Project SSM2019-5225: “Marine 14C levels around the Swedish coast” – Additional gamma spectrometric measurements and ICP-MS analysis of brown algae (Fucus spp.)

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Project SSM2019-5225:

"Marine ^{14}C levels around the Swedish coast" –

Additional gamma spectrometric measurements and ICP-MS analysis of brown algae (*Fucus* spp.)

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Summary

The results of the project SSM2019-5225, “Marine ^{14}C levels around the Swedish coast”, financed by the Swedish Radiation Safety Authority (SSM), are presented in the paper “Spatial and temporal variations of ^{14}C in *Fucus* spp. in Swedish coastal waters” published in the Journal of Environmental Radioactivity. The project SSM2019-5225 focussed on spatial variations in ^{14}C in *Fucus* spp. samples collected in 2020 along the entire Swedish west coast, in coastal waters in the very south of Sweden, and for the east coast up into the Gulf of Bothnia. In this report we present results from additional measurements of the *Fucus* spp. samples, using gamma-ray spectrometry (of ^{7}Be, ^{40}K, ^{60}Co, ^{137}Cs and ^{131}I) and inductively coupled plasma mass spectrometry (ICP-MS) (of Co, Ni, Cd, Gd, Hg and Pb). The gamma spectrometry showed in addition to the expected content of natural ^{40}K also ^{137}Cs and ^{7}Be, and at some places also ^{60}Co and ^{131}I. The ICP-MS analysis revealed a correlation between $F^{14}C$ and Gd, and the highest values of $F^{14}C$ and Gd were found close to Ringhals nuclear power plant. This strengthens the hypothesis that that a significant part of the observed peak in $F^{14}C$ on the west coast originates from Ringhals nuclear power plant.

Abbreviations and notations

AMS	Accelerator Mass Spectrometry
d.w.	Dry weight
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
NPP	Nuclear Power Plant
SD	Standard Deviation
SSM	Swedish Radiation Safety Authority
SUM	Standard Uncertainty of the Mean
$T_{1/2}$	Physical half-life
w.w.	Wet weight
%RSD	Percent Relative Standard Deviation

Contents

1. Introduction

The results of the project SSM2019-5225, "Marine ^{14}C levels around the Swedish coast", financed by the Swedish Radiation Safety Authority (SSM), are presented in the paper "Spatial and temporal variations of ^{14}C in *Fucus* spp. in Swedish coastal waters", published in the Journal of Environmental Radioactivity [1]. That paper describes all objectives of the project, which were to answer the questions:

- Are there differences in ^{14}C levels between Swedish coastal waters?
- Are the ^{14}C levels in Swedish coastal waters influenced by discharged or dumped radioactive wastes? Can releases of ^{14}C from Sellafield (UK) or La Hague (France) be detected on the Swedish west coast?
- What are representative marine background levels of ^{14}C to be used by the different Swedish nuclear power plants? There are three nuclear power plants in the country, with a total of six reactors in operation. These NPPs are the Forsmark (3 reactors), Oskarshamn (1) and Ringhals (2) plants. The two reactors at the Barsebäck NPP were shut down in 1999 and 2005, respectively. These reactors are now undergoing decommissioning, together with Oskarshamn's units 1 and 2. Apart from the reactors in operation or undergoing decommissioning, there are a number of other nuclear installations in Sweden, used to manufacture nuclear fuel and store spent nuclear fuel and radioactive waste.
- Do different parts of a specific brown algae show varying ^{14}C activity concentration (Bq/kg d.w.) and specific activity (Bq/kg C), e.g. depending on the age of the specific part of the algae?

The project SSM2019-5225 focussed on spatial variations in ^{14}C in *Fucus* spp. samples collected in 2020 along the entire Swedish west coast, in coastal waters in the very south of Sweden, and for the east coast up into Gulf of Bothnia. The paper [1] also includes ^{14}C data from another SSM project, SSM2020-797 (P.I. Sören Mattsson), providing temporal variations in ^{14}C at the sampling station Särdal (56.76N, 12.63E) on the Swedish west coast during the period 1967-2020 (27 *Fucus* spp. samples from various years have been used in this report).

In this report we present additional measurements of the *Fucus* spp. samples collected in 2020, using gamma-ray spectrometry (of ^{7}Be, ^{40}K, ^{60}Co, ^{137}Cs, and ^{131}I) and inductively coupled plasma mass spectrometry (ICP-MS) (of Co, Ni, Cd, Gd, Hg, and Pb).

2. Materials and Methods

Fucus spp. samples were collected at 45 sampling sites (Table 1) between 3 March and 13 May 2020. Details about the sampling sites (including maps), sampling strategy and specific sampling dates can be found in [1].

Carbon-14 analysis

Methods or sample pre-treatment, sample preparation, measurement and analysis for ^{14}C can be found in [1].

Gamma-ray spectrometry

Prior to gamma analysis, the *Fucus* spp. samples (typically 1-2 kg w.w.) were dried in a heating cabinet at 70 °C, then grounded and put in either a 250 ml or a 60 ml sample beaker. The gamma spectrometric measurements were performed at Medical Radiation Physics in Malmö (Lund University) using different HPGe-gamma spectrometers in lead shielded cavities [2]. Measurement time was typically 24-48 hours. Pulse height distributions obtained for each sample were evaluated using an in-house evaluation sheet, which was cross-checked with the detector software evaluation program, Gamma Vision 7.1^{TM}. For further information, see Ref [2].

ICP-MS

0.5 g of each dried sample (consisting of leftovers from the samples prepared for ^{14}C analysis, fraction A: vesicles and above, i.e. the new vegetative fronds, mainly representing growth since last spring) was submitted to the Department of Biology (Instrumental Chemistry), Lund University, for sample digestion and to the Department of Geology, Lund University, for Inductively Coupled Mass Spectrometry (ICP-MS) analysis using a Bruker Aurora Elite ICP-MS instrument (see [3]). The stable isotopes analysed were ^{59}Co, ^{60}Ni, ^{114}Cd, ^{111}Cd, ^{157}Gd, ^{202}Hg and ^{208}Pb. The results reported are elemental concentrations, assuming that the isotopes have their natural abundance in the seaweed samples.

Analysis of a certified reference material (ERM-CD200 Bladder wrack) 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) showed agreement within estimated uncertainty intervals for Pb, Cd, Hg, Se and As (Cu and Zn were not analysed). For further details on quality assurance in the ICP-MS measurements, see [3].

Table 1 Sampling sites for the collection of *Fucus* spp. samples.

Sea	Site number	Site name	Latitude	Longitude
Skagerrak	1	Strömstad, Båteviken	58.95N	11.13E
	2	Fjällbacka, Sälvik	58.59N	11.28E
	3	Smögen, Kleven	58.35N	11.22E
	4	Tjörn, Stockevik	57.96N	11.55E
Kattegat	5	Hällsvik	57.70N	11.74E
	6	Smarholmens badplats	57.43N	11.92E
	7	Frillesås	57.31N	12.15E
	8	Sallebacka	57.28N	12.14E
	9	Ringhals norr Gloppen	57.27N	12.11E
	10	Videbergshamn (Ringhals)	57.25N	12.11E
	11	Bua (Ringhals)	57.24N	12.10E
	12	Skeatången, Årnäshalvön	57.20N	12.17E
	13	Getterön	57.12N	12.21E
	14	Glommen	56.94N	12.36E
	15	Särdal	56.76N	12.63E
	16	Kattvik	56.46N	12.75E
	17	Mölle	56.28N	12.50E
Öresund Strait	18	Ålabodarna/Örenäs	55.94N	12.78E
	19	Barsebäcks camping	55.77N	12.92E
	20	Barsebäcks hamn Skansen	55.75N	12.90E
	21	Barsebäck between NPP and Skansen	55.75N	12.91E
	22	Barsebäck north of NPP	55.75N	12.92E
	23	Vikhög north fort	55.73N	12.95E
	24	Vikhög harbour	55.73N	12.96E
	25	Gamla Bjärred, Pilevägen	55.71N	13.03E
	26	Lomma	55.68N	13.06E
	27	Limhamn	55.58N	12.91E
	28	Lilla Hammar (Höllviken)	55.45N	12.95E
Baltic Sea	29	Smygehuks fyr	55.34N	13.35E
	30	Skillinge	55.47N	14.28E
	31	Hasslö	56.09N	15.46E
	32	Sandvik	56.55N	16.22E
	33	Dragskär	57.31N	16.57E
	34	Simpevarp, Åkvik	57.41N	16.64E
	35	Kråkelund	57.45N	16.75E
	36	Händelöp	57.67N	16.75E
	37	Jogersö	58.66N	17.05E
	38	Senneby, Sandviken, Väddö	59.98N	18.88E
Gulf of Bothnia	39	Öregrund	60.34N	18.45E
	40	Valudden	60.35N	18.37E
	41	Stenskär	60.36N	18.33E
	42	Norrboda, Gräsö	60.47N	18.39E
	43	Bönan	60.74N	17.30E
	44	Hölick	61.63N	17.43E
	45	Svenskärs fiskeläge	62.51N	17.88E

3. Results and discussion

Carbon-14

The results of the ^{14}C measurements are presented and discussed in [1] and briefly summarised here. The results are expressed as Fraction Modern, $F^{14}C$ (see [4, 5] for definition). The relation between specific activity of carbon, $\frac{A}{m_C}$, at the year of measurement, y, and $F^{14}C$ is given by [5]:

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

where $\delta^{13}C$ is the isotope fractionation of the sample (typical values of various environmental sample types are presented in Table B4 in [2]). For the *Fucus* spp. samples in [1], $\delta^{13}C$ varied between -20.6‰ and -12.9‰ (measured in 13 samples). The advantage of using $F^{14}C$ instead of specific activity ($\frac{A}{m_C}$) in high precision measurements of ^{14}C is that:

1) $F^{14}C$ is independent of the year of measurement, whereas $\frac{A}{m_C}$ is not;

2) $F^{14}C$ eliminates the effect of isotope fractionation, making various environmental sample types directly comparable.

As an example, for $F^{14}C = 1$, y = 2020 and $\delta^{13}C = -16‰$:

$\frac{A}{m_C}$ = 228.3 Bq (kg C)$^{-1}$.

The main results were:

1) The ^{14}C levels in *Fucus* spp. were significantly higher on the west coast than on the east coast (see Figure 1, from [1]). $F^{14}C$ values in *Fucus* spp. were up to about 1.10 in Skagerrak, and about 1.01, close to atmospheric levels, in the Baltic Sea and the Gulf of Bothnia.

2) The highest value of $F^{14}C$ observed was close to the Ringhals nuclear power plants (NPP) on the west coast, $F^{14}C \approx 1.3$ (see Figure 1), from [1]). This is higher than expected in the terrestrial environment of Ringhals NPP.

3) $F^{14}C$ showed a strong positive correlation with salinity on the west coast, and both increased towards the north (see Figure 2). This indicates that ^{14}C may be carried from other anthropogenic sources (e.g. from the nuclear fuel reprocessing plants at La Hague and Sellafield).

4) The values of $F^{14}C$ in the Särdal marine samples collected after the 1990s are clearly higher than $F^{14}C$ in clean air CO_2, indicating contributions of ^{14}C of anthropogenic origin (see Figure 3).

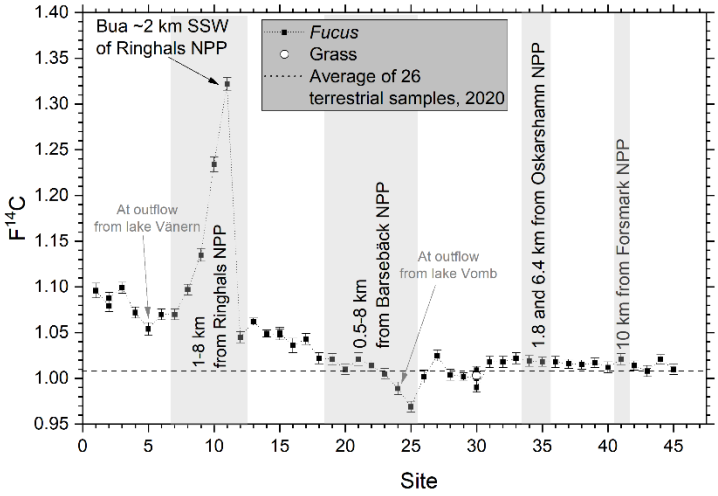

Figure 1 $F^{14}C$ values obtained from samples of *Fucus* spp. and grass (the latter at Skillinge, Site 30) collected along the Swedish coast, starting at the northernmost site on the west coast (Strömstad, Site 1) and continuing to the northernmost site on the east coast (Svenskärs Fiskeläge, Site 45). Only data from samples composed of the top parts of the *Fucus* spp. individuals (corresponding to growth during 2020) are included. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas. The dashed line shows the average $F^{14}C$ value of 26 terrestrial samples collected in southern Sweden for 2020, representing the atmospheric CO_2 reference value for 2020 (from Bernhardsson *et al.* [6]). Uncertainties represent 1 SD, obtained from repeated measurements of the same sample. *Picture from [1] with modifications.*

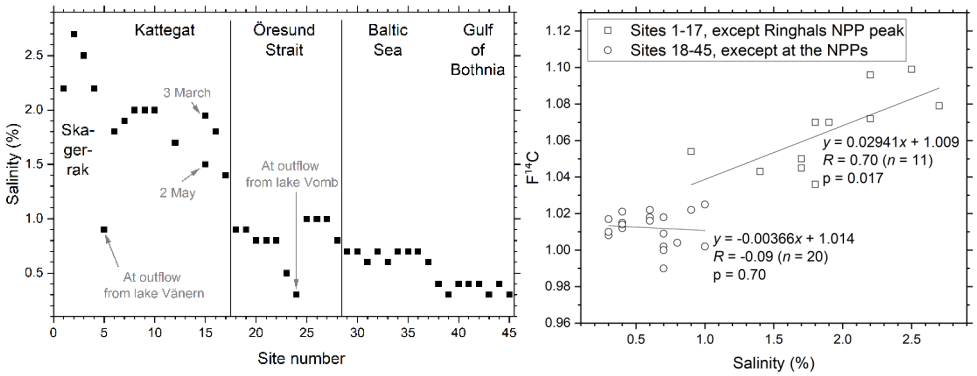

Figure 2 Left: Salinity measured at the time of sampling of *Fucus* spp. at the different sites in 2020. **Right:** Linear correlation between $F^{14}C$ and salinity for samples in the current study, divided into two sets separated by the border between Kattegat and the Öresund Strait. Sites 8-11 near Ringhals NPP have been excluded, as well as sites located close to all other NPPs (Sites 19-25, 34, 35 and 41; grey shading in Figure 1). The salinity at Särdal (Site 15) was measured using a portable seawater refractometer RHS-10ATC. *Picture from [1] with modifications.*

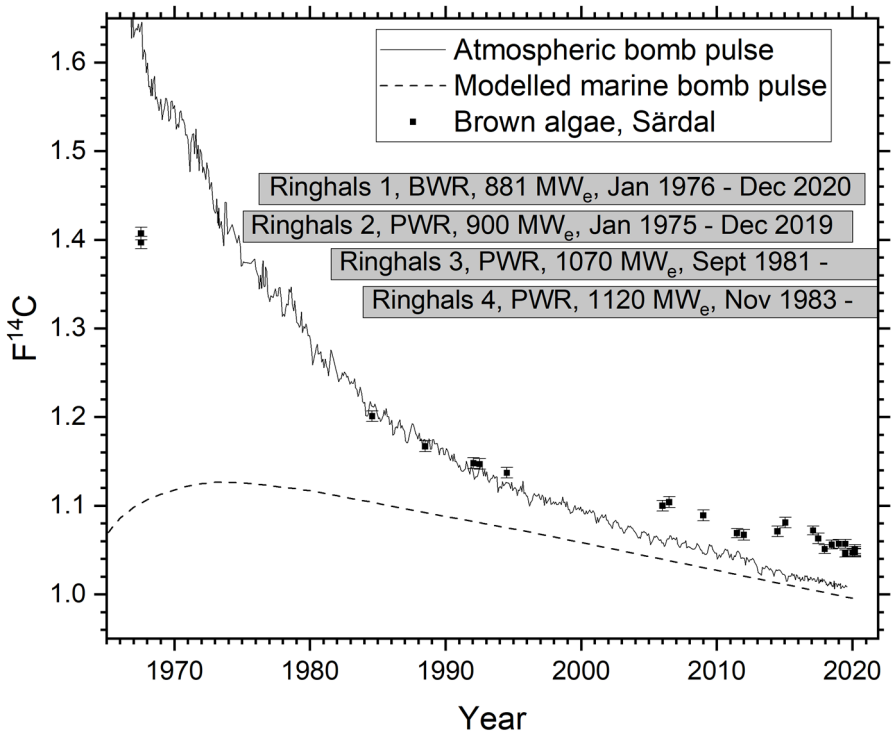

Figure 3 ^{14}C in *Fucus* spp. from Särdal in the period 1967-2020. Uncertainty represents 1 SD. Values of $F^{14}C$ in atmospheric CO_2 (atmospheric bomb pulse representative of the Northern Hemisphere) and oceans (modelled marine bomb pulse) are shown for comparison. Atmospheric data are Central European clean air CO_2 from Conen *et al.* [7], Hammer *et al.* [8], Levin and Kromer [9], and Levin *et al.* [10]. Modelled marine values up to 1996 are from Reimer *et al.* [11], representing average global oceanic water down to a depth of 75 m. Values from 1996 to 2020 are estimates based on linear regression of the marine data from 1987 to 1996. The periods of operation of the four reactors at Ringhals NPP, located ~70 km NNW of Särdal, are also given. *Picture from [1].*

Gamma spectrometry

Brown algae are known to display seasonal variations in activity concentrations of certain radionuclides, such as ^{137}Cs and ^{99}Tc [12-14]. The sampling in the current study was performed over a period of 2 months and 11 days. In an attempt to account for seasonal differences, sampling started in the south, and samples were collected from the northern sites during the latter part of the sampling period. The site Skillinge (in the very south-east of Sweden) was sampled at the very beginning (11 March 2020) as well as on the final day of sampling (13 May 2020) to assess seasonal variations. At Särdal samples were taken 3 March and 2 May.

The results of the gamma spectrometry are presented in Table A1 in Appendix 1 and summarized in Table 2 below. Of particular interest in this project is to link increased levels of ^{14}C found at the west coast with increased levels of other radionuclides, which may contribute to source apportionment of ^{14}C (mainly to releases from Ringhals NPP and to from long-range transport e.g. from Sellafield and LaHague).

Table 2 Summary of the results from the gamma-ray spectrometry. A: Activity concentration. MDA: Minimum detectable activity concentration. *N*: Number of measurements. SD: standard deviation. SUM: Standard uncertainty of the mean.

	^{7}Be (Bq/kg d.w.)		**^{40}K (Bq/kg d.w.)**		**^{60}Co (Bq/kg d.w.)**		**^{134}Cs (Bq/kg d.w.)**		**^{137}Cs (Bq/kg d.w.)**		**^{131}I (Bq/kg d.w.)**	
	A	**MDA**	**A**	**MDA**	**A**	**MDA**	**A**	**MDA**	**A**	**MDA**	**A**	**MDA**
N	46	46	47	47	3	47	42	46	47	47	22	47
Mean	30.6	6.3	934	20	3.1	1.8	1.5	1.0	7.9	1.4	5.7	0.8
Median	29.4	5.8	947	12	3.2	1.6	1.4	0.85	7.5	1.4	1.55	0.6
SD	12.3	2.5	165	16	0.4	1.0	0.5	0.6	5.8	0.8	11.3	0.5
SUM	1.8	0.4	24	2	0.2	0.1	0.1	0.1	0.8	0.1	2.4	0.1

^{7}Be

The results of the gamma spectrometry of ^{7}Be are shown in Figure 4. ^{7}Be is produced by interaction of cosmic rays with molecules of N, O and C in the atmosphere [15]. Once produced, ^{7}Be 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 ^{7}Be deposition has earlier been demonstrated [16, 17]. The mean yearly deposition in Sweden has been estimated to around 1500 Bq/m^2.

The difference between *Fucus serratus* and *F. vesiculosus* and between Särdal, Skillinge and Hasselö (and there also for seaweed with and without ongrowth) is within the range of the variation. Long-term measurements in Särdal show typical activity concentrations of 20-40 Bq/kg d.w. in *F. serratus* and 10-40 Bq/kg d.w. in *F. vesiculosus* [18]. Higher values, up to 100 Bq/kg were reached in 2018 and 2019. Still higher values were found in the environmental control programs for Ringhals with average values between 150 and 300 Bq/kg d.w. in 1995-2019 [19]. At Barsebäck the levels have varied between 19 and 150 Bq/kg in *F. vesiculosus* with an average of about 60 Bq/kg in the period 2011 to

2019 [19]. It would be interesting to further investigate the fraction of possible Ringhals produced ^{7}Be in *Fucus* taken in the immediate vicinity of the plant.

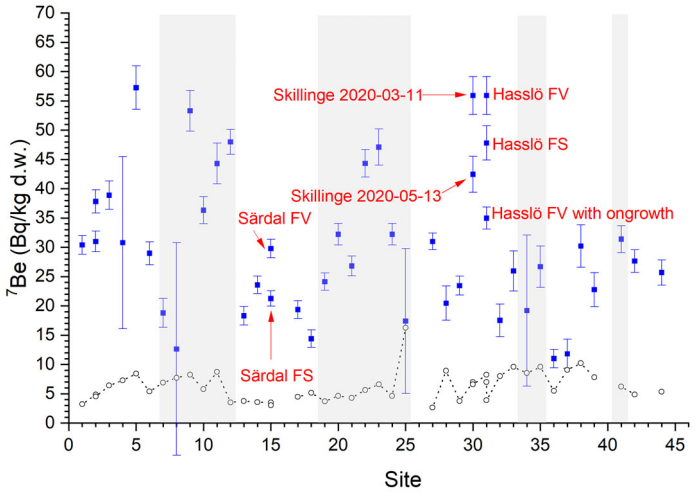

Figure 4 ^{7}Be in *Fucus* spp. collected in 2020. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas (see Figure 1). Open circles: MDA.

^{40}K

Obtained activity concentrations for ^{40}K are shown in Figure 5. The mean value for the ^{40}K activity concentration is similar to that for the long-term measurements at Särdal. The mean value for *Fucus serratus* from Särdal during the period 1967-2020 was 885±150 Bq/kg d.w. which corresponds to 29 g potassium per kg of dry seaweed [20]. There is a tendency of decreasing concentrations towards the Baltic Sea. *Fucus* spp. from the Baltic Sea showed a mean ^{40}K activity concentration of 817±33 Bq/kg d.w. in the period 1978-1983 [21].

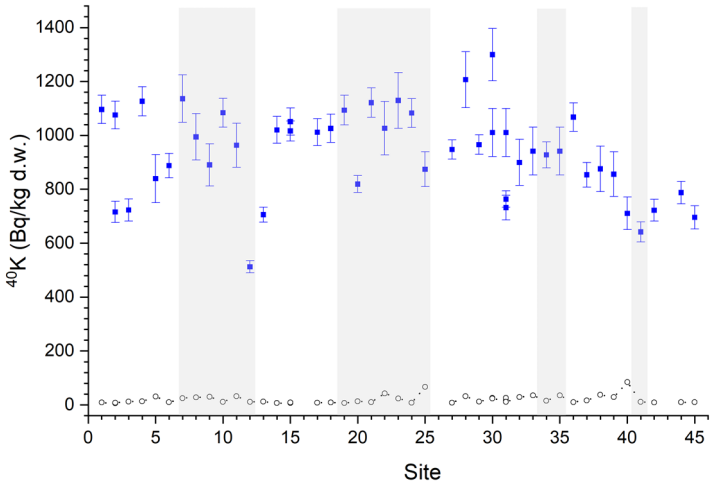

Figure 5 ^{40}K ($T_{1/2}$ = 1.28·10^{9} years) in *Fucus* spp. at the various sites. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas (see Figure 1). Open circles: MDA.

^{60}Co

Fucus vesiculosus and *F. serratus* have since long been used to map the spatial and temporal distribution of the activation products ^{60}Co, ^{58}Co, ^{54}Mn and ^{65}Zn around nuclear power plants [22, 23]. In the current measurement series, only ^{60}Co was detected and when detected, only in a few samples close to Barsebäck NPP (Figure 6), now under decommissioning. The levels were low and are probably explained by remobilization of ^{60}Co from sediments or fresh releases in connection with the ongoing decommissioning. The measuring points around the other nuclear power plants have been too far from the reactors to give detectable results.

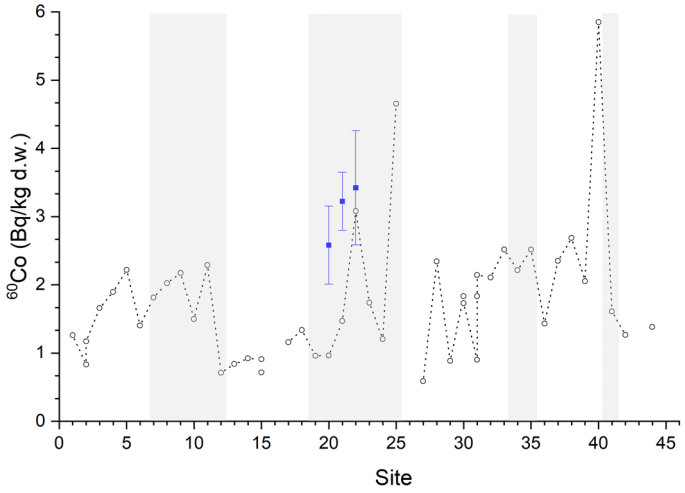

Figure 6 ^{60}Co ($T_{1/2}$ = 5.3 y) in *Fucus* spp. at the various sites. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas (see Figure 1). Open circles: MDA.

^{137}Cs

^{137}Cs was higher on the east coast than on the west coast (Figure 7). Lower salinity is known to increase the uptake of ^{137}Cs in *Fucus* spp. [24, 25].

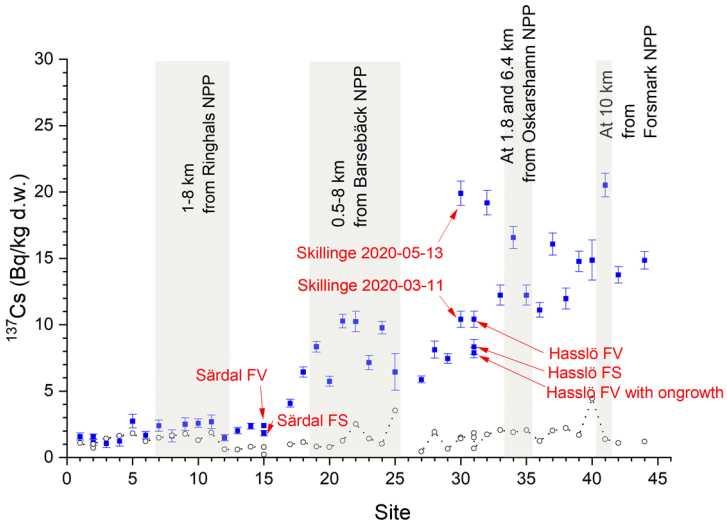

Figure 7 ^{137}Cs ($T_{1/2}$ = 30.2 y) in *Fucus* spp. at the various sites. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas (see Figure 1). Open circles: MDA.

Figure 8 shows the ^{137}Cs activity concentrations in *Fucus* spp. as a function of collection date for all sites. In general, the ^{137}Cs activity concentrations are higher towards the end of the sampling period (May) compared to the beginning of the sampling period (March). The site Skillinge (sampled at the very beginning as well as on the final day of sampling) shows almost a doubling of the ^{137}Cs activity concentration from March to May. At Särdal, the activity concentration (mean for *F. serratus* and *F. vesiculosus*) also almost doubled from 2.1 to 4.0 Bq/kg d.w. from March 3 to May 2, 2020. This is in accordance with the seasonal variations seen earlier [18].

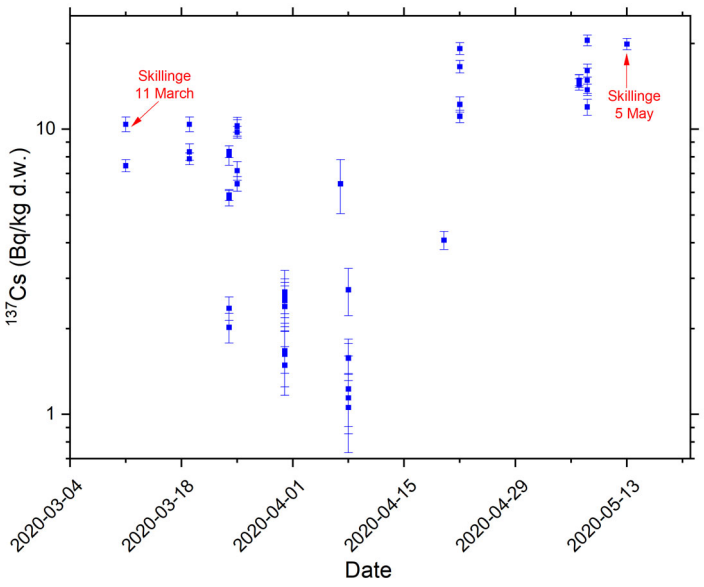

Figure 8 ^{137}Cs ($T_{1/2}$ = 30.2 y) in *Fucus* spp. as a function of collection date. Note the logarithmic scale.

^{131}I

The activity concentrations of ^{131}I in *Fucus vesiculosus* are increased around Barsebäck NPP compared to the other NPPs (see Figure 9). This is most likely a result of releases from the sewage treatment plants at Sjölunda (Malmö) and Källby (Lund) due to the presence of ^{131}I in excreta from patients treated with ^{131}I mainly for thyrotoxicosis and thyroid cancer). In the same way, the still higher level at station 5 (Hällsvik) is likely to be due to releases from patients at hospitals and patients who have returned home from the hospitals in Gothenburg and along the Göta älv. The sewage treatment plant Ryaverken serving Gothenburg and its surrounding municipalities was at least earlier known to have a very short delay time for the waste water [26].

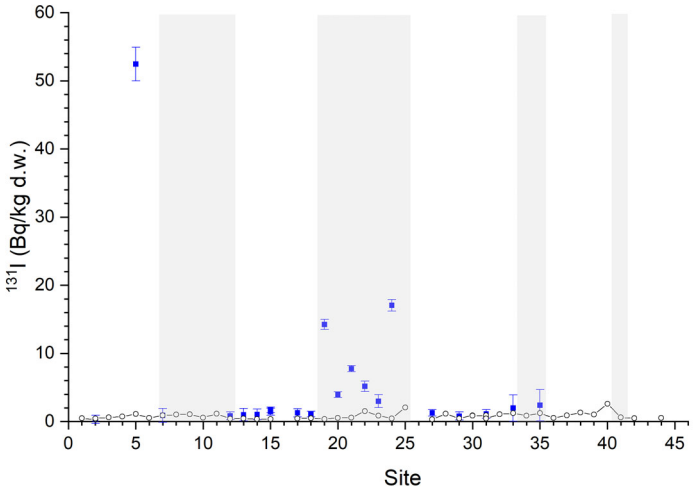

Figure 9 ^{131}I ($T_{1/2}$ = 8.0 d) in *Fucus* spp. at the various sites. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas (see Figure 1). Open circles: MDA.

Correlation between ^{14}C and gamma emitters

A correlation matrix for F^{14}C and the measured gamma emitters (except ^{60}Co due to the low number of data above the MDA) for all samples is presented in Table 3. A negative correlation is found between F^{14}C and ^{137}Cs, which most likely is due to completely different sources resulting in higher F^{14}C on the west coast and higher ^{137}Cs on the east coast. The high F^{14}C on the west coast most likely originates from Ringhals NPP and Sellafield/La Hague, while the high ^{137}Cs on the east coast is Chernobyl-derived. In Table 4, only samples collected in Skagerrak and Kattegat are included in the correlation analysis: F^{14}C shows no correlation to any of the gamma emitters. The same is valid when separately investigating the Öresund Strait, the Baltic Sea and the Gulf of Bothnia (Table 5).

Table 3 Correlation matrix for $F^{14}C$ and ^{7}Be, ^{40}K, ^{134}Cs, ^{137}Cs and ^{131}I for *Fucus* spp. samples for all sites. p-values <0.05 are shaded in grey.

		^{7}Be	**^{40}K**	**^{137}Cs**	**^{131}I**
$F^{14}C$	Pearson Corr.	0.21	0.04	-0.54	2.0E-4
	p-value	0.16	0.809	1.5E-4	0.99
^{7}Be	Pearson Corr.		-0.119	-0.15	0.55
	p-value		0.469	0.31	0.01
^{40}K	Pearson Corr.			-0.10	-0.03
	p-value			0.52	0.91
^{137}Cs	Pearson Corr.				-0.02
	p-value				0.91

Table 4 Correlation matrix for $F^{14}C$ and ^{7}Be, ^{40}K, ^{134}Cs, ^{137}Cs and ^{131}I for *Fucus* spp. samples collected in Skagerrak and Kattegat. p-values <0.05 are shaded in grey.

		^{7}Be	**^{40}K**	**^{137}Cs**	**^{131}I**
$F^{14}C$	Pearson Corr.	0.31	0.15	0.16	-0.06
	p-value	0.20	0.56	0.53	0.87
^{7}Be	Pearson Corr.		-0.42	-0.02	0.74
	p-value		0.08	0.93	0.02
^{40}K	Pearson Corr.			0.29	-0.16
	p-value			0.25	0.67
^{137}Cs	Pearson Corr.				0.19
	p-value				0.62

Table 5 Correlation matrix for $F^{14}C$ and ^{7}Be, ^{40}K, ^{134}Cs, ^{137}Cs and ^{131}I for *Fucus* spp. samples collected in the Öresund Strait, the Baltic Sea and the Gulf of Bothnia. p-values <0.05 are shaded in grey (none).

		^{7}Be	**^{40}K**	**^{137}Cs**	**^{131}I**
$F^{14}C$	Pearson Corr.	0.01	-0.19	0.25	-0.45
	p-value	0.97	0.34	0.21	0.14
^{7}Be	Pearson Corr.		0.11	-0.14	0.04
	p-value	--	0.57	0.48	0.91
^{40}K	Pearson Corr.			-0.23	0.46
	p-value		--	0.23	0.12
^{137}Cs	Pearson Corr.				0.14
	p-value				0.65

ICP-MS

The results of the ICP-MS analysis are presented in Table A2 in Appendix 1. The results are summarized in Table 6 and in Figure 10. Typical uncertainties in terms of relative standard deviation in percent (%RSD) were on average 5%-8% for most samples and isotopes analysed (^{59}Co, ^{60}Ni, ^{114}Cd, ^{111}Cd, ^{155}Gd, ^{157}Gd, ^{208}Pb). For ^{202}Hg, the %RSD was generally higher than for the other metals (average %RSD: 59% with a standard deviation 40%). Forsberg et al. [27] have reported metal concentrations in *Fucus vesiculosus* from the archipelago of Stockholm. The ranges of concentrations for Ni, Cd, Pb and Co are of the same order of magnitude as results in Table 6. This is also the situation for the same metals in *Fucus vesiculosus* and *F. serratus* collected at Tjärnö (between Båteviken, Strömstad och Sälvik, Fjällbacka) in 2014 [28].

Table 6 Summary of the results from the ICP-MS measurements (elemental concentrations). %RSD: relative standard deviation. SD: standard deviation. SUM: Standard uncertainty of the mean. * From ^{114}Cd. ** From ^{111}Cd.

	Co (ppb)	% RSD	Ni (ppb)	% RSD	Cd* (ppb)	% RSD	Cd** (ppb)	% RSD	Gd (ppb)	% RSD	Hg (ppb)	% RSD	Pb (ppb)	% RSD
Mean	1725	5	7709	6	1271	7	1289	7	246	8	13	59	746	7
Median	1588		6790		1075		1086		162		12		444	
SD	893	1	4347	2	581	2	594	2	227	3	5	40	833	3
SUM	126		615		82		84		32		1		118	

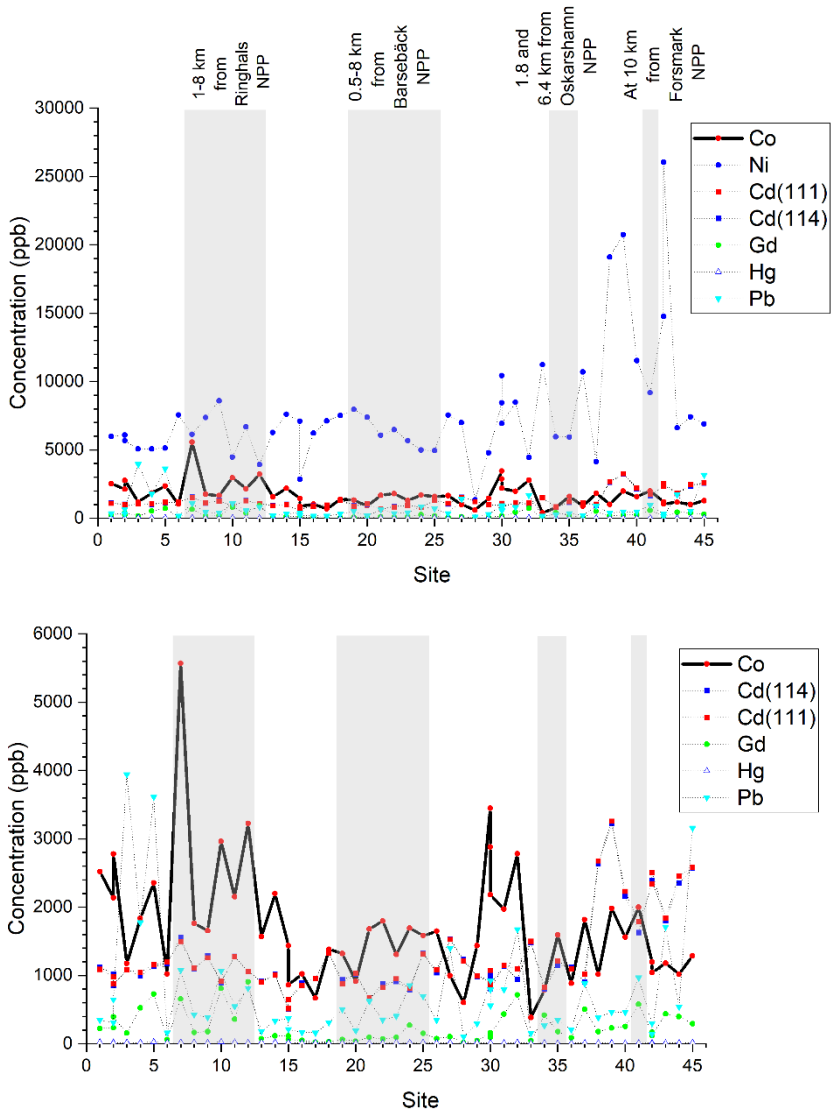

Figure 10 Concentrations of various elements in *Fucus* spp. obtained with ICP-MS analysis. Relative standard deviations are typically 5-8 % for all elements except Hg (average %RSD 59%). Upper graph: all results. Lower graph: all results except Ni.

The average concentration was highest for Ni, followed by Co, Cd, Pb, Gd and Hg. The concentrations of Ni and Cd were highest in the northern Baltic Sea and in the Gulf of Bothnia. In the current report we focus on possible correlations between these elements and $F^{14}C$. The correlation between $F^{14}C$ and Co, Ni, Cd, Gd and Pb is presented in Table 7 (Hg has been excluded due to its high uncertainties). The only significant correlation (p-value < 0.05) for $F^{14}C$ can be seen for Gd. Other significant correlations can be seen between Co and Gd, Ni and Cd, and Gd and Pb.

Table 7 Correlation matrix for $F^{14}C$ and Co, Ni, Cd, Gd and Pb. p-values <0.05 are shaded in grey. *From ^{114}Cd. **From ^{111}Cd.

		Co	Ni	Cd*	Cd**	Gd	Pb
$F^{14}C$	Pearson Corr.	0.25	-0.15	-0.14	-0.15	0.29	0.08
	p-value	0.08	0.30	0.34	0.30	0.04	0.59
Co	Pearson Corr.		-0.15	-0.13	-0.13	0.58	0.19
	p-value		0.28	0.38	0.37	8.63E-06	0.20
Ni	Pearson Corr.			0.71	0.71	-0.21	-0.22
	p-value			7.04E-09	7.04E-09	0.15	0.12
Cd*	Pearson Corr.				1.00	0.04	0.09
	p-value				7.04E-09	0.76	0.54
Cd**	Pearson Corr.					0.07	0.10
	p-value					0.62	0.50
Gd	Pearson Corr.						0.45
	p-value						7.04E-09

The correlation between $F^{14}C$ and Gd is further visualized in Figure 11. The highest Gd concentrations and $F^{14}C$ values are both found in Kattegat, close to Ringhals NPP, hence the correlation between the two. Gd is present in Ringhals NPP in fuel rods (as Gd_2O_3) [29]. The Gd concentration varies significantly at nearby sites in Skagerrak, Kattegat, the Baltic Sea and the Gulf of Bothnia. The variation in Gd concentration between nearby sites is less pronounced in southern Kattegat and in the Öresund Strait. Site 5 is located at the outflow of Göta älv and 24 is located north of the outflow from Sjölunda wastewater treatment plant and Höje å carrying the releases from the Källby wastewater treatment plant. Both these sites are characterized by lower salinity than the nearby sites (see Figure 2) and with higher Gd concentrations than in nearby sites. These increases in Gd concentration thus most likely stem from river runoff of Gd originating from the use of Gd-containing contrast agents used for magnetic resonance imaging in hospitals.

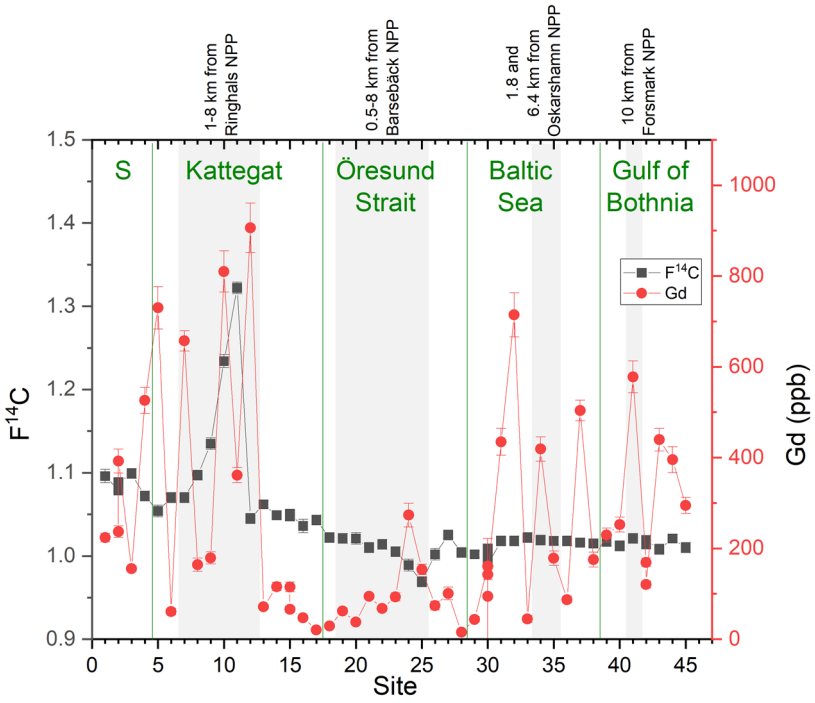

Figure 11 $F^{14}C$ and Gd concentration in *Fucus* spp. collected along the Swedish coast in 2020. S: Skagerrak.

4. Summary and conclusions

The gamma spectrometric measurements of *Fucus* spp. showed the well-known presence of naturally occurring ^{40}K (algae contain high concentrations of potassium), likewise naturally produced ^{7}Be among which there may also be a component caused by releases from the local nuclear power plants. The activation product ^{60}Co has been traced in the vicinity of the nuclear power plants in operation and in the vicinity of the decommissioned nuclear power plant in Barsebäck. The activity concentration of ^{137}Cs in *Fucus* spp. is affected by local emissions and above all by inflows from the North Sea and transport of Chernobyl ^{137}Cs from the Baltic Sea. The uptake of ^{137}Cs in *Fucus* spp. is strongly dependent on salinity and also on the season.

The correlation between $F^{14}C$ and Gd, and in particular the finding that the highest values of $F^{14}C$ and Gd are found close to Ringhals NPP, strengthens the hypothesis that that a significant fraction of the peak in $F^{14}C$ on the west coast originates from Ringhals NPP.

5. Outlook

More information can most likely be extracted from the gamma and ICP-MS analyses. It would be interesting to do this in the context of a new project.

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Appendix

Table A1 Results of gamma spectrometry measurements. FV – *Fucus vesiculosus*. FS – *Fucus serratus.* FR – *Fucus radicans.*

						Cs-137 (Bq kg^{-1} d.w.)			Co-60 (Bq kg^{-1} d.w.)			Be-7 (Bq kg^{-1} d.w.)			I-131 (Bq kg^{-1} d.w.)			Ka-40 (Bq kg^{-1} d.w.)		
Site	**Site name**	**Species**	**Sampling date YYY-MM-DD**	**Lab code**	**Detector**	**A**	**SD**	**MDA**	**A**	**SD**	**MDA**	**A**	**SD**	**MDA**	**A**	**SD**	**MDA**	**A**	**SD**	**MDA**
1	Båtevikens badplats. Strömstad	FV	2020-04-08	M645	5	1.6	0.3	1.1	<MDA		1.3	30.4	1.6	3.2	<MDA		0.5	1096	52	9
2	Fjällbacka. Sälvik. norr om hamn. med påväxt	FV	2020-04-08	M644	5	1.6	0.2	0.7	<MDA		0.8	31.0	1.8	4.9	0.3	0.6	0.3	1076	51	6
2	Fjällbacka. Sälvik. hamn och söder om hamn. utan påväxt	FV	2020-04-08	M641	5	1.1	0.2	1.0	<MDA		1.2	37.8	2.0	4.5	<MDA		0.4	716	39	8
3	Smögen. Kleven	FV	2020-04-08	M642	5	1.1	0.3	1.4	<MDA		1.7	38.9	2.4	6.4	<MDA		0.6	723	41	12
4	Stockevik. Tjörn	FV	2020-04-08	M643	5	1.2	0.4	1.6	<MDA		1.9	30.8	14.7	7.3	<MDA		0.7	1127	54	13
5	Hällsvik	FV	2020-04-08	M647	7	2.7	0.5	1.8	<MDA		2.2	57.3	3.7	8.4	52.5	2.5	1.1	840	89	31
6	Smarholmens badplats	FV	2020-03-31	M639	5	1.7	0.3	1.2	<MDA		1.4	29.0	2.0	5.4	<MDA		0.5	888	45	10
7	Frillesås	FV	2020-03-31	M638	7	2.4	0.4	1.5	<MDA		1.8	18.8	2.5	6.9	0.9	1.0	0.9	1136	88	25
8	Sallebacka	FV	2020-03-31	M634	7	1.6	0.5	1.7	<MDA		2.0	12.6	18.1	7.7	<MDA		1.0	994	86	28
9	Ringhals norr Gloppen	FV	2020-03-31	M635	7	2.5	0.5	1.8	<MDA		2.2	53.3	3.5	8.3	<MDA		1.1	890	78	30
10	Videbergshamn (Ringhals)	FV	2020-03-31	M636	5	2.6	0.3	1.3	<MDA		1.5	36.3	2.3	5.8	<MDA		0.6	1084	53	11
11	Bua (Ringhals)	FV	2020-03-31	M640	7	2.7	0.5	1.9	<MDA		2.3	44.3	3.5	8.7	<MDA		1.1	963	82	32
12	Skeatången. Ärnäshalvön	FV	2020-03-31	M637	4	1.5	0.2	0.6	<MDA		0.7	48.0	2.1	3.5	0.9	0.6	0.4	512	22	11
13	Getterön	FV	2020-03-24	M630	4	2.0	0.2	0.6	<MDA		0.8	18.3	1.6	3.8	1.0	0.9	0.5	706	28	12
14	Glommen	FV	2020-03-24	M629	5	2.4	0.2	0.8	<MDA		0.9	23.6	1.5	3.5	1.1	0.8	0.3	1020	49	7
15	Särdal V-A	FV	2020-03-03	M612	5	2.4	0.2	0.8	<MDA		0.9	29.8	1.6	3.5	1.7	0.4	0.3	1051	50	6
15	Särdal S	FS	2020-03-03	M611	4	1.8	0.2	0.2	<MDA		0.7	21.3	1.3	3.0	1.4	0.5	0.4	1016	37	9
17	Mölle	FV	2020-04-20	M654	5	4.1	0.3	1.0	<MDA		1.2	19.4	1.5	4.5	1.3	0.6	0.4	1012	50	8
18	Ålabodarna/Örenäs	FV	2020-03-25	M626	5	6.4	0.4	1.2	<MDA		1.3	14.4	1.5	5.1	1.1	0.4	0.5	1026	52	9

19	Barsebäcks camping	FV	2020-03-24	M625	5	8.3	0.4	0.8	<MDA		1.0	24.1	1.5	3.7	14.3	0.8	0.4	1093	55	7
20	Barsebäcks hamn Skansen	FV	2020-03-24	M621	4	5.7	0.4	0.8	2.6	0.6	1.0	32.2	1.9	4.6	3.9	0.4	0.5	819	32	13
21	Barsebäck between NPP and Skansen	FV	2020-03-25	N624	5	10.3	0.5	1.3	3.2	0.4	1.5	26.8	1.7	4.3	7.8	0.4	0.6	1121	54	10
22	Barsebäck north of NPP	FV	2020-03-25	M627	7	10.2	0.8	2.5	3.4	0.8	3.1	44.3	2.4	5.7	5.2	0.8	1.5	1027	100	43
23	Vikhög fort	FV	2020-03-25	M628	7	7.2	0.5	1.4	<MDA		1.7	47.1	3.1	6.6	3.0	0.9	0.9	1129	103	24
24	Vikhög hamn	FV	2020-03-25	M620	5	9.8	0.5	1.0	<MDA		1.2	32.2	1.9	4.6	17.1	0.8	0.5	1083	53	9
25	Gamla Bjärred. Pilevägen	FV	2020-04-07	M646	7	6.4	1.4	3.5	<MDA		4.7	17.4	12.4	16.3	<MDA		2.1	874	65	67
27	Limhamn	FV	2020-03-24	M622	4	5.9	0.3	0.5	<MDA		0.6	31.0	1.4	2.7	1.3	0.5	0.3	947	36	8
28	Lilla Hammar (Höllviken)	FV	2020-03-24	M623	7	8.1	0.6	1.9	<MDA		2.3	20.5	2.9	8.9	<MDA		1.2	1207	104	33
29	Smygehuks fyr	FV	2020-03-11	M615	5	7.5	0.4	0.7	<MDA		0.9	23.5	1.6	3.8	0.8	0.6	0.5	966	37	12
30	Skillinge. 1A	FV	2020-03-11	M614	7	10.4	0.6	1.5	<MDA		1.8	55.9	3.2	7.0	<MDA		0.9	1010	89	25
30	Skillinge	FV	2020-05-13	M674	7	19.9	0.9	1.4	<MDA		1.7	42.5	3.1	6.6	<MDA		0.9	1300	97	24
31	Hasslö	FS	2020-03-19	M617 (FS)	5	8.3	0.6	1.8	<MDA		0.9	47.8	2.9	8.3	<MDA		0.8	732	46	15
31	Hasslö	FV	2020-03-19	M618 (FV)	7	10.4	0.6	1.5	<MDA		1.8	55.9	3.2	7.0	<MDA		0.9	1010	89	25
31	Hasslö	FV+FS	2020-03-19	M619 (FV with ongrowth)	4	7.9	0.4	0.7	<MDA		2.1	35.0	1.9	3.9	1.1	0.6	0.5	763	30	12
32	Sandvik	FV	2020-04-22	M658	7	19.2	0.9	1.7	<MDA		2.1	17.5	2.8	8.0			1.0	899	85	29
33	Dragskär	FV	2020-04-22	M655	7	12.2	0.8	2.1	<MDA		2.5	26.0	3.4	9.6	2.0	1.9	1.3	942	88	35
34	Simpevarp Åkvik	FV	2020-04-22	M657	5	16.6	0.8	1.9	<MDA		2.2	19.2	12.9	8.5	<MDA		0.8	928	48	16
35	Kråkelund	FV	2020-04-22	M656	7	12.2	0.8	2.1	<MDA		2.5	26.7	3.5	9.6	2.4	2.3	1.3	942	88	35
36	Händelöp	FV	2020-04-22	M659	5	11.1	0.5	1.2	<MDA		1.4	11.0	1.6	5.5	<MDA		0.5	1068	53	10
37	Jogersö	FV	2020-05-08	M663	5	16.1	0.8	2.0	<MDA		2.4	11.8	2.5	9.1	<MDA		0.9	854	46	17
38	Senneby. Sandviken. Väddö	FV	2020-05-08	M665	7	12.0	0.8	2.2	<MDA		2.7	30.2	3.6	10.2	<MDA		1.3	876	85	37
39	Öregrund	FR	2020-05-07	M664	7	14.8	0.8	1.7	<MDA		2.1	22.8	2.9	7.8	<MDA		1.0	856	83	28
40	Valudden	FR	2020-05-08	M661	7	14.9	1.5	4.5	<MDA		5.8				<MDA		2.6	711	60	84
41	Stenskär	FR	2020-05-08	M662	5	20.5	0.9	1.4	<MDA		1.6	31.4	2.3	6.2	<MDA		0.6	642	37	11
42	Norrboda. Gräsö. A	FR	2020-05-08	M668	5	13.8	0.6	1.1	<MDA		1.3	27.6	1.9	4.9	<MDA		0.5	722	40	9
44	Hölick	FR	2020-05-07	M667	5	14.9	0.7	1.2	<MDA		1.4	25.7	2.1	5.3	<MDA		0.5	788	42	10
45	Svenskärs fiskeläge	FR	2020-05-07	M666	5	14.4	0.7	1.2	<MDA		1.4	24.5	2.1	5.5	4.0	1.8	0.5	696	43	10

Table A2 ICP-MS data in *Fucus* spp. and grass, including details of sampling and sampling preparation conditions. N.D. – No data. *From ^{114}Cd. **From ^{111}Cd.

Site no	Site name	Sampling date YYY-MM-DD	Lab code	Co (ppb)	%RSD	Ni (ppb)	%RSD	Cd* (ppb)	%RSD	Cd** (ppb)	%RSD	Gd (ppb)	%RSD	Hg (ppb)	%RSD	Pb (ppb)	%RSD
1	Båteviken, Strömstad	2020-04-08	KS-1	2522	5.5	5972	7.4	1121	9.4	1088	11	223	4.1	21	98	346	5.3
2	Fjällbacka, Sälvik, north of harbour, with ongrowth	2020-04-08	KS-3	2134	5.5	6090	7.6	1021	6.5	981	9.4	237	5.4	16	19	308	5.9
2	Fjällbacka, Sälvik, harbour and south of harbour, no ongrowth	2020-04-08	KS-2	2777	5.4	5662	8.1	853	4.8	880	4.8	393	6.7	13	19	647	8.0
3	Smögen, Kleven	2020-04-08	KS-4	1176	4.2	5076	11	1087	5.4	1081	4.0	155	3.7	16	44	3941	5.0
4	Stockevik, Tjörn	2020-04-08	KS-5	1838	5.8	5063	7.1	994	7.1	1046	11	526	5.4	14	20	1775	7.3
5	Hällsvik	2020-04-08	KS-7	2354	5.5	5133	2.7	1130	7.1	1156	6.9	730	6.4	27	14	3610	8.5
6	Smarholmens badplats	2020-03-31	KS-8	1022	4.7	7545	6.2	1170	7.5	1196	6.7	61	12	12	65	161	9.2
7	Frillesås	2020-03-31	KS-13	5570	2.9	6131	8.6	1558	3.0	1497	7.4	657	3.4	19	13	1079	4.1
8	Sallebacka	2020-03-31	KS-6	1761	4.0	7374	5.2	1090	5.8	1110	7.4	164	8.9	18	19	426	5.5
9	Ringhals, Gloppen	2020-03-31	KS-9	1656	7.2	8592	6.9	1285	7.7	1257	8.0	179	7.4	15	13	388	7.2
10	Videbergshamn (Ringhals)	2020-03-31	KS-10	2962	8.0	4470	5.4	893	7.3	919	6.3	810	5.6	24	9.4	1066	5.6
11	Bua (Ringhals)	2020-03-31	KS-11	2153	4.4	6684	4.4	1281	7.0	1272	5.3	361	4.6	18	20	546	8.1
12	Skeatången, Årnäshalvön	2020-03-31	KS-12	3223	3.2	3919	3.4	1063	6.8	1058	5.6	906	6.0	15	16	811	5.2
13	Getterön (M630)	2020-03-24	KS-14	1567	5.4	6273	7.0	918	7.4	904	5.1	71	6.7	18	27	183	5.0
14	Glommen (M629)	2020-03-24	KS-15	2198	5.5	7596	8.1	1017	5.7	1005	6.5	116	8.0	18	19	335	8.4
15	Särdal A	2020-03-03	KS-16	1436	2.6	7100	7.3	505	6.5	513	7.8	115	9.3	19	17	370	6.2
15	Särdal b	2020-03-03	KS-17	861	6.7	2858	8.7	653	5.8	648	5.6	66	10	17	28	210	5.6
16	Kattvik	2020-04-20	KS-18	1021	3.7	6226	6.5	886	8.1	855	5.2	47	8.6	14	12	163	8.4

17	Mölle	2020-04-20	KS-19	670	5.1	7110	8.6	958	14	951	8.8	21	12	12	26	159	6.8
18	Ålabodarna	2020-03-25	KS-20	1383	5.9	7513	5.6	1329	8.8	1327	9.9	29	9.0	10	18	308	6.5
19	Barsebäcks camping	2020-03-24	KS-21	1322	4.9	7969	8.6	938	7.6	879	8.3	62	10	11	22	503	5.7
20	Barsebäck Skansen	2020-03-24	KS-23	914	6.1	7392	6.3	1018	7.9	1031	9.4	38	8.1	9	46	190	8.5
21	Barsebäck between NPP and Skansen	2020-03-25	KS-22	1678	4.5	6052	2.8	662	8.6	670	7.9	95	6.0	9	23	624	6.3
22	Barsebäck north of NPP	2020-03-25	KS-24	1796	6.7	6470	5.4	876	6.5	822	6.5	68	8.1	9	33	350	9.1
23	Vikhög fort	2020-03-25	KS-25	1307	3.9	5665	5.1	909	10	950	10	93	9.8	16	66	405	6.8
24	Vikhög hamn	2020-03-25	KS-26	1691	2.7	4982	2.9	791	6.4	812	5.6	273	9.6	15	64	853	6.7
25	Gamla Bjärred, Pilevägen	2020-04-07	KS-27	1584	4.6	4934	6.8	1320	9.0	1318	7.7	154	7.4	16	50	693	8.0
26	Lomma	2020-03-24	KS-28	1645	5.2	7523	3.2	1039	9.0	1083	6.9	74	13	12	65	347	9.2
27	Limhamn	2020-03-24	KS-29	996	7.0	6998	3.2	1533	6.3	1526	7.6	101	14	12	68	1399	6.4
28	Lilla Hammar	2020-03-24	KS-30	603	5.5	1348	3.1	1236	9.5	1214	6.4	16	15	5	114	105	20
29	Smygehuks fyr	2020-03-11	KS-31	1437	4.2	4779	6.6	985	7.0	996	8.9	44	14	17	40	293	8.1
30	Skillinge, 1A	2020-03-11	KS-32	3447	3.7	8437	4.5	940	6.2	895	8.3	161	5.5	8	83	921	7.6
30	Skillinge, 2A	2020-03-11	KS-33	2881	4.4	10421	6.3	1009	8.6	1074	8.0	94	6.7	7	97	557	4.7
30	Skillinge	2020-05-13	KS-34	2181	5.7	6923	2.5	806	7.9	866	6.4	142	7.4	9	79	793	7.1
31	Hasslö	2020-03-19	KS-35	1972	3.7	8477	3.3	1138	7.6	1149	9.7	434	6.8	8	95	793	6.8
32	Sandvik	2020-04-22	KS-36	2781	8.1	4446	5.5	941	9.5	1093	9.2	714	6.8	16	89	1673	6.5
33	Dragskär	2020-04-22	KS-37	385	4.5	11237	5.1	1482	7.9	1499	6.6	44	17	7	94	155	7.4
34	Simpevarp Åkvik	2020-04-22	KS-38	784	4.3	5957	4.9	798	9.1	828	6.1	419	6.4	9	90	265	11
35	Kråkelund	2020-04-22	KS-39	1592	4.5	5937	3.6	1151	8.0	1209	7.1	179	8.9	5	107	348	6.8
36	Händelöp	2020-04-22	KS-40	886	5.0	10705	6.6	1124	5.1	1093	5.8	87	8.6	5	115	205	12
37	Jogersö	2020-05-08	KS-41	1817	3.6	4126	8.6	906	7.1	1021	5.2	504	4.5	10	60	881	8.7
38	Senneby, Sandviken, Väddö	2020-05-08	KS-42	1017	5.2	19110	4.3	2632	7.5	2676	5.2	175	9.4	8	129	384	5.5
39	Öregrund	2020-05-07	KS-43	1983	4.7	20736	5.4	3219	5.8	3258	6.7	229	6.7	8	87	463	8.0
40	Valudden	2020-05-08	KS-44	1559	4.8	11537	6.4	2158	8.6	2230	5.5	253	6.5	5	160	461	5.6

41	Stenskär	2020-05-08	KS-45	1999	3.9	9179	3.9	1628	7.5	1785	6.6	578	6.1	8	81	967	3.1
42	Norrboda, Gräsö, A	2020-05-08	KS-46	1197	3.5	14751	5.0	2378	8.5	2340	6.8	169	5.4	10	85	297	9.1
42	Norrboda, Gräsö, B	2020-05-08	KS-47	1042	7.1	26045	5.4	2389	4.3	2505	4.9	121	6.4	4	147	141	10
43	Bönan	2020-05-07	KS-48	1179	1.7	6619	6.1	1805	6.2	1839	5.2	439	5.7	9	109	1708	7.3
44	Hölick	2020-05-07	KS-49	1018	5.0	7413	4.6	2353	4.8	2451	5.5	395	7.2	9	88	540	5.8
45	Svenskärs fiskeläge	2020-05-07	KS-50	1288	6.1	6896	3.0	2571	4.7	2582	7.9	295	6.0	14	69	3157	6.5
Mean				**1725**	**5**	**7709**	**6**	**1271**	**7**	**1289**	**7**	**246**	**8**	**13**	**59**	**746**	**7**
Median				**1588**		**6790**		**1075**		**1086**		**162**		**12**		**444**	
Standard deviation				**893**	**1**	**4347**	**2**	**581**	**2**	**594**	**2**	**227**	**3**	**5**	**40**	**833**	**3**
Standard uncertainty of the mean				**126**	**0**	**615**	**0**	**82**	**0**	**84**	**0**	**32**	**0**	**1**	**6**	**118**	**0**