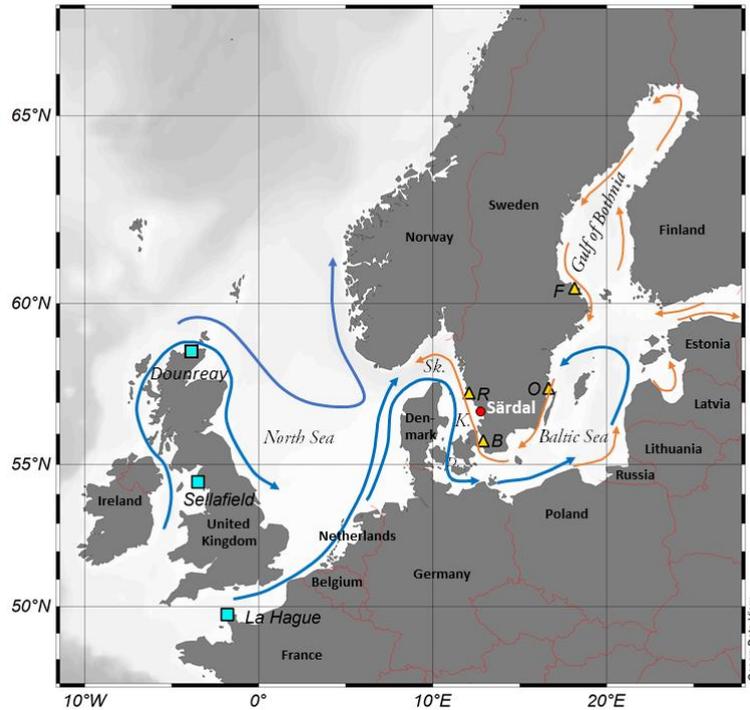


Figure 3: The sampling site (Särda) and the Swedish west coast in a larger perspective. Main transport routes from Sellafield, Dounreay and La Hague, respectively, are indicated. The Swedish nuclear power plants are also shown (R: Ringhals; B: Barsebäck; O: Oskarshamn; F: Forsmark). Sk. – Skagerrak; K. – Kattegat. Arrows represent surface water movement in orange and bottom water movement in blue. Maps: Schlitzer, Reiner, Ocean Data View, <https://odv.awi.de>, 2022.

Marine aerosols are generated at the water–atmosphere interface. This may have implications for coastal land areas. The aerosols are dominated by sea-salt particles. It is of interest to investigate whether also radionuclides in the water are spread in this way. The aerosols move inland, but it is not known how far. Therefore, samples of *Xanthoria parietina* (yellow wall lichen; Sw. *gul vägglav*) have been collected at various distances from the water-front in 2017 and 2018 (Figure 4). This lichen is growing on rocks, stones, tiled roofs, walls and tree bark.



Figure 4: *Xanthoria parietina* is a leafy lichen. It can be found near the shore on rocks and walls (*parietina* means "on walls"), and also on inland rocks, walls, tiled roofs and tree bark.

4.2. Analysis of radionuclides

Fractions of most samples collected since 1967 are still available. Due to limitations in analytical capacity, lack of methods etc., all samples are yet not analysed for all radionuclides indicated. This is still possible for those with long physical half-life, but not for those with shorter, e.g. ^{131}I , ^7Be , ^{134}Cs , $^{110\text{m}}\text{Ag}$, and ^{54}Mn . The desirability and possibility of supplementing the analyses performed for the radionuclides ^3H , ^{14}C , ^{99}Tc , ^{90}Sr , ^{129}I , ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am , ^{228}Ra , ^{236}U and ^{238}U so that they cover the entire time series are discussed in the section Outlooks and future activities at the end of this report.

Table 1: Table showing which radionuclides have been analysed so far in the time series of samples of *Fucus* 1967-2021. x – samples still available for analysis, xx – samples measured, but results not evaluated,  – samples fully analysed. F.s and F.v – number of currently available samples of *Fucus serratus* and *F. vesiculosus* respectively.

Year	F. s.	F. v.	^{137}Cs ^{134}Cs	^{40}K	$^{60,58}\text{Co}$ ^{65}Zn ^{54}Mn $^{110\text{m}}\text{Ag}$	^3H	^{14}C	^{99}Tc	^{90}Sr	^{129}I	$^{239}\text{Pu}+$ ^{240}Pu	^{239}Pu	^{238}Pu	^{241}Am	^{228}Ra ^{228}Ac	^7Be
1967	1	1							x							x
1970	4	4					x		x	x		x	x			x
1972	1	1					x		x	x		x	x			x
1973	1	1					x		x	x		x	x			x
1974	1	1					x		x	x		x	x			x
1975	1	1					x		x	x		x	x			x
1976	2	2					x		x	x		x	x			x
1977	3	2					x		x	x		x	x			x
1978	3	3					x		x	x						x
1979	3	3					x		x	x		x	x			x
1980	6	6					x		x	x		x	x	x		x
1981	4	4					x		x	x		x	x			x
1982	4	3					x		x	x		x	x	x		x
1983	1	1					x		x	x		x	x	x		x
1984	0	0							x			x	x	x		x
1985	2	4					x		x	x		x	x	x		xx
1986	8	3					x		x	x		x	x	x		xx
1987	4	2					x		x	x		x	x	x		xx
1988	4	5							x	x		x	x	x		x
1989	3	1					x		x	x		x	x	x		x
1990	4	4					x		x	x		x	x	x		xx
1991	6	5					x		x	x		x	x	x		xx
1992	6	4							x			x	x	x		xx
1993	6	2					x		x	x		x	x	x		xx
1994	6	4							x	x		x	x	x		xx
1995	5	5					x		x	x		x	x	x		xx
1996	6	3					x		x	x		x	x	x		xx
1997	5	1					x		x	x		x	x	x		xx
1998	5	3					x		x	x		x	x	x		xx
1999	6	4					x		x	x		x	x	x		xx
2000	5	7					x		x	x		x	x	x		xx
2001	10	8							x	x		x	x	x		xx
2002	7	6							x	x		x	x	x		xx
2003	6	7					x	x	x		x	x	x	x		xx
2004	3	3					x		x	x		x	x	x		xx
2005	6	6							x	x		x	x	x		xx
2006	8	5							x	x		x	x	x		xx
2007	6	5					x		x	x		x	x	x		xx
2008	10	9							x	x		x	x	x		xx
2009	9	9							x	x		x	x	x		xx
2010	8	8					x		x	x		x	x	x		xx
2011	7	7							x	x		x	x	x		xx
2012	8	8					x	x	x			x	x	x		xx
2013	8	5					x	x	x		x	x	x	x		xx
2014	8	7						x	x		x	x	x	x		xx
2015	11	6						x	x		x	x	x	x		xx
2016	7	7						x	x		x	x	x	x		xx
2017	6	6						x	x		x	x	x	x		x
2018	8	7						x	x		x	x	x	x		x
2019	6	6						x	x		x	x	x	x		x
2020	7	7						x	x	x		x	x	x		x
2021	7	7					x	x	x	x		x	x	x		x

4.2.1. Analysis of ^{137}Cs , ^{134}Cs , ^{40}K , ^7Be , ^{60}Co , ^{228}Ra , ^{131}I and other γ -emitters

For the assessment of the radionuclide concentration in the various samples, different HPGe-gamma spectrometers inserted into lead-shielded cavities were used. In the current project a detector (ORTEC, USA) with the relative efficiency of 55% has been used for most of the measurements during 2018-2021.

The recorded pulse height distributions were evaluated using an in-house evaluation sheet (cross-checked with the detector software evaluation program, Gamma Vision 7.1™) in order to have control of all the steps in the evaluation of each individual gamma line.

4.2.2. Analysis of ^3H

Samples of *Fucus serratus* and *F. vesiculosus* as well as of sea water were collected at Särö and other places along the Swedish west coast from September 2020 to March 2022 to determine their tritium content. Several hundreds of grams of seaweed samples were collected, frozen and stored in sealed plastic bags at -18°C . The frozen samples were freeze-dried at the laboratory in Malmö for 8 hours and the extracted water was collected in plastic bottles. The sea water samples were analysed directly without preparation.

The water samples were split in two to duplicate the measurements, 10 ml of water were mixed with 10 ml of Ultima Gold LLT scintillation cocktail (Perkin Elmer) in a 20 ml polyethylene vial. Samples were then shaken for 2 min and stored in the dark for 48 hours to decrease chemical quenching before measurement by liquid scintillation counting (LSC). Each duplicate was then measured in a Beckman LS 6500 multi-purpose LSC during a total of 10 hours (5·2h) and evaluated using a dedicated user programme (Pédhontaa-Hiaa et al., 2020).

Samples of deep well water (Grevie-Bulltofta verket, VA Syd) with a well-documented low tritium concentration were used as background. Dilutions of tritiated water samples with certified values (from the inter-comparison exercise Procorad, 2019) were used as control in the tritium measurements. A quenching curve was also obtained using the method described by the cocktail provider Perkin Elmer (Thomson, 2014).

4.2.3. Analysis of ^{14}C

For a selection of samples, a few mg of dried and grained sample material was submitted to the Radiocarbon Dating Laboratory at Lund University. Prior to ^{14}C -measurement, 1-2 mg carbon was extracted from each sample (>2 mg d. wt.) using the graphitization system AGE at the Radiocarbon Dating Laboratory at Lund University (Wacker et al., 2010; Adolphi et al., 2017). After extraction, the carbon from each sample was pressed into separate Al sample holders, which were taken further to ^{14}C analysis in the Single Stage Accelerator Mass Spectrometry (SSAMS) facility at Lund University (Skog, 2007). The precision of the measurements was <1% (Skog et al., 2010), given by measurement of several standard samples of known ^{14}C content (IAEA C7, SRM 4990B and SRM 4990C). The background was assessed by the measurement of ^{14}C -free

samples. The raw data was analysed by the Radiocarbon Dating Laboratory. Uncertainties of the measurements were reported at one standard deviation. A more detailed description of the ^{14}C measurements is given e.g. in Bernhardsson et al. (2018). For some samples, $\delta^{13}\text{C}$ was measured by Elemental Analyser - Isotope Ratio Mass Spectrometry (EA-IRMS) at SLU Stable Isotope Laboratory, SSIL, at the Swedish University of Agricultural Sciences in Umeå.

The results of the ^{14}C analysis are primarily given as Fraction Modern, $F^{14}\text{C}$ (see Reimer et al. (2004) and Eriksson Stenström et al. (2011) for definition). The relation between specific activity of carbon, $\frac{A}{m_c}$, at the year of measurement, y , and $F^{14}\text{C}$ is given by:

$$\frac{A}{m_c} = F^{14}\text{C} \cdot \left(\frac{1 + \frac{\delta^{13}\text{C}}{1000}}{0.975} \right)^2 \cdot e^{\frac{(1950-y)}{8267}} \cdot 226 \frac{\text{Bq}}{\text{kg C}}$$

where $\delta^{13}\text{C}$ is the isotope fractionation of the sample (in ‰). Isotope fractionation occurs when isotopes of the same element are transferred in chemical or physical processes in nature: the mass difference between the isotopes leads to a discrimination of some isotopes compared to others, resulting in small changes in the isotopic ratios. The specific activity of carbon in dissolved inorganic carbon (DIC) in a water body, which is transferred to *Fucus* in the same water body, will not result in the exact same specific activity in *Fucus* as in DIC. One advantage of expressing high precision measurements of ^{14}C as $F^{14}\text{C}$ instead of specific activity of carbon ($\frac{A}{m_c}$) is that $F^{14}\text{C}$ eliminates the effect of isotope fractionation, making various environmental sample types in the same environmental reservoir directly comparable. Typical values of $\delta^{13}\text{C}$ range from about 0‰ for marine carbonates, -9‰ for atmospheric CO_2 , -15‰ for marine organisms and -27‰ for tree leaves (see Table B4 in Bernhardsson et al. (2018) and references therein). Furthermore, $F^{14}\text{C}$ is independent of the year of measurement, whereas specific activity is not. As an example, for $F^{14}\text{C} = 1.000$, $y = 2020$ and $\delta^{13}\text{C} = -16.00\text{‰}$ (relevant for *Fucus*) the specific activity of carbon becomes: $\frac{A}{m_c} = 228.3 \text{ Bq (kg C)}^{-1}$.

4.2.4. Analysis of ^{99}Tc

The samples were analysed at Department of Medical Radiation Physics in Malmö and prepared according to the method described by Lindahl et al. (2003). Three to five gram d.wt. of the samples were ashed at 550 °C after addition of oxalic acid until all the carbon was removed. About 20 kBq of $^{99\text{m}}\text{Tc}$ -pertechnetate was added as a yield determinant. The ash was dissolved in hydrogen chloride acid and Tc was oxidized to +VII oxidation state with potassium persulphate and hydrogen peroxide. The samples were transformed to 5 M sulphuric acid and Tc was extracted with tributylphosphate and hydrogen fluoride. The technetium was then back-extracted with 2 M sodium hydroxide solution and xylene, electroplated on stainless steel discs, and counted by an anticoincidence shielded GM-counter. The radiochemical recovery was determined by counting the activity of $^{99\text{m}}\text{Tc}$ ($E_\gamma = 141 \text{ keV}$) with a HPGe-detector and was normally between 20 and 70%.

4.2.5. Analysis of ^{129}I

The samples were analysed at Centro Nacional de Aceleradores (CAN), University of Seville, Spain. The sample preparation method used for algae and lichen is described in Schmidt et al. (1998). A small amount of sample (typically between 0.1 and 0.2 g) was mixed with six grams of sodium hydroxide and 4 to 7 mg of iodide carrier prepared from Woodward iodine in 1 ml of water in a nickel crucible. The crucible was covered, and alkaline leaching was carried out for 1 h at 150 °C, then for 2 h at 200 °C followed by 3 h at 275 °C. The melt was then extracted by water and centrifuged. To reduce all iodine species to iodide, N_2H_4 was added to the supernatant. The resulting solution was centrifuged again. Then NaNO_2 was added to the supernatant, acidified with HNO_3 and iodine was extracted into chloroform in a separatory funnel. After reduction by N_2H_4 , iodide was back-extracted into water as previously described. The purification by extraction and back-extraction was carried out two more times. Finally, AgI was precipitated by adding AgNO_3 solution, pressed on a copper cathode and measured by Accelerator Mass Spectrometry (AMS) at Centro Nacional de Aceleradores (CAN), University of Seville, Spain. I^- ions were extracted from the sample in a Cs sputter ion source and injected in a 1 MV Tandemron accelerator (High Voltage Engineering Europe, the Netherlands). After passing through a He stripper, I^{2+} was selected in a magnetic deflector. While stable $^{127}\text{I}^{2+}$ was measured as a current in a Faraday cup, $^{129}\text{I}^{2+}$ went through an electrostatic deflector and was detected in an ionization chamber. Standard samples with known $^{129}\text{I}/^{127}\text{I}$ -ratio were continuously measured to correct the isotopic ratio, as well as blank samples (Woodward iodine) for background evaluation. In this work, instrumental background was typically $^{129}\text{I}/^{127}\text{I} = 1\text{-}2 \times 10^{-13}$, while samples showed ratios at least two orders of magnitude higher (López-Gutiérrez et al., 2000).

4.2.6. Analysis of $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am

One of the important goals of this project was to restore and further build up the radiochemical capacity to analyse plutonium and other actinides in *Fucus* and in other parts of the environment. This has taken longer than expected, but we have now come up with a tested sample preparation and analysis method which is described below.

Method development

The radiochemistry laboratory at Medical Radiation Physics Malmö is currently establishing a routine method for the extraction and the analysis of $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am in *Fucus* samples. Such methods have been developed based on a method previously used for soil samples and peat bog samples (Sáez Muñoz, 2016).

In addition to the preparation of those methods and the purchase of equipment, a Master student was recruited for a Master thesis work on the subject for a year starting in September 2019. The results of this work were compiled in a thesis entitled “Development of a separation method using UTEVA and DGA resins for the quantification of plutonium and americium in brown algae by alpha spectrometry” (Norberg, 2020). The main tasks and achievements of the Master’s project were:

- Large volumes of *Fucus* were collected in prevision of the numerous tests required during the method development.
- Operating parameters (temperature and time) of the ashing of seaweed samples were optimised.
- The leaching method was adapted from the soil sample to fit vegetation samples, the possibility of microwave assisted digestion was also investigated.
- Regarding chemical separation, UTEVA and DGA extraction resins were bought from Triskem and used for method development. The method for the extraction and separation of Pu using UTEVA resin is now fully developed and can provide reliable results while the separation of Am on DGA resin is still under development.
- The classically used Hallstadius electrodeposition method was tested, compared and optimised for plutonium and americium (Hallstadius, 1984) as well as the alternative methods developed by Krmpotic et al. (2017, 2018); similar test could be done for additional radionuclides in the future. The electrodeposition method was used to prepare alpha source of the extracted samples but also to characterise various tracer solutions (see Figure 5).
- Alpha detectors were characterised: background measurements, calculation of the limits of detection for this instrument, energy calibration, determination of their efficiency, etc. Tools for data analysis were developed.

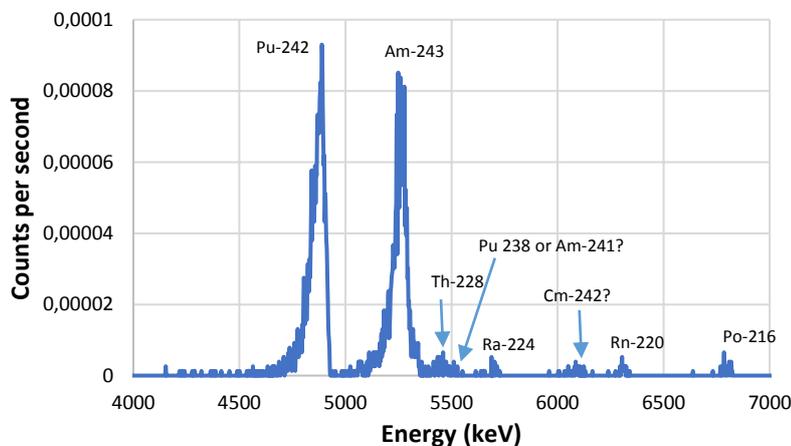


Figure 5: Example of an alpha spectrum: ^{242}Pu tracer solution contaminated with ^{243}Am .

Sample preparation and analysis method

Ashing: Samples of ground *Fucus* are put in ceramic crucibles (5 to 20 g per crucible) and covered with aluminium foil pierced with holes. The crucibles are placed in an oven and heated up to 550 °C with a temperature ramp of 2 °C/min then heated at 550 °C for 10 hours.

Acid leaching: After ashing, 5 g of sample are spiked with a known activity of ^{242}Pu tracer, ^{243}Am tracer and ^{232}U tracer (in case uranium content is also going to be analysed). The ashes are dissolved in 90 mL *aqua regia* (conc. HNO_3 and conc. HCl , ratio 1:3) and stirred for at least 4 h at 50 °C. Then, 24 mL of H_2O_2

are added drop by drop while stirring the mixture. After the end of the effervescence from the decomposition of H_2O_2 , 90 mL of conc. HNO_3 are added and left under stirring overnight without heat. The next day, 60 mL of HNO_3 (8 M) are added to the mixture which is filtered twice under vacuum over a Büchner funnel on a 45 μm cellulose filter. The filtrate is finally evaporated at 100 °C down to approximately 50 mL.

Iron co-precipitation: The remaining slurry is diluted with ultrapure water to approximately 100 mL. 30 mg $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ is added, and the solution stirred for a few minutes. The beaker is then cooled over ice while the pH is slowly adjusted to 8-9 with aqueous NH_4OH (32%) to precipitate the iron as FeOH_3 . The suspension is centrifuged at 3200 RPM for 20 minutes. The supernatant is decanted, and the precipitate is collected. The supernatant is then acidified and the iron co-precipitation and centrifugation steps are repeated. The precipitates are combined and dried at 70 °C for 2–4 hours.

Valence adjustment: The dried precipitate is dissolved in 10 mL hot HNO_3 (8 M). The warm solution is hot-filtered by syringe through a glass microfibre filter (Whatman GF/A, 25 mm) into a clean 25 mL beaker followed by a rinse of 1 mL HNO_3 (8 M). Then, 0.05 g NaNO_2 is added to the sample solution. The mixture is then stirred and heated for 30 minutes. NaNO_2 rapidly oxidises Pu^{3+} to Pu^{4+} and slowly reduces Pu^{6+} to Pu^{4+} (Maillard et al., 2018). The NaNO_2 is totally decomposed when the solution loses its yellow colour and stops producing orange fumes.

Separation on columns (UTEVA): The UTEVA column is prepared for separation according to the instructions from the supplier and conditioned with 5 mL of 8 M HNO_3 eluted by gravity or on a vacuum box connected to a pump at a rate of 1 mL min^{-1} (the same flow rate is applied to the rest of the procedure).

The sample solution is poured through the column. The beaker or bottle containing the sample is rinsed with 10 mL of 8 M HNO_3 and poured into the columns. The UTEVA resin retains Th, U and Pu while Am is eluted.

Pu is eluted from the UTEVA column with 30 mL of a solution of Rongalite (0.1 M) in 2 M HNO_3 . The elution solution is evaporated to dryness and stored before electrodeposition.

Electrodeposition: The procedure employed for actinide electrodeposition is based on Krmpotic et al. (2017, 2018). The dried samples are re-dissolved in a few mL of HNO_3 and H_2O_2 and then heated to destroy any organic compound that may have leaked from the resin and evaporate to dryness. Then, the samples are dissolved again on the electrodeposition electrolyte composed of ammonium oxalate (0.3 M) in 0.3 M HCl. Unlike the classic Hallstadius (1984) method, this procedure is not pH-dependant.

The electrodeposition cell consists of a spiralled platinum electrode as the anode, and stainless-steel discs of 20 mm diameter as the cathode. Electrode distance from plate is about 0.5-1 cm. The cell is covered with Parafilm to limit evaporation. The electroplating is performed at 0.6 A for 75 min. Finally, the discs are taken out of the cell and rinsed successively with 1% NH_3 , water and acetone and they are analysed once dried.

Alpha-spectrometry: The electroplated discs are analysed by alpha-spectrometry. The discs are placed at 2 mm from the surface barrier silicon detector under vacuum in the detector chamber and measured for 2 to 14 days depending on the count rate of the radionuclides of interest.

The minimum detectable activity (*MDA*) for each isotope on a particular detector is calculated using the following formula from Lee et al. (2008) based on Currie (1968):

$$MDA = \frac{2.71 + 3.29(\mu_B)^{\frac{1}{2}}(t_S)^{\frac{1}{2}}(1 + \frac{t_S}{t_B})^{\frac{1}{2}}}{t_S \cdot E \cdot Y \cdot S}$$

where μ_B is the count rate of the background, t_S and t_B are the counting times of the sample and the background, E is the counting efficiency, Y is the chemical yield and S is the amount of the sample. The estimated *MDA* was 0.1 mBq kg⁻¹ (d.wt.).

4.3. Analysis of stable elements (metals)

4.3.1. Samples analysed

Figure 6 shows which metals have been analysed so far in the time series of samples of *Fucus* 1967-2021.

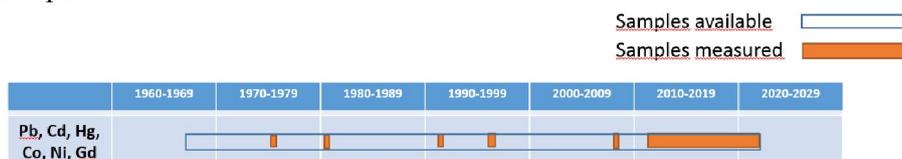


Figure 6: Figure showing which metals have been analysed so far in the time series of samples of *Fucus* 1967-2021.

4.3.2. Analysis method

The most common techniques for the analysis of stable elements are Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and Inductively Couple Plasma Atomic Emission Spectroscopy (ICP-AES). Both techniques have very low detection limits and allow to measure, in a single analysis, a large number of elements. The analysis of stable elements (metals) in the algae, lichen and water samples were carried out at Analytical Services, Department of Geology, Lund University after chemical digestion at Department of Biology, also at Lund University.

Samples were measured on a Bruker Aurora Elite ICP-MS instrument. The following elements and isotopes were measured: ²⁰⁸Pb, ¹¹⁴Cd and ¹¹¹Cd, ⁵⁹Co, ⁶⁰Ni, ²⁰²Hg, ¹⁵⁷Gd, and in some samples also ⁷⁵As and ⁷⁸Se (not reported here). The calculation of the concentration of each element was performed assuming their respective isotopes were present in the sample at their natural abundance.

Samples from a certified reference material (ERM-CD200 Bladderwrack) with specified element concentration of Pb, Cd, Hg, Se, As, Cu and Zn from Unit for Reference materials, EC-Joint Research Center, Institute for Reference Materials

and Methods, Geel, Belgium have been analysed alongside the *Fucus* and *Xanthoria* samples. Agreement within estimated uncertainty intervals were reached for Pb, Cd, Hg, Se and As (Cu and Zn were not analysed).

5. Results and discussion

5.1. Radionuclides in *Fucus*

5.1.1. ^{137}Cs

The time variation of the ^{137}Cs activity concentration in *Fucus serratus* is shown in Figure 7a. The corresponding curve for *F. vesiculosus* is shown in Figure 7b. In the period 1967-1979, most of the ^{137}Cs is assumed to come from the nuclear weapons tests in the atmosphere and to a minor extent from releases from Sellafield. The following unexpected increase, peaking in 1980-81, came 4 years after the peak release of ^{137}Cs from Sellafield. The contribution from Chernobyl is seen as an immediate peak in 1986 and a broader peak around 1990. The following decrease was much slower than after the Sellafield peak indicating a continuous inflow of ^{137}Cs from the Baltic. The Baltic Sea still acts as a secondary source of ^{137}Cs due to outflowing brackish water from the Baltic Sea into the Kattegat. In 2000, the annual net outflow from the Baltic was estimated to 39 TBq (Dahlgard, 2002; Skjerdal et al., 2017). In 2009, HELCOM (2009) estimated the effective half-life of ^{137}Cs in Baltic seawater during the period 1993-2006 to 9.6 years. In the period 1995-2000 the ^{137}Cs concentration in *Fucus* shows – as estimated from Figure 7a – an effective half-life of 10.8 ± 0.9 year. This indicates that the main source of ^{137}Cs in the coastal waters at Särda is dominated by ^{137}Cs from the Baltic Sea. The discharges from Sellafield have decreased in recent years, but contaminated sediments in the Irish Sea from the large discharges in the 1970s and 1980s may have been remobilized and has been estimated to do so for a long time (Cook et al., 1997; Cook et al., 1998; Leonard et al., 1999). In addition to contributions from ^{137}Cs from Chernobyl and Sellafield, the ^{137}Cs -levels in the west coast marine environment are affected by the inflow from la Hague, remaining activity from the nuclear weapons tests in the atmosphere, and run-off from land. These components seem to be small in comparison with the outflow from the Baltic Sea.

The new results regarding ^{137}Cs reported in this project stem from the time period 2008-2021 and have been included in previously presented diagrams of the temporal variation (Figure 7a). For studies of temporal and seasonal variations of ^{137}Cs behavior in *Fucus*, highest precision is reached using the $^{137}\text{Cs}/\text{K}$ -ratio. Currently, the ^{137}Cs concentration and the $^{137}\text{Cs}/\text{K}$ ratio do not change much from year to year: seasonal variation, with a pronounced maximum in summer and minimum in winter, is still clearly measurable (Figure 10).

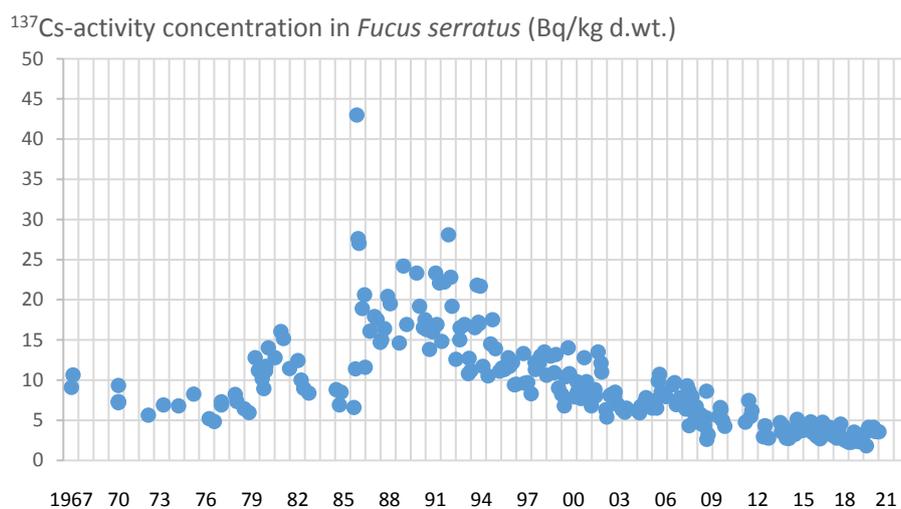


Figure 7a: ¹³⁷Cs-activity concentration (Bq/kg d.wt.) in *Fucus serratus* from Särödal (56.76N,

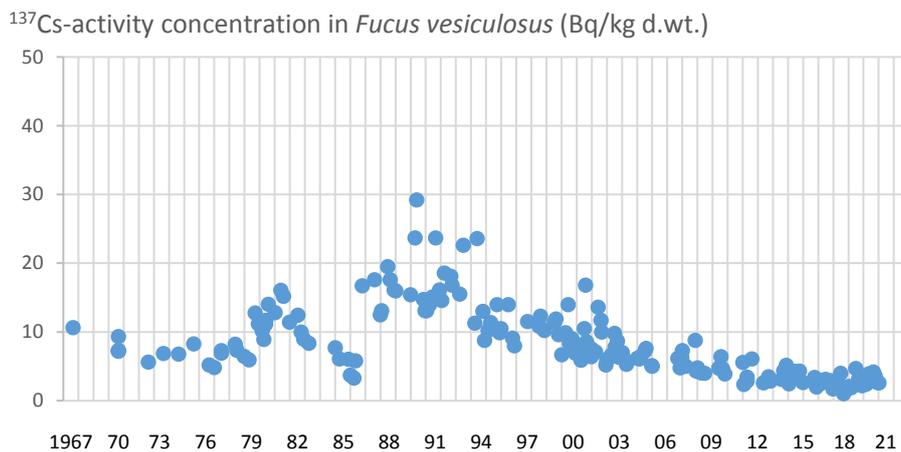


Figure 7b: ¹³⁷Cs-activity concentration (Bq/kg d.wt.) in *Fucus vesiculosus* from Särödal (56.76N,

Assuming a ¹³⁷Cs concentration of 65 Bq/m³ of water in Kattegat in the period 1988-2001 (HELCOM, 2009), the concentration factor can be estimated to 60 for *Fucus serratus* and 50 for *F. vesiculosus*.

5.1.2. ¹³⁴Cs

Measurable amounts of ¹³⁴Cs were registered in the period 1979-1983 which means in connection with the Sellafield peak of ¹³⁷Cs and in 1986-1999 after the Chernobyl fall-out (Figure 8). The ¹³⁴Cs/¹³⁷Cs-activity ratio at max levels at these two occasions were estimated to 5x10⁻³ and 10⁻², respectively.

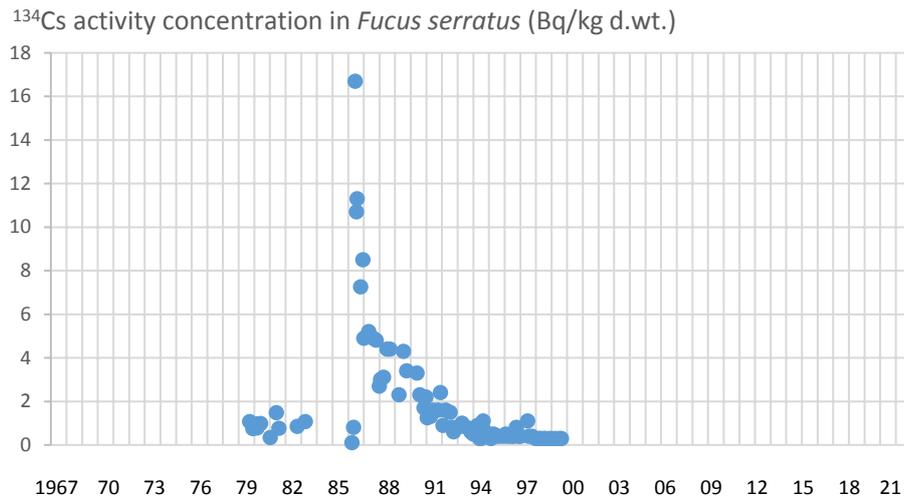


Figure 8: ^{134}Cs -activity concentration (Bq/kg d.wt.) in *Fucus serratus* from Särödal (56.76N, 12.63E) on the Swedish west-coast during the period 1967-2020. For long periods the activity concentration was below the detection limit of 0.1-0.3 Bq/kg d.wt.

5.1.3. ^{40}K and total potassium

Potassium-40 (^{40}K) is a natural radionuclide ($T_{1/2} = 1.27 \cdot 10^9$ year) and constitute today 0.0119% of the total potassium content. Algae have a higher potassium concentration than terrestrial plants. The mean value for *Fucus serratus* from Särödal during the period 1967-2020 was 885 ± 150 Bq/kg d.wt. which corresponds to 29 g potassium per kg of dry seaweed. The results for ^{40}K in the period 1967-2021 have been plotted as a function of sampling month and day in Figure 9. There is no pronounced seasonal variation in the potassium concentration during a year as for ^{137}Cs . There seems however to be a tendency for a minimum in summer (Figure 9).

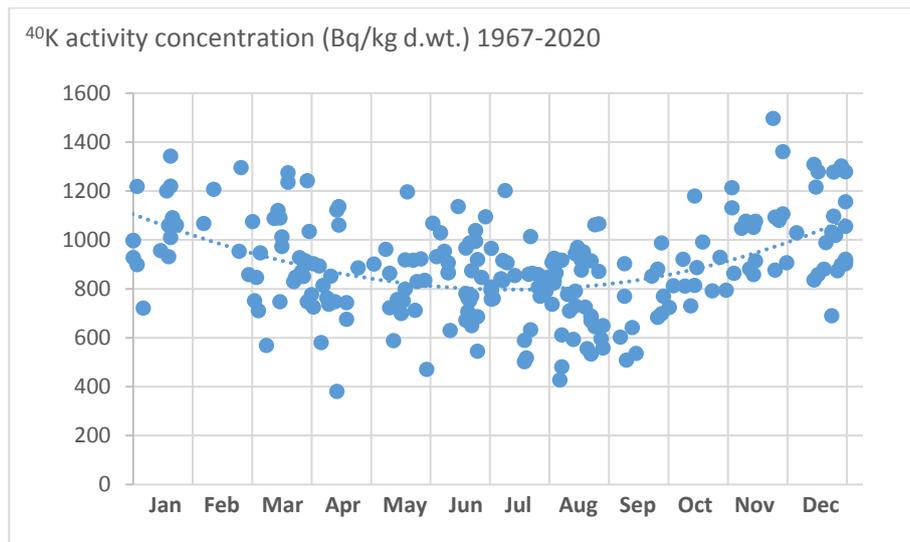


Figure 9: ^{40}K activity concentration in *Fucus serratus* from Särödal (56.76N, 12.63E) in the period 1967-2021, plotted as a function of sampling day during the year. The curve represents a 3rd order polynomial fit. (1 g K has an activity of 30.6 Bq; 1000 Bq/kg d.wt. means 32.7 gK/kg d.wt. or 8 gK/kg w.wt.)

Other authors have reported more pronounced seasonal variations of the potassium concentration in algae (Patti et al., 1990), also with maximum values in winter and minimum values in summer.

Fucus from the Baltic showed a mean ^{40}K activity concentration of 817 ± 33 Bq/kg d.wt. in the period 1978-1983 (Holm et al., 1986). In 2020 a rough mean value for *Fucus* collected along the Swedish coast was 930 Bq/kg d.wt. (Eriksson Stenström et al., 2021).

5.1.4. $^{137}\text{Cs}/^{40}\text{K}$ - ratio

Figure 10 shows the ratio between the ^{137}Cs and the ^{40}K activity concentrations in *Fucus serratus* and *F. vesiculosus* during a recent three-year period with low ^{137}Cs -concentrations. The pronounced seasonal variation of the ^{137}Cs -concentration as well as the $^{137}\text{Cs}/^{40}\text{K}$ -ratio was already seen in the 1970s (Mattsson, 1984; Mattsson and Erlandsson, 1981) and is still easy to register despite the current low ^{137}Cs activity concentration.

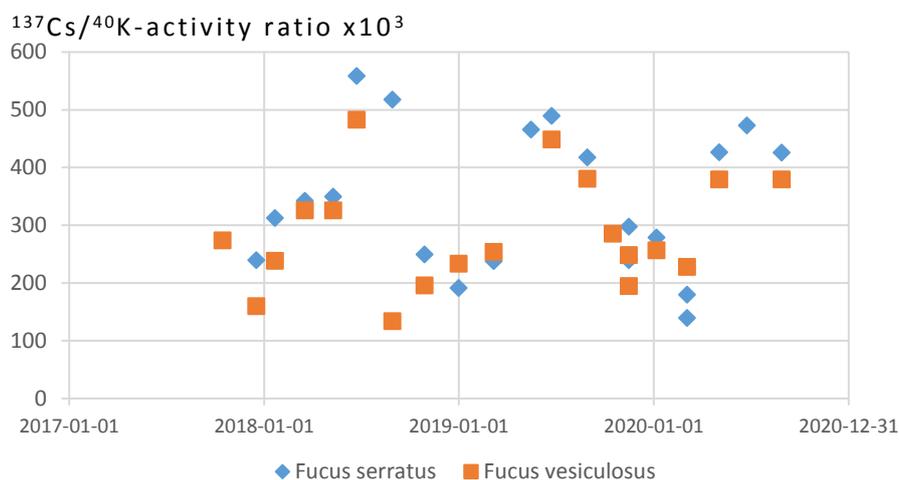


Figure 10: Variations in the activity ratio between ^{137}Cs and ^{40}K in *Fucus serratus* and *F. vesiculosus* from Särödal (56.76 N, 12.63E) from autumn 2017 to autumn 2020. It can be noted that the ratio is generally slightly higher for *F. serratus* than for *F. vesiculosus*.

5.1.5. ^{60}Co

The activity concentration of ^{60}Co in *Fucus serratus* and *F. vesiculosus* is shown in Figures 11a and 11b, respectively. At the same time as ^{60}Co was registered, significant amounts of a number of other neutron activation products were registered (^{58}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$, etc.). These radionuclides have been shown to have their origin in the Barsebäck NPP (Mattsson et al., 1980a). As previously mentioned, Barsebäck's first boiling water reactor was put into use in 1975, the second one in 1977. They were taken out of service in 1999 and 2005, respectively. In the results behind this report, there are no indications of any significant contributions from the Ringhals NPP, which started in February 1969 and still is in use. This is in line with expectations given the dominating northerly coastal currents.

The 1986-12-06 peak in *Fucus serratus* (no *F. vesiculosus* sample was taken at that occasion) comes six months after the ^{137}Cs peak from the Chernobyl fallout and is, like the other ^{60}Co registrations most likely an effect of releases from the Barsebäck NPP.

^{60}Co -activity conc. in *Fucus s.* (Bq/kg d.wt.)

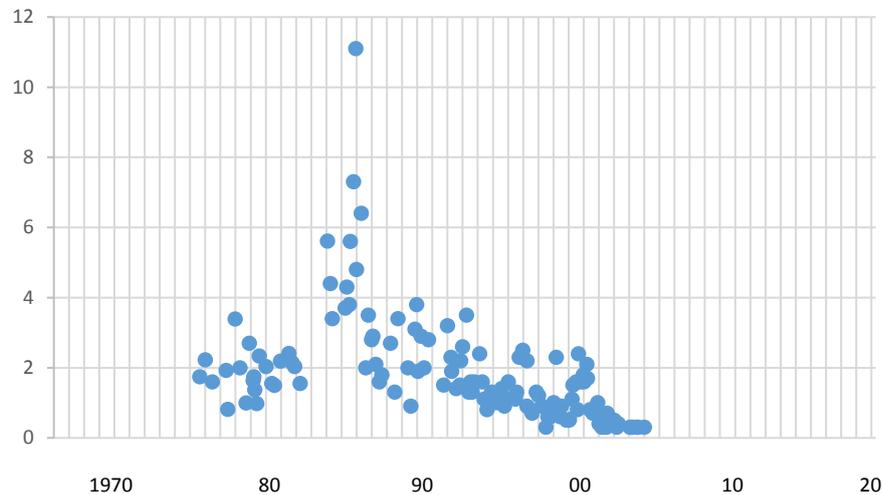


Figure 11a: ^{60}Co -activity concentration in *Fucus serratus* from Särödal (56.76N, 12.63E) 1967-2000. First detected August 21, 1976. Last time for detectable amount December 29, 2005. Minimum detectable activity concentration 0.3 Bq/kg d.wt.

^{60}Co -activity conc. in *Fucus v.* (Bq/kg d.wt.)

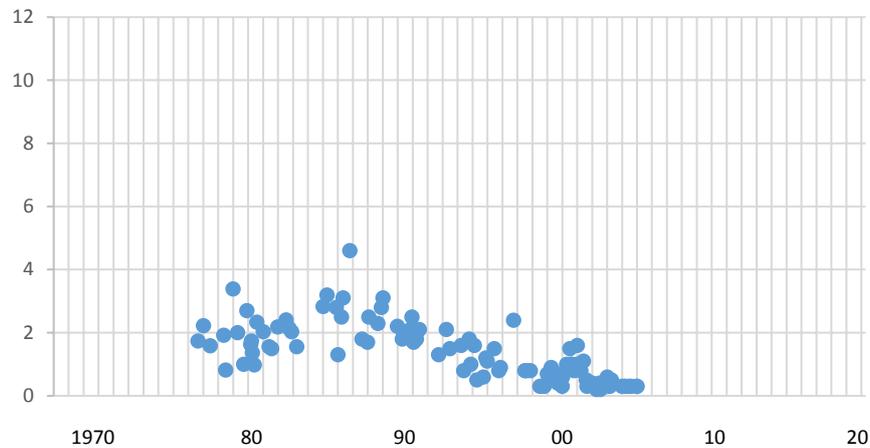


Figure 11b: ^{60}Co -activity concentration in *Fucus vesiculosus* from Särödal (56.76N, 12.63E) 1967-2000.

5.1.6. ^7Be

Beryllium-7 is a gamma emitting (477.6 keV) radionuclide with a physical half-life of 53.4 days. It is mainly produced by spallation reactions through interaction of cosmic rays with molecules of N, O and C in the atmosphere (Lal and Peters, 1967). Once produced, ^7Be rapidly adsorbs onto aerosol particles in the stratosphere and troposphere and is further transported to the Earth's surface by dry and/or wet fallout. An almost linear relation between precipitation and

^7Be deposition has earlier been demonstrated (Ishikawa et al., 1995; Caillet et al., 2001). The yearly deposition in Sweden has been estimated to around 1500 Bq/m^2 .

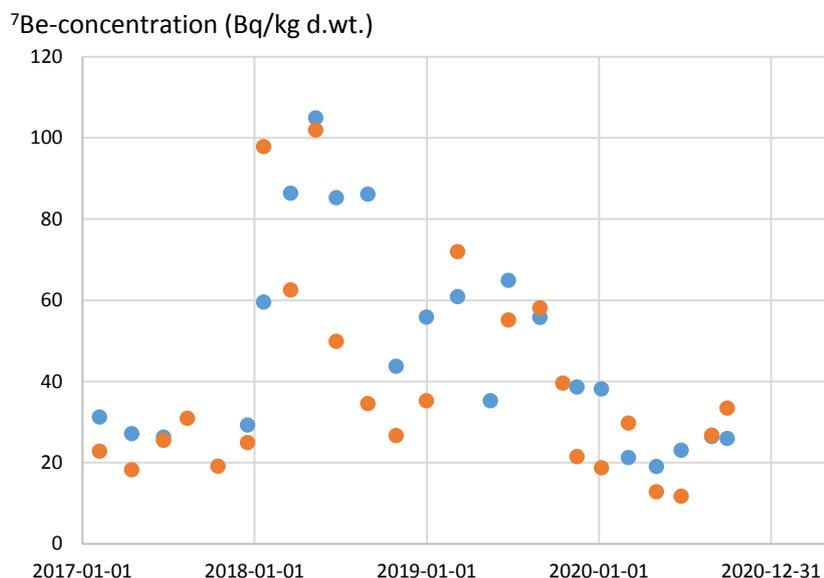


Figure 12: The ^7Be activity concentration (Bq/kg d.wt.) in *Fucus serratus* (blue) and *F. vesiculosus* (orange) from Särðal (56.76N, 12.63E) during the period 2017-2020.

The activity concentration is typically 20-40 Bq/kg d.wt. in *F. serratus* and 10-40 Bq/kg d.wt. in *F. vesiculosus* from Särðal. This was also the case in 2017 (Figure 12). In 2018 however the concentrations were 3 times higher than in 2017 both for *F. serratus* and *F. vesiculosus*. Given the much lower rainfall in 2018 (700 mm) than in 2017 (1100 mm) (see Figure 13), this is a surprising result. Possible explanations could be inflow from areas with higher precipitation or more likely a more efficient uptake of dry deposited ^7Be . Another explanation could be that the ^7Be deposition is more dependent of other meteorological parameters than amount of precipitation e.g. on air temperature, sunshine duration and wind speed (Zalewska et al., 2021).

An interesting observation is that Ringhals 1 and 2 have reported releases of ^7Be ; for the years 2002-2004, 1-50 MBq per year, which is equivalent to the yearly natural deposition on $700\text{-}30\,000 \text{ m}^2$ (3 hektar). Beryllium-7 is available in concentrations of up to about the order of 10 kBq/kg in the reactor water of a PWR (Henning, Å., personal communication, 2021). The main source/production mechanism is $^{10}\text{B}(p,\alpha)^7\text{Be}$. Beryllium-7 is rarely detected in the emission samples and when it is detected, it is in very low concentrations, $<10 \text{ Bq/kg}$ (Henning, Å., personal communication, 2021).

The ^7Be concentration in *Fucus* from Särðal was however significantly lower than in *Fucus* from the station in Ringhals environmental program that is closest to their emissions (Station 3) (Aquilonius, K., personal communication, 2022), where the average value was 150-300 Bq/kg d.wt. between 1995 and 2019. In 2018 it was 189 Bq/kg d.wt. *Fucus* from stations further away from Ringhals showed activity concentrations of 20-80 Bq/kg d.wt. (Aquilonius, K., personal communication, 2022), which is still somewhat higher than the normal concentration at Särðal.

In Barsebäck's environmental control program, ^7Be has also been measured. Between the years 2011 and 2019, the levels have varied between 19 and 150 Bq/kg in *Fucus vesiculosus* with an average of about 60 Bq/kg (Aquilonius, K., personal communication, 2022).

This observation described above motivated further analysis of previously measured samples with respect to ^7Be . The *Fucus* samples measured within three months after collection have been evaluated for their ^7Be content. The result is shown in Figure 15. Since the physical half-life is only 53.4 days, a remeasurement was not possible.

Precipitation (mm/3 months)

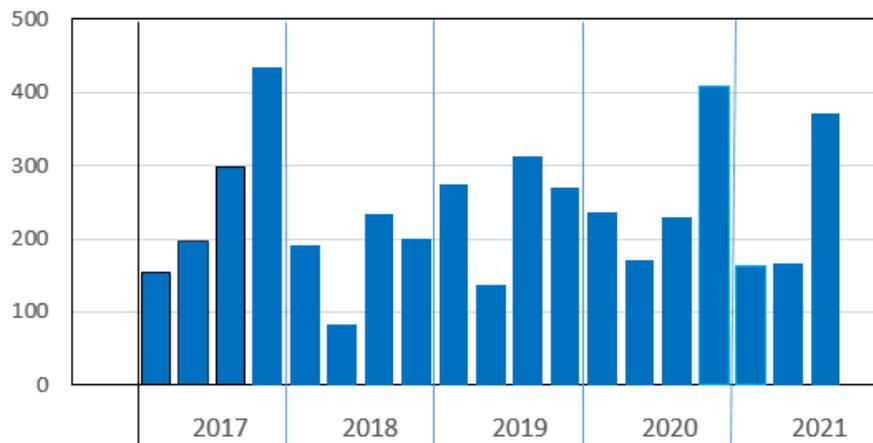


Figure 14: Precipitation (mm/3 months) in Efra, 8 km N Särödal, 2017-2020 (SMHI, 2020).

^7Be -concentration (Bq/kg d.wt.)

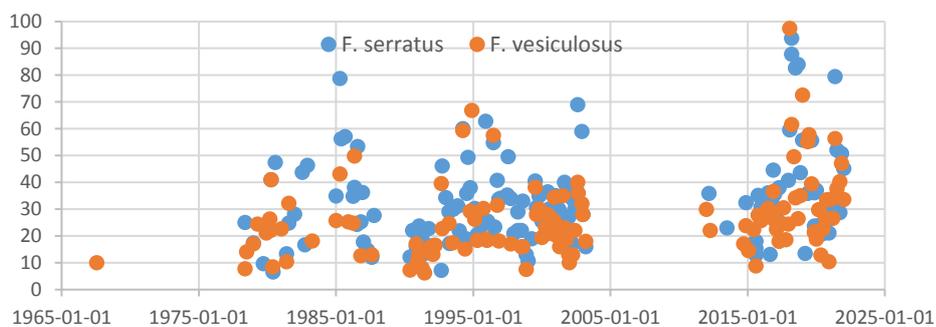


Figure 15. The ^7Be activity concentration (Bq/kg d.wt.) in *Fucus serratus* and *Fucus vesiculosus* from Särödal (56.76N, 12.63E) during the period 1967-2020.

With the exception of the higher levels in 2018, there is nothing that indicates deviations from the normal values during the time period studied.

5.1.7. ^{228}Ra

The content of radium isotopes (^{226}Ra , $T_{1/2} = 1.60 \cdot 10^3$ y; ^{228}Ra , 5.75 y; ^{224}Ra , 3.8 d) in seawater is thought to be mainly due to leaching from coastal sediment and shallow shelves or the mixing of inland waters (Inoue et al., 2005). The

radionuclides occur also i.e. in sludge, deposits and process water from the offshore and coastal oil and gas industry and can also get into the water in connection with dredging and backfilling and may therefore be of interest to follow in the marine environment.

The ^{228}Ra activity was estimated from the full-energy peaks from ^{228}Ac ($T_{1/2} = 6.13$ h) at 338 and 911 keV. Actinium-228 is assumed to be in equilibrium with its mother nuclide ^{228}Ra ($T_{1/2} = 5.75$ year), so the activity concentration of ^{228}Ra is taken as the same as for ^{228}Ac . Radium-228 is a radioactive decay product in the thorium-232 series: $^{232}\text{Th} \rightarrow ^{228}\text{Ra} \rightarrow ^{228}\text{Ac} \rightarrow ^{228}\text{Th} \rightarrow ^{224}\text{Ra} \rightarrow ^{220}\text{Rn} \rightarrow ^{216}\text{Po} \rightarrow ^{212}\text{Pb} \rightarrow ^{212}\text{Bi} \rightarrow ^{208}\text{Tl} \rightarrow ^{208}\text{Pb}$ (stable); with a sub-branch of $^{212}\text{Bi} \rightarrow ^{212}\text{Po} \rightarrow ^{208}\text{Pb}$ (stable). Radium-228 gives rise to many short-lived radionuclides, resulting in a wide range of alpha, beta and gamma radiation. Measured activity concentrations in *Fucus* for the period 2014-2021 is shown in Figure 16. As can be seen in the Figure, there is a clear seasonal variation, somewhat more pronounced for *Fucus serratus* than for *F. vesiculosus*. It would be interesting to investigate whether the variation is due to changes in uptake and retention mechanisms in the algae or reflects the water concentration.

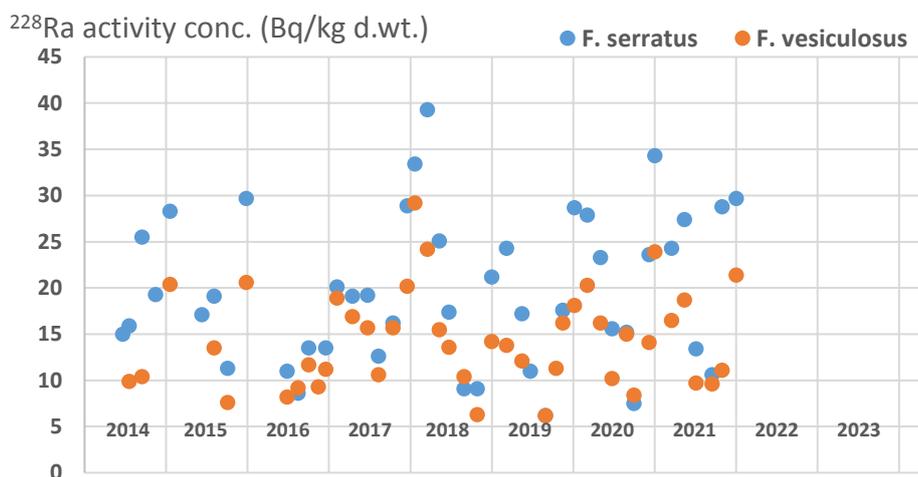


Figure 16: The ^{228}Ra activity concentration (Bq/kg d.wt.) in *Fucus serratus* and *Fucus vesiculosus* from Särda (56.76N, 12.63E) during the period 2014-2020.

It would also be interesting to follow the ^{226}Ra levels in the same samples (Inoue et al., 2005). These measurements should preferably be made by analyses of ^{214}Bi . To enable measurements that way, the sample jars must be gas-tight, and one have to await disintegration equilibrium before measurement so that the ^{226}Ra activity can be evaluated from the two peaks of ^{214}Pb at 352 and 295 keV ($T_{1/2} = 3.8$ d for ^{222}Rn). (The 186 keV peak from ^{226}Ra is often influenced by the nearby peak from ^{235}U).

5.1.8. ^{131}I

Occasionally, ^{131}I is present in detectable amount in the *Fucus* samples. The measured activity concentrations have been very low, typically 0.5-5 Bq/kg d.wt. The most likely explanation is uptake of ^{131}I from discharges of excreta from patients after nuclear medicine therapy with ^{131}I -iodide. Its short half-life

and the delayed discharge via sewers mean that only low activities of ^{131}I reach the marine environment.

5.1.9. ^3H

The sampling sites for the collection of *Fucus* and sea water samples are listed in Table 2. The tritium activity concentration of the water of the *Fucus* samples is presented in Table 3. Most of the values are below the calculated minimum detectable activity concentration (MDA) of 1.65 Bq/L. Only three samples are above this limit with a maximum of 2.48 ± 0.81 Bq/L (Table 3). Sea water samples were also collected on other places on the west coast during the same period. All of them except one present values below the MDA (Table 4). All these values for seaweed and sea water are at the expected environmental level so it can be concluded that the analysed samples did not contain any measurable extra amounts of tritium.

Table 2: Sampling sites for the collection of *Fucus* and sea water samples.

Sea	Site name	Latitude	Longitude
Skagerrak	Båteviken, Strömstad	58.95N	11.13E
	Lysekil	58.27N	11.42E
Kattegat	Hällsvik	57.70N	11.74E
	Smarholmens badplats	57.43N	11.92E
	Frillesås	57.31N	12.15E
	Sallebacka	57.28N	12.14E
	Gloppe	57.27N	12.11E
	Videbergshamn (Ringhals)	57.25N	12.11E
	Bua (Ringhals)	57.24N	12.10E
	Särdal	56.76N	12.63E
	Laxvik	56.60N	12.92E
Öresund Strait	Kattvik	56.46N	12.75E
	Ålabodarna	55.94N	12.78E
	Barsebäcks hamn, Skansen	55.75N	12.90E

Table 3: Tritium activity concentration in water extracted from *Fucus* samples from the Swedish west coast in 2020.

Sample ID	Location	Date	Species	^3H activity concentration (Bq/L)	
M686	Gårda brygga	2020-09-30	<i>Fucus s</i>	0.99 ± 0.76	<MDA
M700	Gloppe	2020-10-01	<i>Fucus v</i>	0.93 ± 0.99	<MDA
M685	Getterön	2020-09-30	<i>Fucus v</i>	1.49 ± 0.98	<MDA
M680	Särdal	2020-09-29	<i>Fucus s</i>	1.67 ± 0.57	
M681	Särdal	2020-09-29	<i>Fucus v</i>	0.70 ± 0.83	<MDA
M732	Särdal	2020-12-06	<i>Fucus s</i>	2.48 ± 0.81	
M714-15	Laxvik	2020-10-10	<i>Fucus</i>	1.50 ± 0.80	<MDA
M718-19	Torekov	2020-10-10	<i>Fucus</i>	2.11 ± 0.76	
M724	Ålabodarna	2010-10-18	<i>Fucus v</i>	1.08 ± 0.58	<MDA

Table 4. Tritium activity concentration in sea water collected on the Swedish west coast in 2020.

Sample ID	Location	Date	Activity concentration (Bq/L)	
SW28	Båteviken	2020-10-15	0.00 ± 0.61	<MDA
SW29	Lysekil	2020-10-15	1.60 ± 0.79	<MDA
SW30	Hällsvik	2020-10-15	1.87 ± 0.99	
SW31	Smarholmen	2020-10-15	0.80 ± 0.78	<MDA
SW8	Frillesås	2020-10-01	0.50 ± 0.78	<MDA
SW9	Sallebacka	2020-10-01	0.37 ± 0.86	<MDA
SW10	Gloppe	2020-10-01	1.56 ± 0.85	<MDA
SW11	Videbergshamn	2020-10-01	1.09 ± 0.63	<MDA
SW12	Bua	2020-10-01	0.00 ± 1.22	<MDA
SW7	Arild	2020-09-30	1.28 ± 0.81	<MDA
SW13	Barsebäck Skansen	2020-10-04	0.00 ± 0.92	<MDA

5.1.10. ^{14}C and $\delta^{13}\text{C}$

The ^{14}C analyses have been extended and 23 new samples within the time interval 1967–2020 have been analysed. For 22 of the samples, $\delta^{13}\text{C}$ was also analysed. Most samples were *Fucus serratus*. One sample of *F. vesiculosus* was divided into two fractions, and one sample was *F. spiralis*. The results are presented as $F^{14}\text{C}$ values in Table 5 (see definition of $F^{14}\text{C}$ in section 2.4.3) and summarized in Figure 17 together with the results of the previously performed analyses of 8 samples. Table 5 also contains the corresponding specific activities of carbon (in Bq/kg C), calculated using the mathematical expressions given in section 4.2.3, as well as activity concentration (in Bq/kg d. wt.). The latter has been calculated using a typical carbon content for *Fucus* of 0.392 g C/g d.wt., obtained from literature (Douville et al., 2004) (the carbon content of the samples of the current study was not measured).

The samples collected in June 2018 were analysed in duplicate at two different occasions: the results overlap within 2 standard deviations. Three samples collected on 3 March 2020 were analysed, and one of the samples were split in three parts. As seen in Table 5, no statistically significant difference can be observed in the different species (*Fucus serratus*, *F. vesiculosus* and *F. spiralis*) or in the different parts of *F. vesiculosus* sample M612.

The ^{14}C results are thoroughly discussed in the paper by Eriksson Stenström and Mattsson (2022). One of the most important findings are, as seen in Figure 17, that all Särö samples taken after 1992 have higher $F^{14}\text{C}$ values than the atmospheric CO_2 curve, whereas $F^{14}\text{C}$ in the samples from 1967 are lower than the atmospheric CO_2 curve. This suggests influence by anthropogenic ^{14}C from activities other than atmospheric nuclear tests in the 1950s and 1960s. The main sources of ^{14}C in Särö are believed to be long-range transport of ^{14}C from the spent nuclear fuel reprocessing plants in La Hague and Sellafield, but also contributions from Ringhals NPP. In the paper by Eriksson Stenström and Mattsson (2022) we make an attempt of source apportionment by using estimated dilution factors for Sellafield/Särö (200) and La Hague/Särö (40) found for other radionuclides at Särö (Lindahl et al., 2003; Dahlgaard et al., 1995). These dilution factors were applied to literature data of environmental ^{14}C concentrations in the marine environment of La Hague and Sellafield. The resulting predicted excess $F^{14}\text{C}$ values at Särö were found to be of the same order of magnitude as the observed increased $F^{14}\text{C}$ values observed at Särö (in comparison to a modelled marine $F^{14}\text{C}$ curve for surface water).

Table 5. $F^{14}C$ and $\delta^{13}C$ in *Fucus* sampled at Särödal between 1967 and 2020. For samples with no measured $\delta^{13}C$, the average $\delta^{13}C$ value of -14.8‰ has been used in the calculation of specific activity. Uncertainty in $F^{14}C$ and specific activity represent one standard deviation. The carbon content in the samples has not been measured. The activity concentration (Bq/kg d.wt.) has been estimated from literature data of carbon content in *Fucus* ($0.392 \text{ g C}/(\text{g d.wt.})$) according to Douville et al. (2004). All data are presented in Eriksson Stenström and Mattsson, 2022), except those from 2001 and 2002 (marked with *), which are presented in Stenström et al. (2006). *F. v.* – *Fucus vesiculosus*; *F. s.* – *Fucus serratus*; *F. sp.* – *Fucus spiralis*.

Date	Sample number		$F^{14}C$	$\delta^{13}C$ (‰)	Specific activity Bq/(kg C)	Activity conc. Bq/(kg d.wt.)
1967-07-15	V759	<i>F. v.</i>	1.407 ± 0.007	-13.5	325 ± 2	127
1967-07-15	V760	<i>F. v.</i>	1.397 ± 0.007	-14.3	322 ± 2	126
1984-08-05	V3763	<i>F. s.</i>	1.201 ± 0.006	-12.5	277 ± 1	109
1988-06-26		<i>F. s.</i>	1.167 ± 0.006	-13.5	269 ± 1	105
1992-01-19		<i>F. s.</i>	1.148 ± 0.006	-13.9	264 ± 1	104
1992-07-04		<i>F. s.</i>	1.147 ± 0.006	-12.5	265 ± 1	104
1994-07-09		<i>F. s.</i>	1.137 ± 0.006	-13.8	262 ± 1	103
*2001-07-03		<i>F. s.</i>	1.132 ± 0.022		260 ± 5	102
*2001-07-07		<i>F. s.</i>	1.113 ± 0.016		255 ± 4	100
*2002-08-07		<i>F. s.</i>	1.090 ± 0.012		250 ± 3	98
*2002-08-08		<i>F. s.</i>	1.124 ± 0.015		258 ± 4	101
2005-12-29		<i>F. s.</i>	1.100 ± 0.006	-15.4	252 ± 1	99
2006-06-25		<i>F. s.</i>	1.104 ± 0.006	-15.1	253 ± 1	99
2008-12-31		<i>F. s.</i>	1.089 ± 0.006	-12.8	251 ± 1	98
2011-06-26		<i>F. s.</i>	1.069 ± 0.005	-14.8	245 ± 1	96
2011-12-30		<i>F. s.</i>	1.067 ± 0.006	-13.9	245 ± 1	96
2014-06-20	M330	<i>F. s.</i>	1.071 ± 0.006	-15.6	245 ± 1	96
2015-01-19	M342	<i>F. s.</i>	1.081 ± 0.006	-14.5	248 ± 1	97
2017-02-06	M509	<i>F. s.</i>	1.072 ± 0.005		245 ± 1	96
2017-06-22	M527	<i>F. s.</i>	1.063 ± 0.006	-14.9	243 ± 1	95
2017-12-17	M537	<i>F. s.</i>	1.051 ± 0.005		241 ± 1	94
2018-06-23	M552	<i>F. s.</i>	1.056 ± 0.005		242 ± 1	95
2018-12-31	M571	<i>F. s.</i>	1.057 ± 0.005	-15.3	242 ± 1	95
2019-06-23	M587	<i>F. s.</i>	1.057 ± 0.005	-16.3	241 ± 1	95
2019-06-23	M587	<i>F. s.</i>	1.046 ± 0.004	-15.6	239 ± 1	94
2020-01-06	M606	<i>F. s.</i>	1.047 ± 0.004	-17.2	238 ± 1	93
2020-03-03	M611	<i>F. s.</i>	1.048 ± 0.004	-16.1	239 ± 1	94
2020-03-03	M612	<i>F. v.</i>	1.047 ± 0.004	-16.4	239 ± 1	94
2020-03-03	M612 ^(x)	<i>F. v.</i>	1.048 ± 0.006		240 ± 1	94
2020-03-03	M612 ^(xx)	<i>F. v.</i>	1.050 ± 0.006		240 ± 1	94
2020-03-03	M613	<i>F. sp.</i>	1.050 ± 0.004	-18.2	239 ± 1	94

^(x) - air bladders and above, ^(xx) - top

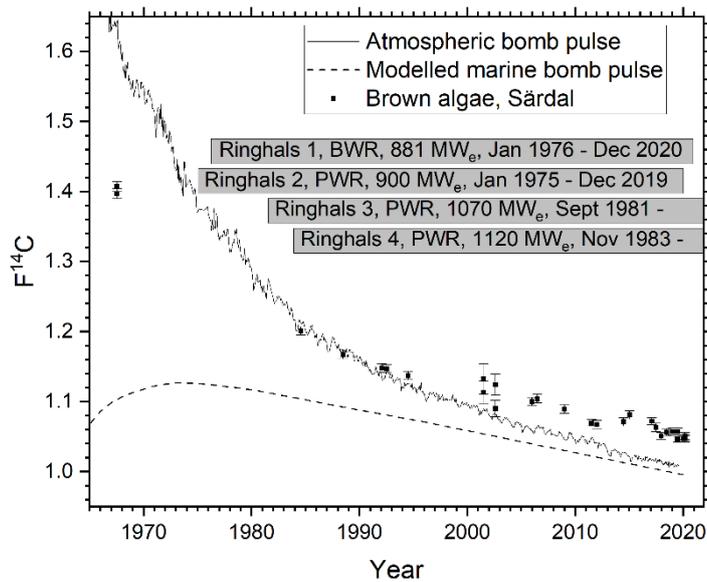


Figure 17: ^{14}C in *Fucus* spp. from SärDAL in the period 1967-2020. The atmospheric bomb pulse representative of CO_2 the Northern Hemisphere is also shown ((Conen et al., 2019); (Hammer and Levin, 2017), (Levin and Kromer, 2004) and (Levin et al., 2013)) as well as a modelled marine bomb pulse representing average global oceanic water down to a depth of 75 m (for values up to 1996 from Reimer et al. (2009); values from 1996 to 2020 are based on linear regression of the data from 1987 to 1996). The periods of operation of the four reactors at Ringhals NPP are also given.

Figure 18 shows annual liquid ^{14}C releases from the nuclear fuel reprocessing plants Sellafield and La Hague (OSPAR, 2012-2018). The possible peak of $F^{14}\text{C}$ in the SärDAL data around 2016 (see Figure 17) may possibly be explained from variations in the releases from Sellafield and La Hague, taking transportation time and the dilution factors from above into account. To fully explore the possible correlation between releases from La Hague and Sellafield the gaps in SärDAL ^{14}C data in Figure 18 needs to be completed by analysis of additional *Fucus* samples (sample availability given in Table 1 in section 4.2).

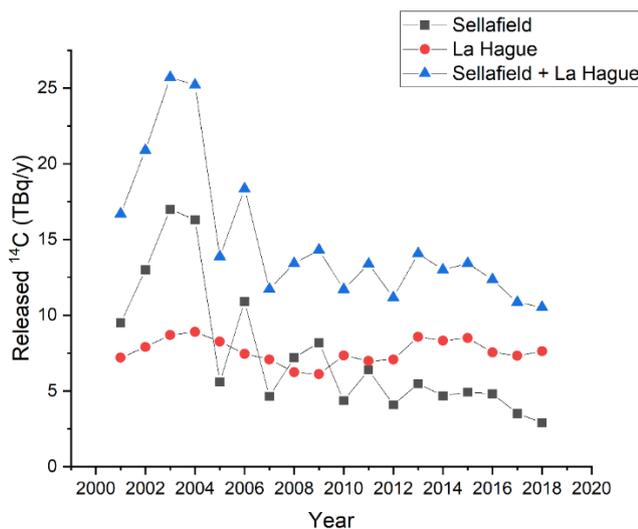


Figure 18: Annual liquid ^{14}C release from Sellafield and La Hague (OSPAR, 2012-2018).

Seasonal variation of the ^{14}C - and ^{13}C -concentration in *Fucus*

Figure 19 displays the $F^{14}\text{C}$ data from the samples collected in 2017-2020. No obvious seasonal trend can be observed, but more samples may be analysed (sample material is available). ^{14}C is transferred to seaweed due to photosynthesis, and Fiévet et al. (2006) estimate that the biological half-life of ^{14}C in *Fucus serratus* is about 5 months. Any seasonal variations of $F^{14}\text{C}$ in seaweed is thus expected to be less pronounced than in the seawater (Fiévet et al., 2006).

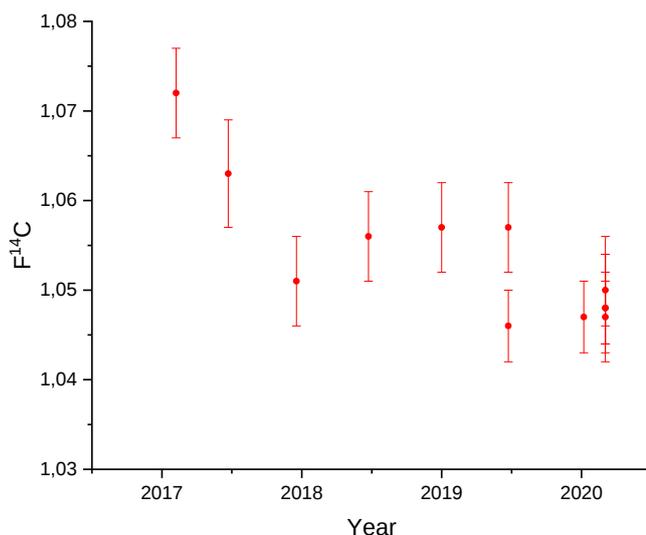


Figure 19: ^{14}C in *Fucus serratus* and *F. vesiculosus* from Särödal in the period 2017-2020 (enlarged from Figure 17).

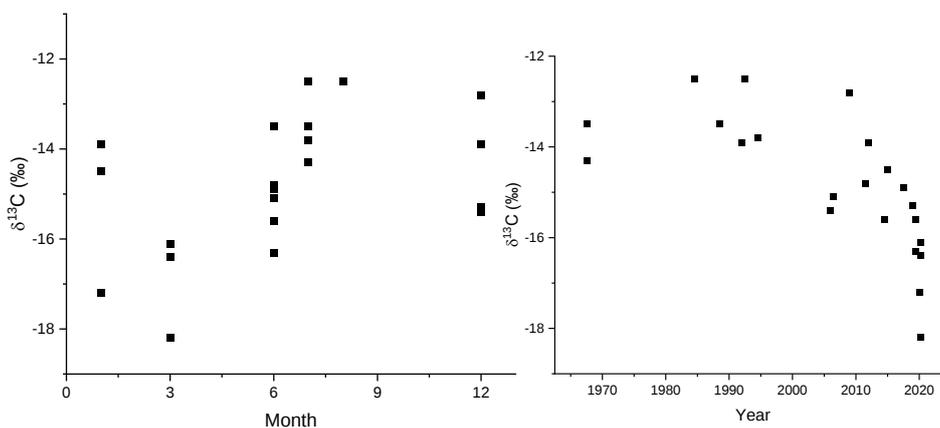


Figure 20: $\delta^{13}\text{C}$ in Särödal seaweed as a function of collection date (left) and collection month for all years (right).

Figure 20 shows all $\delta^{13}\text{C}$ data for the Särödal seaweed samples. $\delta^{13}\text{C}$ values can give information about the uptake rate of inorganic carbon into the algae depending on seasonal variations in environmental factors. Brenchely et al. (1997) found seasonal variations in *Fucus serratus* collected on a monthly basis in NE Scotland for 1.5 years, with higher values in the summer (-13.5 ‰) than

in the winter (-18 ‰). The Särđal $\delta^{13}\text{C}$ values are in the same range and order as those reported in Brencely et al. (1997). Seasonal variations of the hitherto more sparsely analysed Särđal data can however not be clearly distinguished. The reason for the decrease in $\delta^{13}\text{C}$ with time (Figure 20 (right)) calls for further investigations.

5.1.11. ^{99}Tc

^{99}Tc ($T_{1/2} = 210\,000$ year) is produced in nuclear fission with a yield approximately comparable to that of ^{90}Sr and ^{137}Cs , and it has been released into the environment during the last 40 years primarily as a result of nuclear fuel reprocessing. In 1978 as well as in 1995, the discharge of ^{99}Tc from the reprocessing facility at Sellafield in the UK increased sharply (Figure 21). In oxygenated seawater, ^{99}Tc is transported over long distances as highly soluble pertechnetate TcO_4^- . In 2004, the implementation of a new treatment process for spent nuclear fuel led to a reduction in the discharges of 90 %.

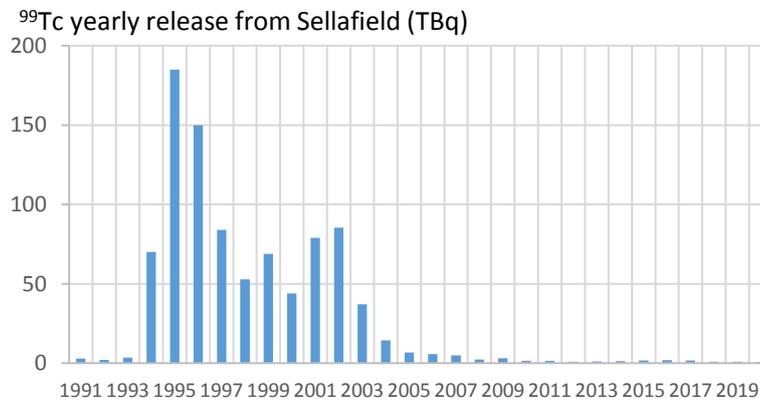


Figure 21: Reported liquid releases of ^{99}Tc from Sellafield 1991-2020 (OSPAR).

The results for ^{99}Tc in *Fucus serratus* from Särđal for the period 1967-2000 have earlier been published (Lindahl et al., 2003) indicating peaks in the activity concentration in *F. serratus* with 4 years' delay in relation to the Sellafield releases as was earlier shown for ^{137}Cs (Figure 22).

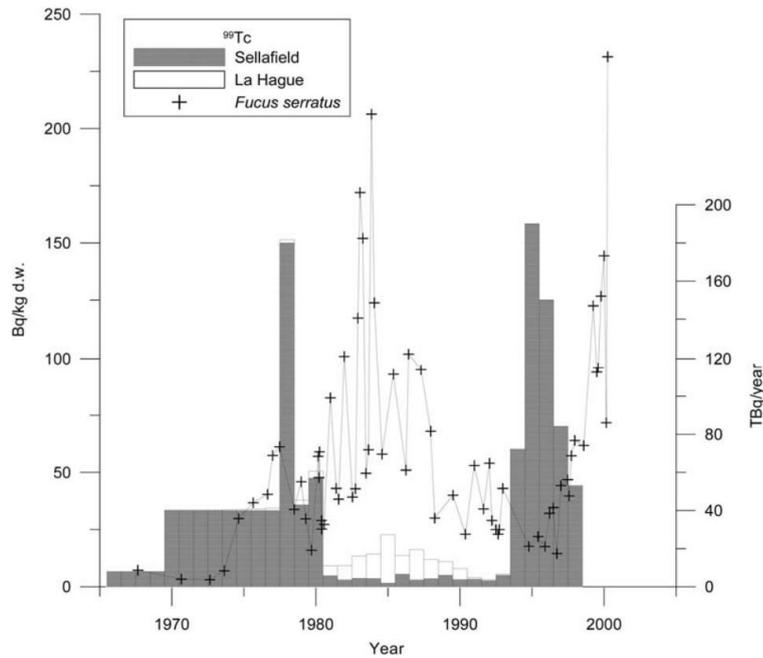


Figure 22: ^{99}Tc activity concentration in *Fucus serratus* from Särödal 1967-2000 and annual ^{99}Tc discharges from Sellafeld (UK) and La Hague (France) reprocessing plants (right y-axis). (From Lindahl et al., 2003).

Since the published results, additional measurements for the period 2000-2010 have been carried out for *Fucus serratus* as well as for *F. vesiculosus* showing decreasing concentrations.

An interesting observation is the very clear difference in ^{99}Tc concentration between *Fucus serratus* and *F. vesiculosus* with an average 68% (19-125%) higher concentration in *F. vesiculosus* (Figure 23).

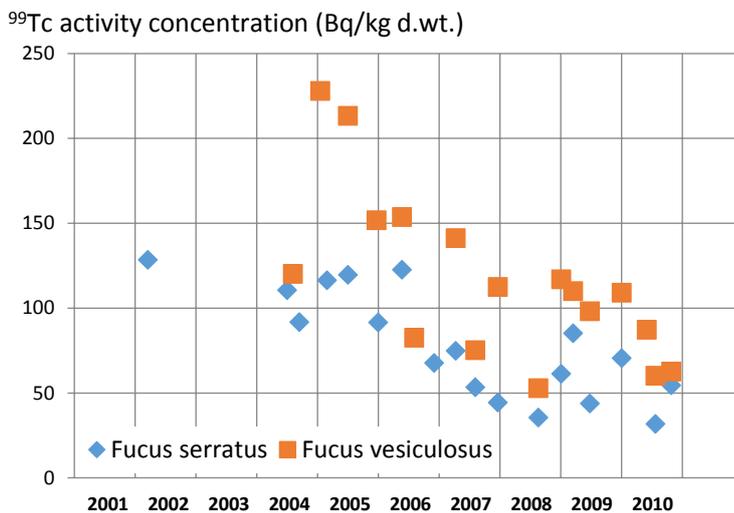


Figure 23: ^{99}Tc in *Fucus serratus* and *F. vesiculosus*, respectively from Särödal during the period 2001-2010.

5.1.12. ^{129}I

Iodine-129 is a long-lived cosmogenic radionuclide ($T_{1/2}=15.7\times 10^6$ year) for which the natural abundances have been altered in a significant way by human actions.

Iodine-129 has also been produced anthropogenically during the last 60 years during fission processes associated with civil and military nuclear activities and accidents, such as Chernobyl (1986). The most significant contribution to the ^{129}I inventory in the environment comes from the liquid and gaseous releases from the two major European nuclear fuel reprocessing facilities, namely Nuclear Decommissioning Authority (NDA)'s plant at Sellafield (UK) and AREVA's plant at Cap de La Hague (France), which after 1985 has been the dominating one (Fréchou and Calmet, 2003; Vivo-Vilches et al., 2018). In spite of trapping systems, ^{129}I (and other radionuclides as ^{85}Kr , ^{14}C , and ^{99}Tc) is partly discharged in the environment as gaseous and liquid radioactive effluents.

Brown algae are known to accumulate iodine from seawater to a remarkable degree with internal levels ranging from 0.05–5% dry weight which constitutes an increase from the concentrations in seawater (near 500 nM) of up to 105 (Küpper and Carrano, 2019)! The ^{129}I concentration in brown algae in relation to that in water has been estimated to 10 000 (Gómez-Guzmán et al., 2011; and references therein). Studies of ^{129}I in water, sediments and seaweed have given important information about water circulation and exchange rates (Vivo-Vilces et al., 2018; Peng et al., 2014; López-Gutiérrez et al., 2004).

The first results related to ^{129}I from this project were reported at the 5th International Conference on Environmental Radioactivity – ENVIRA 2019 in Prague, Czech Republic (Mattsson et al., 2019). Since then, more samples have been analysed and the results up to now are summarised in Figure 24.

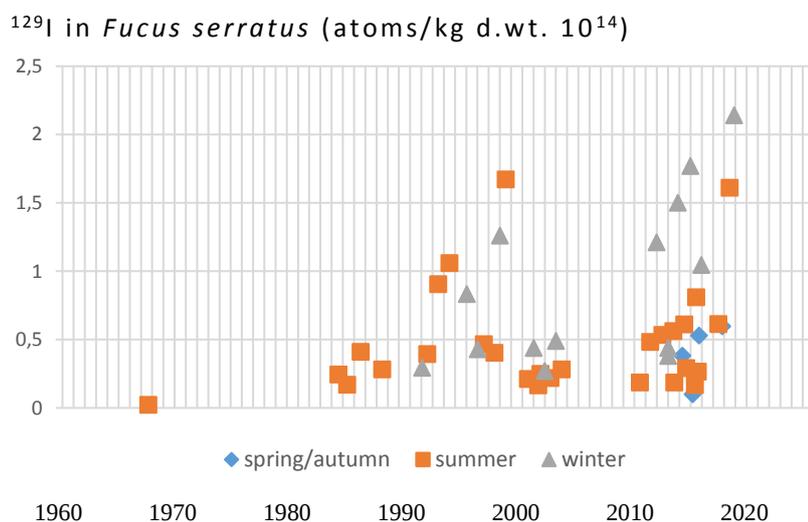


Figure 24: ^{129}I concentration (atoms/kg d.wt.) in *Fucus serratus* from Särda (1 atom = $1.40 \cdot 10^{-15}$ Bq).

The results have to be seen in the light of the reported releases from La Hague and Sellafield (Figure 25) indicating that the main reason for the increase ^{129}I

levels at the Swedish west coast is an effect of the increased releases from La Hague.

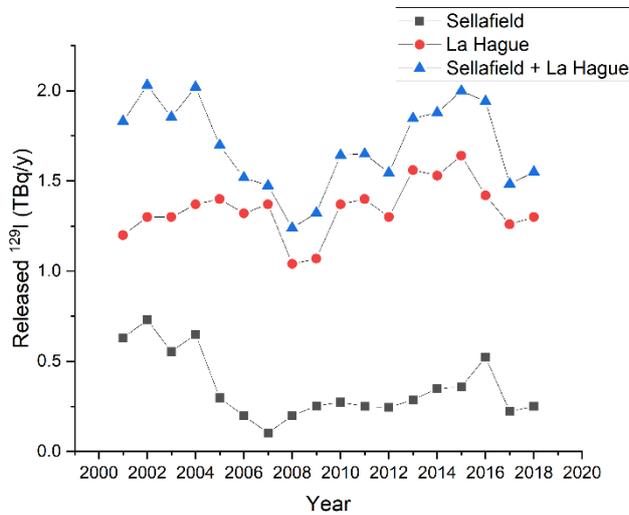


Figure 25: Reported liquid releases of ¹²⁹I from La Hague and Sellafield 2000-2018 (OSPAR).

In 1992-1999, the summer concentration is higher than the concentration in winter. This could be explained by a higher metabolic activity in summertime. On the other hand, in the period 2010-2020, the concentration of ¹²⁹I in *Fucus serratus* in winter is 2-3 times higher than during summer. This is a new observation, which is of importance for careful studies of the time variation of ¹²⁹I in the coastal waters. Further analyses of the correlation between releases and activity concentration in *Fucus* will be done.

¹²⁹I in *Xanthoria parietina*

The question whether ¹²⁹I is transported by air and whether ¹²⁹I is transported from the waterfront up on land is illustrated in Figure 26 below. The estimated inland concentration is marked with a dashed line. Close to the waterfront the levels are about 10 times higher and the distance for measurable influence of sea spray is limited to around a kilometre.

¹²⁹I concentration (atoms/kg dry weight x 10¹³)

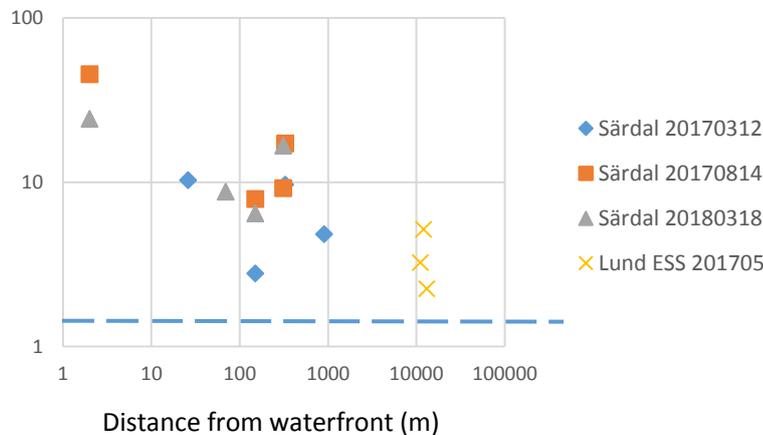


Figure 26: ¹²⁹I concentration in *Xanthoria parietina* collected at different distances from the coastal line (1 atom = 1.40 · 10⁻¹⁵ Bq). Note! Logarithmic scale.

5.1.13. $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am

The method for extraction, separation and analysis of Pu isotopes was repeatedly tested on a *Fucus serratus* sample collected in Särödal on 2018-10-28. The method provides a satisfactory recovery of the Pu and consistent activity concentration. The method was then applied to two additional *F. serratus* samples. An example of spectrum is presented in Figure 27. The measured activity concentrations of $^{239+240}\text{Pu}$ in these samples are presented in Table 6. No signal of ^{238}Pu could be measured on those samples above our detection limit of 0.1 mBq/kg d.wt.

The method development for ^{241}Am was not as successful yet, the obtained results showed a low recovery of the Am tracer added to the sample, thus no ^{241}Am could be detected. Additional work would be necessary to improve this recovery.

Table 6: $^{239+240}\text{Pu}$ activity concentration in dried *Fucus serratus* samples.

Sample ID	Sampling date	Activity concentration in $^{239+240}\text{Pu}$ (mBq/kg d.wt.)	Number of measurements
M26	2012-07-03	14.5 ± 1.8	1
M556	2018-10-28	15.8 ± 3.4	5
M676	2020-06-23	22.5 ± 2.0	1

The measured $^{239+240}\text{Pu}$ activity concentrations fit the trend of a decrease since the 1970's of the Pu concentration in seaweeds collected in Särödal. The activity concentration of $^{239+240}\text{Pu}$ in *F. serratus* collected at Särödal in the period 1967 to 1992 was analysed earlier (Holm et al., 1980; Roos et al., 1993) (Figure 28). The measured values are in the same order of magnitude as those reported in the literature. Levels between 20 and 100 mBq/kg d.wt. were usually reported in *F. vesiculosus* on the Norwegian coast in the last two decades (Skjerdal et al., 2017).

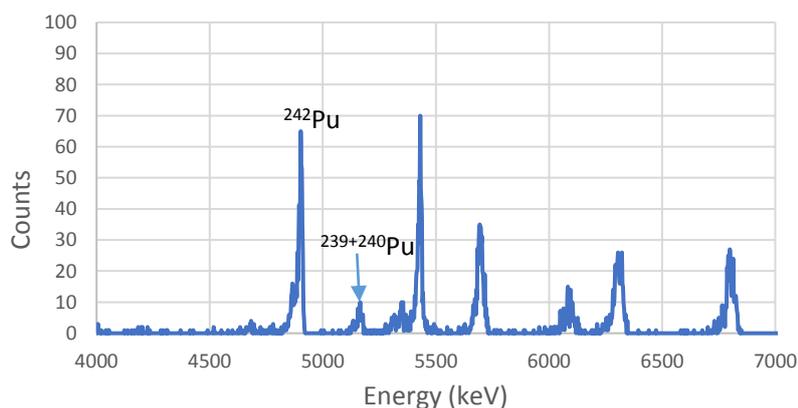


Figure 27: Alpha spectrum of sample M26 (Särödal, 2012-07-03).

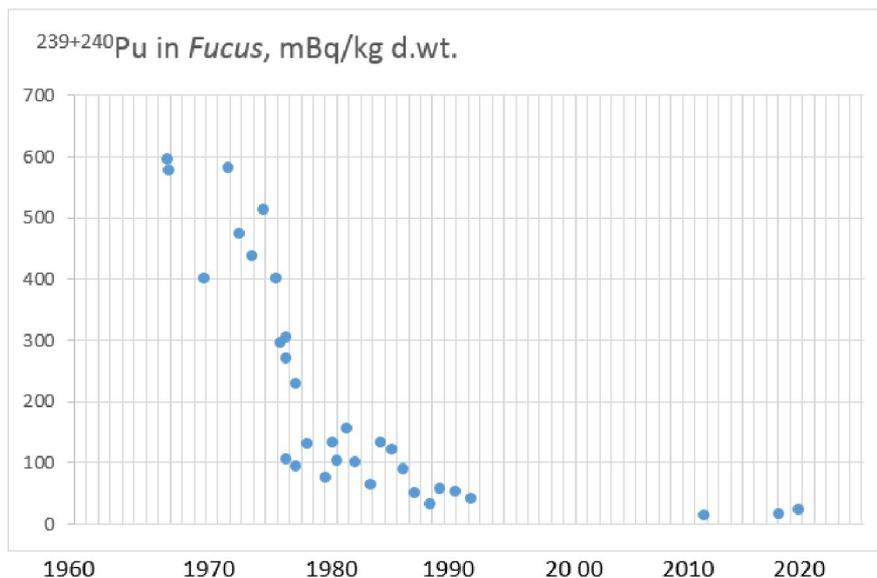


Figure 28: The activity concentration (mBq/kg d.wt.) of $^{239+240}\text{Pu}$ in *Fucus* collected at Särödal in the period 1967 to 2020. Samples from the period 1967-1992 were analysed earlier (Holm et al., 1980; Roos et al., 1993). The figure is based on original data behind these publications and the results of the measurements of the new samples from this project.

To go further with the analysis of Pu, the alpha spectrometry sources prepared from *Fucus* samples could be re-analysed by AMS to obtain the ratio of ^{239}Pu and ^{240}Pu . This method has already applied with success on peat samples by our group in collaboration with the University of Seville (Chamizo et al., 2020).

5.2. Stable elements (metals)

5.2.1. Lead, cadmium, cobalt and nickel

The analyses have up to now included lead (Pb), cadmium (Cd), cobalt (Co), nickel (Ni), mercury (Hg), and gadolinium (Gd). In some samples also arsenic (As) and selenium (Se) were measured. For the period 2011-2020, 1 to 7 samples per year were analysed, but for the period 1967 to 2010 just a few samples have been analysed up to now.

The levels of Pb, Cd, Co and Ni in the period 2011-2020 are shown in Figure 29 and reveals a doubling of the levels of lead and nickel and a 50% increase in the levels of Cd and Co. The variation in the concentration of the same elements during the 40-year period 1979-2020 is shown in Figure 30 and indicate almost constant levels for Co and slightly decreasing levels for Pb, Cd and Ni.

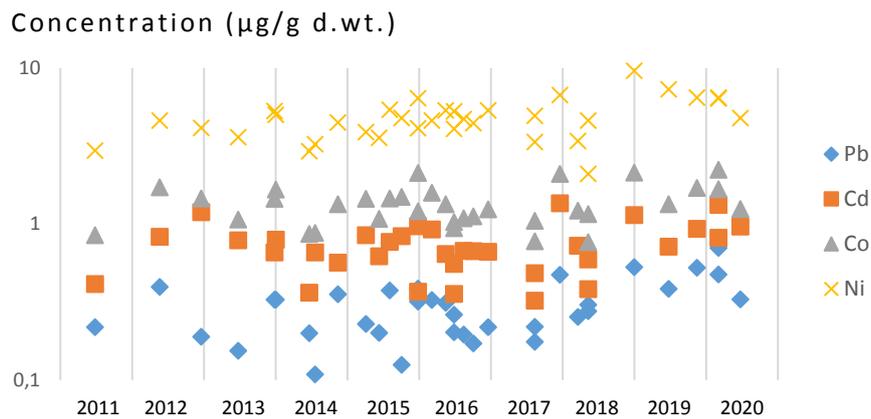


Figure 29: Concentration of lead, cadmium, cobalt and nickel in *Fucus serratus* from Särödal 2011-2020. Note! Logarithmic scale.

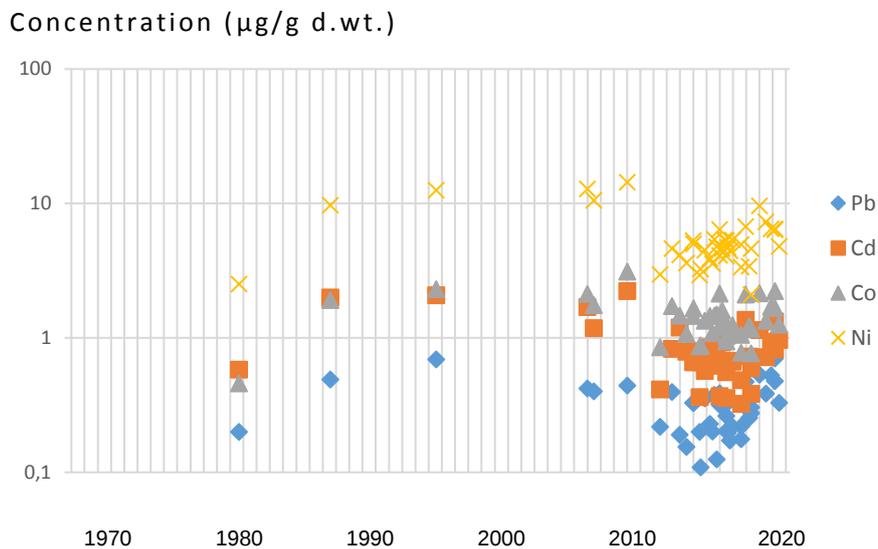


Figure 30: Concentration of lead, cadmium, cobalt and nickel in *Fucus serratus* from Särödal 1979-2020. Note! Logarithmic scale.

Forsberg et al. (1988) have reported metal concentrations in *Fucus vesiculosus* from the archipelago of Stockholm. For the metals we have in common - Ni, Pb, and Cd - the levels correspond well.

Jayasekera and Rossbach (1996) found 2 mg/kg d.wt. of cobalt in *F. serratus* from the North Sea and 1.7 mg/kg from the Baltic. The Cd level in the Baltic samples was 1.2 mg/g d.wt.

Already in 1952, Black and Mitchell (1952) reported 10 mg/kg of Pb in *F. serratus* and 2 mg/kg d.wt. in *F. vesiculosus* from Argyllshire, Scotland collected in May 1949. The Ni levels were 3.2 and 3.8 mg/kg d.wt. respectively and the Co levels 0.84 and 0.65 mg/kg d.wt. In January the same year, the Co concentration was 0.47 mg/kg d.wt.; Ni 1.6 mg/kg d.wt. and Pb 21 mg/kg d.wt. in *F. serratus*.

5.2.2. Mercury and gadolinium

The levels of Hg and Gd in the period 2011-2020 are shown in Figure 31. For Hg, a 10-fold decrease is registered between 2011 and 2016 and then a doubling of the concentration between 2016 and 2020. For Gd there is an increase with a factor of around 5 from 2011 to 2020.

The decreasing trend for Hg probably indicates an effect of an effect of the gradual phasing out of mercury in Sweden and other countries. However, some businesses and companies have an exemption for continued use. It will be interesting to continue to follow the future levels.

For gadolinium, there is a trend of increasing concentrations. The cause of this increase is most likely the increased use of Gd-complexes, such as (Gd-DTPA)²⁻ that are used as contrast agents in magnetic resonance imaging (MRI) in our hospitals. The first gadolinium-based contrast agent became available for clinical use in 1988 and since then a number of new gadolinium chelates have been developed and approved (Lohrke et al., 2016). These compounds are water soluble and are characterized by a very high stability.

The contrast agent is injected into the human bloodstream and excreted via the kidneys. Due to the high stability of these compounds, the anthropogenic Gd reacts neither in the human body (which is the prerequisite for its use as a contrast agent, because Gd³⁺ is toxic), nor in the sewage system or in waste and is introduced into rivers and lakes (Hatje et al., 2016).

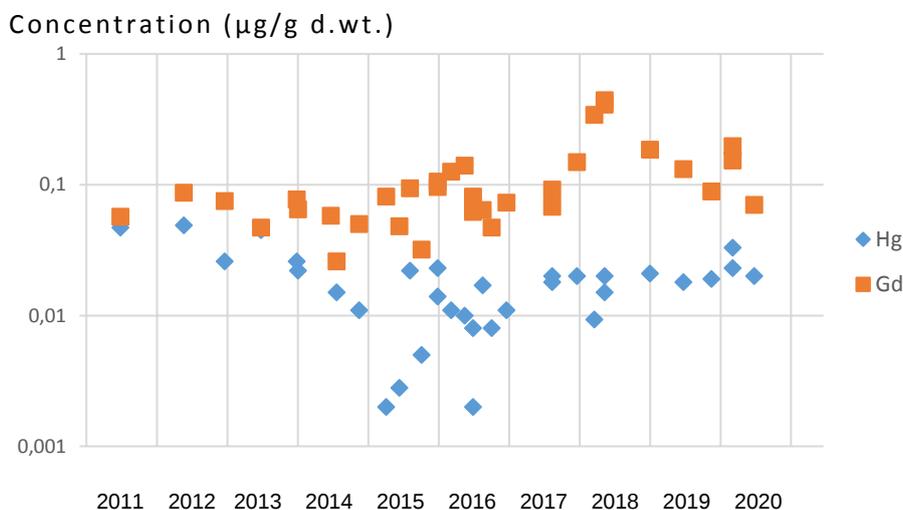


Figure 31: Concentration of mercury and gadolinium in *Fucus serratus* from Särödal 2011-2020. Note! Logarithmic scale.

The variation of the concentration of the same elements during the 40-year period 1979-2020 is shown in Figure 32, also indicating a decrease of Hg concentration and an increase in the Gd concentration. In view of the time variation observed during the period 2011-2020 - and then especially for mercury and gadolinium - it would be important to intensify the analyzes of *Fucus* samples from the period 1967-2010 as well as of samples from 2021 and 2022.

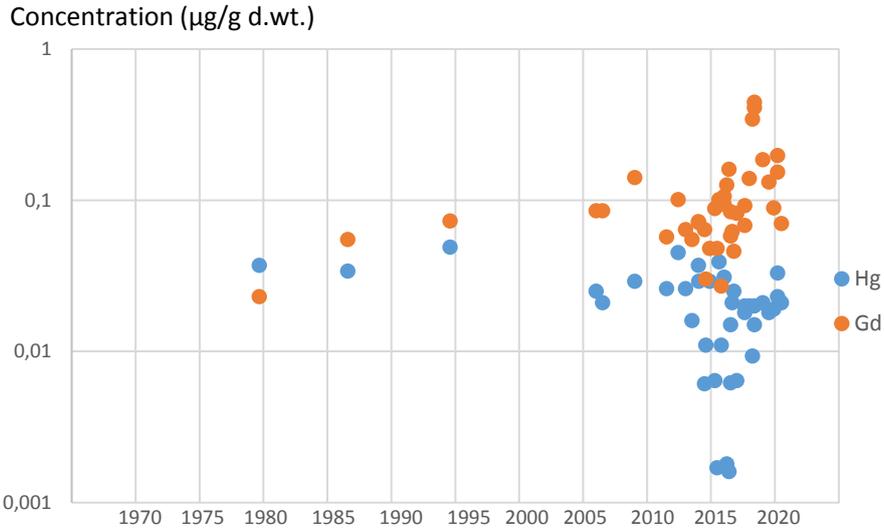


Figure 32: Concentration of mercury and gadolinium in *Fucus serratus* from Särðal 1979-2020. Note! Logarithmic scale.

The finding has a bearing on the expected releases from the upcoming European Spallation Source (ESS) outside Lund. A number of rare earth radionuclides will be produced and also released. An example is ^{148}Gd (pure alpha emitter, 70 year physical half-life) but there are many others (Pédehontaa-Hiaa et al., 2021). It will be very important to try to understand whether these anthropogenic rare earth elements behave differently than the natural rare earth elements as is obviously the case for Gd from contrast agents.

The concentration of the six elements have also been analysed in a sample of *Xanthoria parietina* growing 80 meters from the waterfront. The results are shown together with the corresponding results for *Fucus serratus* in Figure 33.

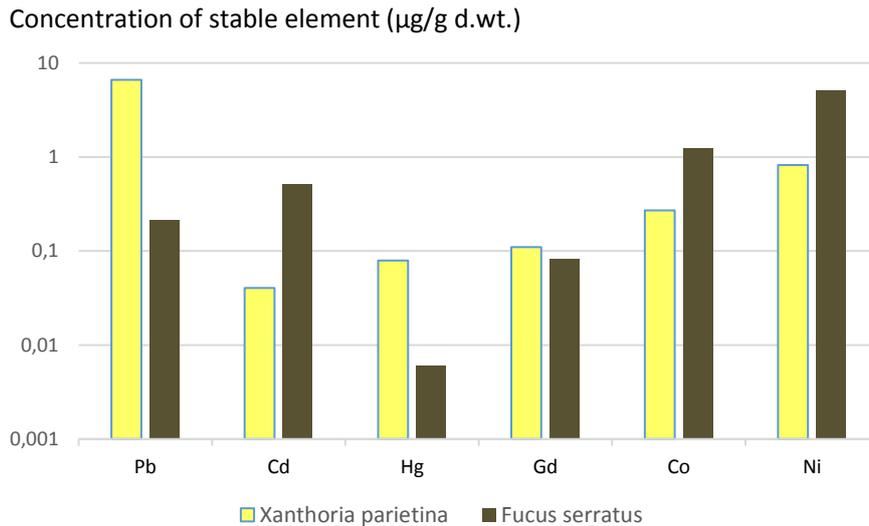


Figure 33: Comparison of the concentration µg/kg d.wt. of lead, cadmium, mercury, gadolinium, cobalt and nickel in *Xanthoria parietina* collected 80 m from the waterfront and in *Fucus serratus* at Särðal in March 2017. Note! Logarithmic scale.

The concentration of stable elements was distributed in the following order:

in *Xanthoria parietina*: Pb>Ni>Co>Gd>Hg>Cd and

in *Fucus serratus*: Ni>Co>Cd>Pb>Gd>Hg

In the project, *Xanthoria parietina* was used primarily to investigate dispersion and to quantify dispersion distances for ^{129}I from sea to land. Figure 33 also illustrates *Xanthoria parietina*'s capacity as a biomonitor for a number of stable trace elements (metals) of importance for air quality assessments. Together with the possibility of obtaining information on water quality with the help of *Fucus* at the same location, the collection and analysis of these two common bioindicators could be used to map larger areas regarding both air quality and water quality in a coastal environment. Further studies could also shed light on the possible spread of stable trace elements from sea to land.

6. Summary and conclusions

The project has revealed new information about long-time trends for various radionuclides in the marine environment of the Swedish west coast through measurements on brown algae *Fucus serratus* and *F. vesiculosus*.

The measurements indicate that after 1986, the main source of ^{137}Cs in the coastal waters at the Swedish west coast is dominated by outflow of Chernobyl-deposited ^{137}Cs from the Baltic Sea.

The program has made it possible to restart the analysis of transuranic elements at Lund University in Malmö and demonstrate a decrease in activity concentration of $^{239+240}\text{Pu}$ from 600 mBq/kg d.wt. in 1967 to 20 mBq/kg d.wt. in 2020. The method development part of the project led to a modernisation of the methods used in our laboratories for the extraction and separation of actinides and the preparation of alpha spectroscopy sources. By the end of the project, a method for the analysis of Pu isotopes in seaweed was fully developed and huge progresses have been made regarding the analysis of Am.

For the first time it has been able to carry out measurements of ^{129}I on parts of the new and historical sample material showing an unexpected increase in the ^{129}I levels during the last ten years.

The project has given new indications about the excess of ^{14}C on the Swedish west coast, most likely a result of inflow of North Sea water.

Tritium levels in *Fucus* and in sea water sample have been determined and all the measured values were at the expected environmental activity concentration ($< 3 \text{ Bq/L}$) so it can be concluded that the analysed samples were not subjected to any significant tritium contamination.

The study also shows that the brown algae could be important for the monitoring of emissions of naturally occurring radioactive materials (NORM) from the offshore oil and gas industry by systematically studying long-term and seasonal variations in the content of ^{228}Ra and in the future also ^{226}Ra and ^{210}Pb .

The project has also demonstrated the value of access to long time series of regularly collected samples of algae for other analyses than of radionuclides. Measurements on *Fucus* samples has also made it possible to describe time trends for a number of heavy metals like lead and cadmium. It has demonstrated a decrease of mercury levels, probably an effect of the gradual phasing out of mercury in Sweden and other countries and an increase of the concentrations of gadolinium, which most likely is a consequence of the increased use of gadolinium-based contrast agents for MR imaging in our hospitals.

The project has given new knowledge about the function and capacity of *Fucus* as a bioindicator especially for the degree of seasonal variations for various radionuclides and stable elements.

7. Outlooks and future activities

The project has confirmed brown algae as good bioindicators for radioactive substances and metals in the marine environment. The availability of half-century-long time series of this kind is of value, not least when it comes to assessing the extent of new future emissions and being able to put these in the right perspective.

The project also shows that precision and accuracy can be increased if differences between different algal species regarding e.g. concentration factors and seasonal variations are known in advance.

^{137}Cs levels should continue to be monitored.

Regarding ^{14}C , like ^{137}Cs , an important radionuclide from a radiation protection point of view, issues of sources and source terms should be further elucidated by analysis of newly collected samples and densification of the measurements of previously collected samples. To further investigate the possible influence from the reprocessing plants in Sellafield and La Hague, we suggest to analyse ^{14}C in Särö samples for the years still lacking from 1970 to 2000; we also propose to reanalyse the samples from 2001 and 2002, since those data were obtained with a previous less accurate instrumentation. The Särö series also enables further studies of seasonal variations of ^{14}C in *Fucus*.

Iodine-129 is certainly a well-known component of the environment, but which now shows an increasing concentration in the algae. The levels should be followed in future samples and the results obtained up to now be supplemented with analysis of samples from the periods 1968-83 and 2005-2010.

An important complement is to analyse ^{90}Sr in representative samples from the entire time series.

Although the Pu and Am levels have fallen sharply, it is important for a preparedness for future events to be able to measure these transuranic elements. The use of modern chromatographic resins to replace liquid/liquid separation was of great benefit for the analysis of Pu in seaweed sample. This strategy also shown promising results for the assessment of Am isotopes even if the analytical method could not be finalized in the duration of the project. Additional efforts would be necessary to understand the mechanism of separation of Am on chromatographic resins. The end goal of such a method development work would be a relatively fast method to sequentially separate U, Pu and Am before their analysis by alpha spectroscopy. To go further with the analysis of Pu, the alpha spectrometry sources prepared from *Fucus* samples could be reanalysed by AMS to obtain the ratio of ^{239}Pu and ^{240}Pu . This method has already applied with success on peat samples by our group in collaboration with the University of Seville (Chamizo et al., 2020).

Further studies of the naturally occurring radionuclide ^{228}Ra and in the future also ^{226}Ra and ^{210}Pb could give new information about water movements and/or

emissions of radioactive materials (NORM) from the offshore oil and gas industry.

It is also important to continue the ^{99}Tc analyses so that they also cover the time after 2010.

Continued studies of some stable elements may increase our understanding of how radioactive isotopes of the same element behave in the environment. In view of the time variation observed during the period 2011-2020 - and then especially for mercury and gadolinium - it would be important to intensify the analyzes of *Fucus* samples from the period 1967-2010 as well as of future samples.

Maintaining the test bank and continuing the collections is also important for the possibility of using the samples in the future for analysis of other radionuclides (such as ^{22}Na , ^{36}Cl , ^{41}Ca , ^{55}Fe , ^{63}Ni , ^{79}Se , ^{107}Pd , ^{151}Sm , ^{147}Pm , ^{236}U , etc.) and several stable elements and substances. There are very few long-term series of biosphere materials of this kind.

The material in this report could and should be analysed in more detail e.g. by studies of the possible correlation between different radionuclides.

8. Acknowledgements

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