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Paper

Radionuclides in algae from Swedish coastal waters for over half a century

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Abstract

Samples of *Fucus serratus* and *Fucus vesiculosus* have been regularly collected at Särdal (56.76 N, 12.63E) on the Swedish west coast since 1967, for most of the time, every two months. In 2020, sampling of *Fucus* spp. was extended to 44 other locations along the Swedish west, south, and east coast for comparison with Särdal data. At seven of these locations, water samples and extra samples of *Fucus* spp. were also taken for ^{3}H analysis. Measurements have been performed by gamma spectrometry, or by radiochemical separation followed by low background beta measurements, alpha spectrometry or accelerator mass spectrometry. Time variations of the measured concentrations of various radionuclides (^{3}H, ^{14}C, ^{60}Co, ^{99}Tc, ^{129}I, ^{131}I, ^{134}Cs, ^{137}Cs, ^{236}U, ^{238}U, ^{239}Pu, and ^{240}Pu) are reported. The concentrations have been correlated with emissions from the nuclear fuel reprocessing facilities at Sellafield (UK) and La Hague (France). Contributions from the nuclear power plants in Ringhals and Barsebäck have also been identified, as has fallout from Chernobyl still stored mainly in the sediments and water of the Baltic Sea. In recent decades, studies have shown increasing levels of ^{129}I and ^{236}U and decreases in ^{137}Cs, ^{99}Tc, and $^{239+240}$Pu concentrations over time. The ^{14}C analyses show an impact of anthropogenic ^{14}C from activities other than atmospheric nuclear weapons tests in the 1950s and 1960s. The considerable variation in the concentration of different radionuclides over time and along the coasts warrants further studies to determine the possible origin of these radionuclides and to map background data in the event of future releases.

Introduction

Marine macroalgae do not only retrieve nutrients from water. They are also known to efficiently absorb waterborne pollutants, such as different radionuclides and heavy metals [1] and are thus good bioindicators, e.g. of anthropogenic radionuclides in the marine environment. In particular, algae that are permanently attached to rocks and stones, such as the brown algae *Fucus serratus* (toothed wrack) and *Fucus vesiculosus* (bladderwrack), reflect environmental conditions for long periods and are relatively easy to collect. Uptake, turnover and retention of the pollutants in the algae depend on the chemical properties of the substances, their concentration in the water and many other parameters such as algae uptake mechanisms, growth rate, light intensity, temperature, and salinity of the water. Knowledge of past and present concentrations of natural as well as anthropogenic radionuclides in the marine environment is of great importance in assessing values measured in connection with future additions, e.g. from discharges of radionuclides from nuclear installations, and from nuclear and non-nuclear accidents and emergencies. The fact that products based on algae are used in various foodstuffs as thickening agents and in health

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food supplements further emphasizes the significance of knowledge of potentially hazardous elements and compounds and their concentrations in algae. Furthermore, being rich in potassium, nitrogen and phosphorus, algae have since long been used as fertilizers in small-scale sustainable farming and gardening. This is yet another pathway from which pollutants, radioactive as well as non-radioactive, accumulated in algae, can reach humans.

In this paper we report temporal variations of the concentrations of various radionuclides (^{3}H, ^{14}C, ^{60}Co, ^{99}Tc, ^{129}I, ^{131}I, ^{134}Cs, ^{137}Cs, ^{236}U, ^{238}U, ^{239}Pu, and ^{240}Pu) in *Fucus* spp. collected at the same sampling site, Särdal (56.76 N, 12.63E), at the Swedish west coast for more than half a century. Parts of the Särdal datasets have previously been published in separate papers [2–20]. This paper now presents additional, recent data and summarizes and discusses all the findings. What is new is: all results for ^{236}U, ^{238}U, ^{129}I, and ^{134}Cs, results for ^{99}Tc during the last 20 years 2004–2023, updated ^{14}C data from Särdal and the whole west and south coast of Sweden, results for ^{60}Co from 1984 till now, results for ^{137}Cs from 2006 to 2022. Additional data on naturally occurring radionuclides and stable elements in the Särdal samples can be found elsewhere [17, 18, 20].

Apart from the Särdal data, the paper also presents activity concentration of ^{137}Cs in *Fucus* spp. collected along the Swedish west-, south-, and east-coast in 2020 (data on ^{14}C in these samples has been presented earlier [19] as has data for gamma-emitters and stable elements [20]) and of ^{3}H in *Fucus* spp. and in sea water collected at seven places on the west coast north and south of Särdal in 2020 [17].

Some information regarding the various investigated radionuclides is given below.

Tritium ($T_{1/2}$ = 12.32 years) is a naturally occurring cosmogenic radionuclide that is produced by interactions of cosmic-ray particles with nuclei of nitrogen and oxygen in the stratosphere.

Carbon-14 ($T_{1/2}$ = 5730 years) is produced naturally in the stratosphere by nuclear reactions of atmospheric nitrogen with cosmic rays' thermal neutrons. It mainly resides as CO_2 in the atmosphere from where it enters the marine compartment as dissolved inorganic carbon (DIC). Carbon-14 in DIC is transferred to *Fucus* spp. and other algae through photosynthesis. The main anthropogenic sources of ^{14}C are past atmospheric testing of nuclear weapons and the nuclear industry.

Technetium-99 ($T_{1/2}$ = 210 000 years) is a fission product produced in amounts comparable to ^{90}Sr and ^{137}Cs, is released into the environment primarily as a result of nuclear fuel reprocessing. Technetium-99 is transported over long distances in oxygenated seawater as highly soluble pertechnetate TcO_4^-.

Iodine-129 is a long-lived cosmogenic radionuclide ($T_{1/2}$ = $15.7 \cdot 10^6$ years) for which the natural abundances have been significantly affected by anthropogenic actions. Iodine-129 has been produced during the last 60 years during fission processes associated with civil and military nuclear activities, accidents, and disasters, such as Chernobyl (1986). Despite capture systems at reprocessing plants for spent nuclear fuel, ^{129}I (along with other radionuclides such as ^{85}Kr, ^{14}C and ^{99}Tc) is released into the environment. The ^{129}I abundance in the environment come from emissions by the two major European nuclear fuel reprocessing plants, Sellafield (UK) and Cap de La Hague (France), which after 1985 have been the dominant contributors [21, 22].

Uranium-236. Significant amounts of ^{236}U ($T_{1/2}$ = $23.4 \cdot 10^6$ years) have been released into the environment by human nuclear activities. ^{236}U is mainly produced by thermal neutron capture reactions ^{235}U(n,γ)^{236}U in nuclear reactors and nuclear explosions, and in fast neutron reactions ^{238}U(n,3n)^{236}U in thermonuclear detonations [23, 24].

Plutonium is formed by the capture of neutrons in ^{238}U in nuclear fuel. Part of the ^{239}Pu and ^{240}Pu that is formed is burned (through nuclear fission) on site while the fuel remains in the reactor and thus contributes to energy production. Plutonium-239 and ^{240}Pu cannot be chemically separated and their peaks cannot be resolved by alpha spectrometry.

Material and methods

Sampling sites

The sampling site Särdal (56.76 N, 12.63E) is located on the west coast of Sweden, at Kattegat (Fig. 1). Särdal is situated in the countryside between the two cities Halmstad (56.7 N, 12.9E; 18 km SE of Särdal) and Falkenberg (56.9 N, 12.5E; 20 km NNW of Särdal). Two major rivers, Nissan and Ätran, are of particular relevance for the Särdal site. The river Nissan flows into the Kattegat in Halmstad (average water discharge rate ~ 41 m^3/s) whereas the river Ätran has its mouth in Falkenberg (average discharge rate ~ 47 m^3/s). Suseån (~10 km north of Särdal has an average discharge rate of ~9 m^3/s) [25].

The sampling site Särdal is heavily exposed to wind and waves. The seabed in the area is dominated by stones, with gravel and sand in between, often overgrown with *Fucus* spp. Three smaller drainage ditches with low water flow open in the area.

As can be seen in Fig. 1, which also shows marine water currents, the Kattegat surface water is influenced by brackish Baltic Sea water and its bottom water by saline North Sea water. Figure 1 also includes some facilities associated with significant radioactive

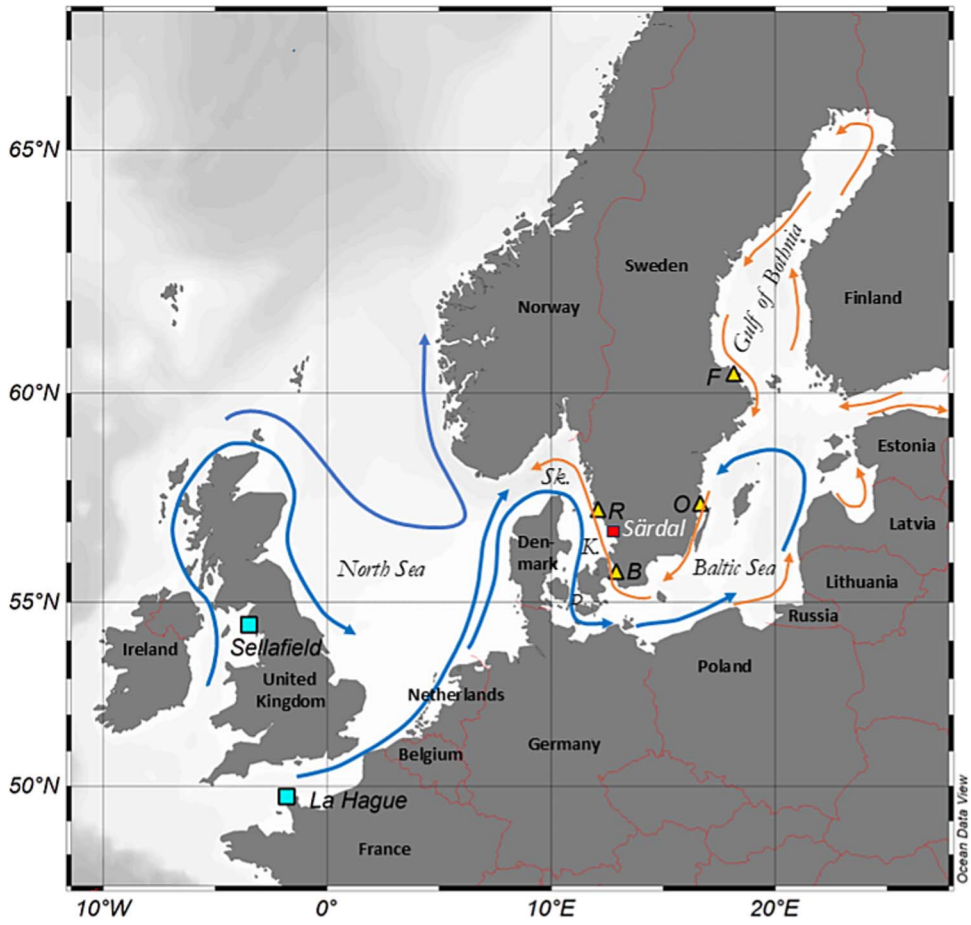

Figure 1. Map including the sampling place at Särdal (56.76 N, 12.63E) on the Swedish west-coast regularly used since 1967. Major ocean currents (adapted from Qiao *et al.* [27] and Qiao *et al.* [28]) are indicated (surface water movement in thin water flow arrows and bottom water movement in thick water flow arrows). The Swedish nuclear power plants (R: Ringhals, B: Barsebäck, O: Oskarshamn and F: Forsmark), and the two nuclear facilities at Sellafield (UK) and La Hague (France) are also marked. The site Winfrith (W) in UK, previously hosting research reactors and performing reactor development, is under decommissioning. Several other sites for potential releases of radioactive discharges to water exist in the area [21, 26]. In addition to those inputs, the Baltic Sea is exposed to other potential local and/or regional sources, as e.g. the controlled or accidental discharges from nuclear facilities (e.g. historical releases from the Swedish nuclear facility at Studsvik (58.77 N, 17.39E) have been recently studied [29]). There is also a number of radioactive dumping sites reported all around the Baltic Sea. Most of them not well-documented. *Sk.*, Skagerrak; *K.*, Kattegat; *D.,* Danish straits. Barsebäck NPP is located at the Öresund Strait (Öresund Strait is not marked in the figure). Map: Schlitzer, Reiner, ocean data view, https://odv.awi.de, 2021 [30].

liquid releases like the reprocessing facilities for spent nuclear fuel at Cap de La Hague, France, and Sellafield, United Kingdom, that are of relevance for the Swedish west coast. Furthermore, the Swedish nuclear power plants, all light-water reactors, are shown in Fig. 1 (*R*: Ringhals; *B*: Barsebäck; *O*: Oskarshamn and *F*: Forsmark). Ringhals nuclear power plant (57.26 N, 12.11E, Kattegat) is located 62 km NNW of Särdal, and has one boiling water reactor (R1, 881 MW_e, in operation from 1976 to the end of 2020) and three pressurized water reactors (R2, 900 MWe, in operation from 1975 to the end of 2019; R3, 1070 MW_e, in operation since 1981; and R4, 1120 MW_e, in operation since 1983). The other Swedish nuclear power plant of relevance to the Särdal site is Barsebäck (55.74 N, 12.92E; ~115 km south of Särdal) at the Öresund Strait. Barsebäck nuclear power plant had two boiling water reactors (each 600 MW_e) which were in operation from 1975 to 1999 and from 1977 to 2005, respectively. Other potential sources of radioactive discharges that may be of relevance for the Särdal site can be found at the homepages of HELCOM [26] and OSPAR [21] and in [28].

In 2020, 44 other sampling sites along the Swedish west-, south-, and east-coasts were included for a study focusing on spatial variations of ^{14}C in the marine

environment [19]. Data on ^{137}Cs in *Fucus* spp. collected at these sites are now also presented (data on other gamma emitters and metals can be found elsewhere [20]).

Since 2020, additional samples of *Fucus* spp. as well as of sea water have been collected for ^{3}H analysis at Särdal and some other places (Torekov (56.42 N, 12.62E), Laxvik (56.60 N, 12.92E), Tjuvahålan at Tylösand (56.64 N, 12.73E), Glommen (56.93 N, 12.35E), Getterön (57.12 N, 12.21E), Gloppe (57.27 N, 12.11E), and Gårda brygga (57.36 N, 12.08E) covering a distance of 140 km along the Swedish west-coast, south and north of Särdal. These results will now also be presented.

Sampling and sample preparation

Samples of *F. serratus* and *F. vesiculosus* have been regularly collected at the same place in Särdal by the same person since 1967. "Regularly" means for most of the 57 years' time (1967–2024) "every other month". Separate samples of *F. serratus* and *F. vesiculosus* have been collected each time.

The algae, growing on rocks or stationary bottom stones, were collected by hand from a water depth of 0.3–0.9 m. The salinity of the water ranged from 15‰ to 23‰ at sampling. The salinity peaked in January–March. More than 25 plants per sample were usually collected. Individuals with visible surface growth of other algae were avoided. After run-off and drying in air, the samples were dried at 70°C, ground into a fine powder and homogenized. The dry-weight to wet-weight (at sampling) ratio was normally between 24 and 27%.

For the *Fucus* spp. samples from 44 other sites at the Swedish coast and collected 2020, further information on sampling strategy, sampling dates and sample pre-treatment has been published together with the ^{14}C results [19]. Additional samples from some of these sites were collected between 24 September and 15 October 2020. At some sites, grass was also collected to compare terrestrial and marine levels of ^{14}C.

The dried and grounded algae material was stored in plastic jars and archived in a locked and well-ventilated room until used for analyses. All subsamples were first dried overnight at 70 or 80°C to remove any water adsorbed during storage and to get the dry weight.

Analysis of ^{137}Cs, ^{134}Cs, ^{131}I, ^{60}Co, and other γ-emitters

The dried and grounded algae were put in 200 ml plastic containers and gamma spectrometric measurements were performed using different HPGe-gamma spectrometers in lead shields [17, 31] at the Department of Medical Radiation Physics in Malmö. In the current project a detector (ORTEC, USA) with the relative efficiency of 55% has been used for most of the measurements performed since 2018. The measurement time was typically 24–48 hours. Pulse height distributions obtained for each sample were evaluated using an in-house developed excel evaluation sheet, which was cross-checked with the automatic spectrum analysis program in the software GammaVision 7.1™ (ORTEC, USA). Solutions with certified activity concentrations of different gamma emitters (Eckert & Ziegler Isotope Products, Type 7603, Valencia, CA 91355, U.S.A.) were used for mixing in different materials for determination of the detection efficiency for various radionuclides and sample densities. Samples were analysed alongside certified reference materials of algae such as the IAEA-446 (Baltic Sea Fucus, IAEA) for quality assurance purpose. The laboratory has regularly participated in intercomparison programs such as the IAEA-TERC proficiency test or the Procorad intercomparison for gamma emitters and tritium to validate our analytical methods.

Analysis of ^{3}H

Sea water samples for ^{3}H analysis were collected in plastic bottles. Several hundred grams of *Fucus* spp. were collected and frozen to −18°C in sealed plastic bags. Water was extracted from the algae by freeze-drying, and collected in plastic bottles. The sea water samples as well as the water extracted from the algae were distilled and split in two to duplicate the measurements. Liquid scintillation counting (LSC) was performed using a Beckman LS 6500 multi-purpose LSC [17]. With 10 ml of water mixed with 10 ml the Ultima Gold LLT (PerkinElmer, USA) scintillation cocktail and measured for 10 h to minimum detectable activity concentration of ∼1.5 Bq L^{-1}.

Analysis of ^{14}C

A few mg of dried and grounded *Fucus* spp. from most years since 1967 were submitted to the Radiocarbon Dating Laboratory at Lund University for analysis of ^{14}C. Sample preparation and analysis of ^{14}C at the Single Stage Accelerator Mass Spectrometry facility at Lund University was performed as earlier described [19]. Analytical uncertainties were reported at one standard deviation. The obtained ^{14}C data are reported as Fraction Modern, F^{14}C([19, 32] and references therein). F^{14}C = 1 corresponds to a specific activity of ∼228 Bq (kg C)$^{-1}$.

Analysis of ^{99}Tc

The ^{99}Tc content of the samples from the period 1967–2010 was determined at Department of Medical Radiation Physics in Malmö according to the method described by Lindahl *et al.* [14]. Samples for the period

2011–2024 were extracted and analysed by liquid scintillation counting by the Service de Mesure et d'Analyse de la Radioactivité et des éléments Traces (SMART, part of the UMR 6457 SUBATECH) in Nantes, France.

Analysis of ^{129}I

The samples were analysed regarding their ^{129}I concentration at Centro Nacional de Aceleradores (CNA), University of Seville, Spain. Sample preparation was based on the method published in Schmidt *et al.* [33] and later evaluated by Szidat *et al.* [34]. In summary, 0.15 g of dry sample, joined to 0.6 g NaOH and 4 mg Woodward iodine carrier dissolved in Milli-Q water were added to 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 450°C, using a slow heating ramp. After this, the melt was extracted and centrifuged to discard solid. The supernatant was transferred to a bottle and Milli-Q water was added until a volume of 100 ml, then adding 1 ml 12% NaOCl. After shaking the sample for 1 h it was kept in the dark overnight. Next day, 5 ml 1 M $Na_2S_2O_5$ and 1 ml 1 M $NH_2OH{\cdot}HCl$ were added and the sample was shaken for 1 h to adjust all iodine species. Then iodine purification was carried out by a double liquid–liquid extraction. First iodine was extracted twice in 100 ml of Cl_3CH previously oxidized with 2 ml 0.45 g/l $NaNO_2$ in acidic environment. Second, it was back extracted in a reducing aqueous solution (containing 0.7 ml of 80% NH_2-NH_2 + 30 ml H_2O-MQ). Finally, AgI was precipitated by adding $AgNO_3$ solution, dried, mixed with Nb powder for AMS target preparation and measured by Accelerator Mass Spectrometry (AMS) at CNA. In this technique, I^- ions are extracted from the sample in a Cs sputter ion source, and then injected in a 1 MV Tandetron accelerator (High Voltage Engineering Europe, Netherlands). After passing through a He stripper, I^{2+} was selected in a magnetic deflector. While stable $^{127}I^{2+}$ were detected as a current in a Faraday cup, $^{129}I^{2+}$ ions went through an electrostatic deflector and was detected in an ionization chamber. Standard samples with known $^{129}I/^{127}I$ were continuously measured to correct the isotopic ratio, as well as blank samples (Woodward iodine) for background evaluation. The instrumental background was typically $^{129}I/^{127}I = 1–2{\cdot}10^{-13}$, while samples showed ratios in the order of 10^{-12} to 10^{-10}. The CNA has also carried out tests with reference materials such as IAEA-446, obtaining results that ratify the validity of the methodology. More details on the AMS facility can be found in Chamizo Calvo *et al.* [35] and Scognamiglio *et al.* [36].

Analysis of ^{236}U, ^{239}Pu and ^{240}Pu

The samples were analysed for $^{239+240}$Pu using radiochemistry and α-spectrometry as described earlier [17]. For some samples separate determinations of ^{239}Pu and ^{240}Pu as well as ^{236}U were done on the 1 MV AMS at the CNA. About 10 g of dry weight of algae were spiked with ~1 pg of ^{242}Pu and ^{233}U, calcined at 650°C and leached with *aqua regia*. Actinides were then coprecipitated with $Fe(OH)_2$, and actinides species sequentially purified using TEVA® (Pu) and UTEVA® (U) cartridges in tandem in a vacuum box, following the guidelines given in López-Lora *et al.* [37]. AMS measurement procedures have been described elsewhere (i.e. see Chamizo and López-Lora [38] for ^{236}U and López-Lora and Chamizo [39] for ^{239,240}Pu). Briefly, actinides were extracted as monoxide ions from a Cs-sputter ion source (e.g. UO^-), mass analysed on a 90° sector magnet, stripped to 3+ in He gas at the accelerator terminal at 650 kV (e.g. UO^-/U^{3+}), mass and energy analysed in a 90° sector magnet and a 120° electrostatic deflector, and finally, rare isotopes counted from the total energy signal provided by a gas ionisation chamber.

As for ^{239,240}Pu and ^{236}U, the reliability of the AMS analysis method has been widely demonstrated through the analysis of IAEA reference materials in different publications. Very recently, CNA has reported results for the seaweed sample IAEA-446 [37]. The CNA has also participated in the certification process of different IAEA- Reference Materials [40, 41].

Results and discussion

Radionuclides in *Fucus* spp.

Gamma emitters

^{137}Cs. The temporal variation of the ^{137}Cs ($T_{1/2}$ = 30.2 years) activity concentration in *F. serratus* and *F. vesiculosus* from Särdal is shown in Fig. 2. In the period 1967–1979, most of the ^{137}Cs is assumed to originate from the nuclear weapons tests in the atmosphere with some contributions from Sellafield. The following unexpected increase, peaking in 1981, came 4 years after a peak release of ^{137}Cs from Sellafield [2, 12, 15]. The discharges from Sellafield (Fig. 3) have steadily decreased since the second and larger peak ending in the mid-1980ies, but contaminated sediments in the Irish Sea from the large discharges in the 1970s and 1990s may have been remobilized and have been estimated to do so for a long time [42–44]. The discharges from La Hague were two orders of magnitude lower than from Sellafield [21, 45–47]. A smaller reprocessing facility also existed until the middle of the 1990s in connection with the research reactor in Dounreay (58.58 N, 3.75E), United Kingdom. Compared with the

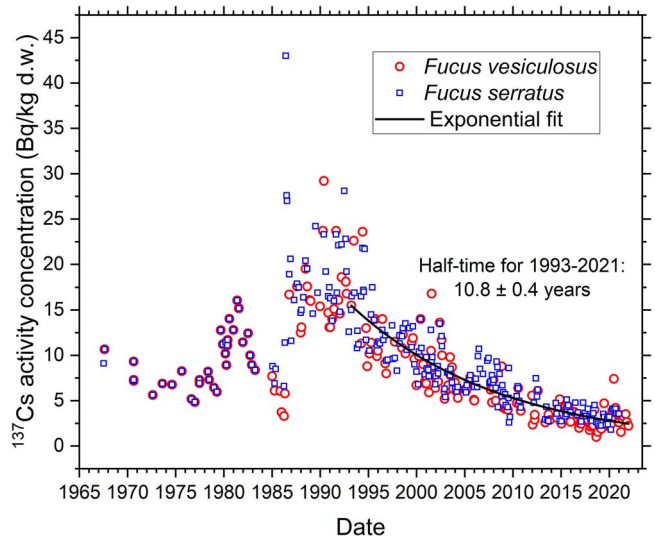

Figure 2. ^{137}Cs-activity concentration (Bq/kg d.w.) in *Fucus serratus and F. versiculosus* from Särdal (56.76 N, 12.63E) on the Swedish west-coast during the period 1967–2021. The uncertainty (1 SD) in the individual measurements is typically ±0.3 Bq/kg d.w.

discharges from the other two reprocessing plants, the discharge from Dounreay was small [48].

The Chernobyl disaster contributed with an immediate peak in the ^{137}Cs-concentration in 1986, followed by a broader peak around 1990 interpreted as the effect of Sellafield releases and an inflow of water from more Chernobyl contaminated areas and especially from the Baltic Sea [12]. The subsequent decrease is much slower than after the 1979–1983 Sellafield peak indicating a continuous inflow of ^{137}Cs from the Baltic Sea. In 2000, the annual net outflow from the Baltic Sea was estimated to 39 TBq [49].

For the period 1987–1994 similar results are reported from Klint (55.97 N,11.58E) in southwestern Kattegat [27]. We have not seen any report in the literature corresponding to the 1979–1983 peak in *Fucus* spp. from Särdal (most had not started measurements on *Fucus* spp. at that time). Measurements on water from Klint suggest an increase in ^{137}Cs levels in the current time window [27].

HELCOM [50] estimated an effective half-time of 9.6 years for ^{137}Cs in Baltic Sea water during the period 1993–2006. In the period 1993–2021, the ^{137}Cs concentration in *Fucus* spp. from Särdal shows—as estimated from Fig. 2—an effective half-time of 10.8 ± 0.4 years. This supports the hypothesis that the majority of the west coast's ^{137}Cs after 1986 originates from the Baltic Sea. In addition to contributions of ^{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.

Assuming a ^{137}Cs concentration of 65 Bq/m^3 of water in the Kattegat in the period 1988–2001 [50], the concentration factor (L/kg wet weight) can be estimated to 60 for *F. serratus* and 50 for *F. vesiculosus*, which is similar to e.g. which was found by Qiao *et al.* from measurements of ^{137}Cs in both seaweed and water in the Danish Straits [27].

From the study of the ^{137}Cs concentration in *Fucus* spp. collected in spring 2020 along the Swedish coast, the ^{137}Cs-concentration in *Fucus* spp. was significantly higher on the east coast than on the west coast (Fig. 4). This observation provides further evidence that Chernobyl-^{137}Cs residing in the Baltic Sea is a source of ^{137}Cs at Särdal. However, it should be noted that the uptake of ^{137}Cs in *Fucus* spp. increases with decreasing salinity [52].

As seen in Fig. 4, the ^{137}Cs activity concentrations were generally lower in the beginning of the sampling period (March) than at the end of the sampling period (May). The Baltic Sea site Skillinge (sampled in March as well as in May) shows almost a doubling of ^{137}Cs in *Fucus* spp. from March to May. The same is valid for Särdal, where the ^{137}Cs activity concentration (mean for *F. serratus* and *F. vesiculosus*) almost doubled from 2.1 to 4.0 Bq/kg d.w. from March 3 to May 2, 2020. Such seasonal variations of ^{137}Cs in *Fucus* spp. have been observed since the beginning of the studies [2, 12, 15].

For ^{137}Cs, the seasonal variation is supposed to rely on biological factors as light, temperature and salinity

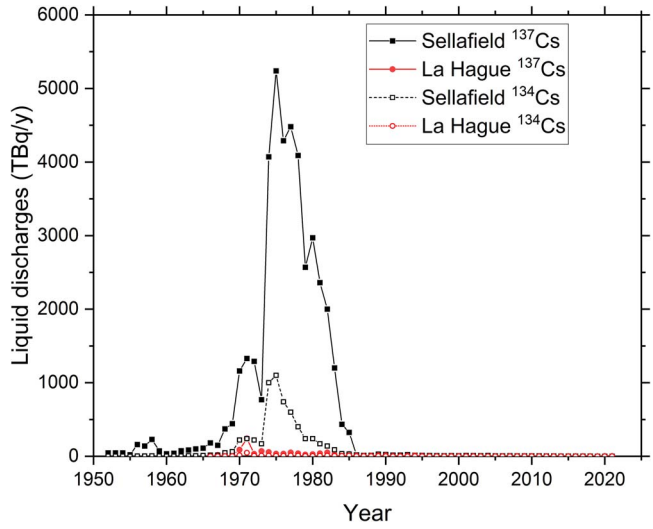

Figure 3. The annual releases of ^{134}Cs and ^{137}Cs from the Sellafield reprocessing plant located in the Irish Sea and from the Cap de La Hague reprocessing plant in the English Channel [21, 42–47, 50, 51].

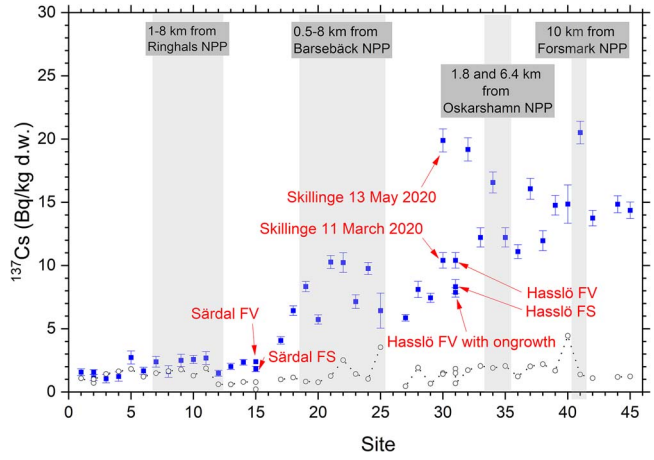

Figure 4. ^{137}Cs in *Fucus* spp. at the various sites along the Swedish coast in spring 2020. Samples collected within 10 km of the Swedish NPPs are highlighted in the grey areas. Uncertainty bars correspond to ±1 SD. Open circles: MDA. FS, *Fucus serratus*; FV, *Fucus vesiculosus*. Site numbers are the same as in [19].

to a larger extent than to the ^{137}Cs-concentration in seawater [53].

^{134}Cs. Caesium-134 ($T_{1/2}$ = 2.07 years) was registered in measurable amounts in *Fucus* spp. at Särdal in the period 1979–1983 (Fig. 5), coinciding with the two Sellafield peaks of ^{137}Cs. In the period 1986–1999, fallout from Chernobyl gave rise to measurable concentrations of ^{134}Cs. The ^{134}Cs/^{137}Cs-activity ratios at maximum levels at these two occasions were estimated to 0.08 ± 0.02 and to 0.40 ± 0.01 respectively.

At measurements on *F. vesiculosus* from south-west Sweden, Carlsson and Holm [54] found ^{134}Cs/^{137}Cs activity ratios in 1987 of 0.35 ± 0.01, which is close to the Särdal value for *Fucus* spp.

^{60}Co. The activity concentrations of ^{60}Co ($T_{1/2}$ = 5.27 years) in *F. serratus* and *F. vesiculosus* from Särdal are shown in Fig. 6. On 6 December 1986, ^{60}Co peaked in *F. serratus* (no *F. vesiculosus* sample was taken at that occasion). The source of this ^{60}Co was first considered a probable large release from Barsebäck NPP. Previous studies had demonstrated the presence of significant amounts of several neutron activation products (^{60}Co, ^{58}Co, ^{65}Zn, ^{110m}Ag, etc.) in the marine environment of Barsebäck NPP [3]. Comparing the liquid discharges

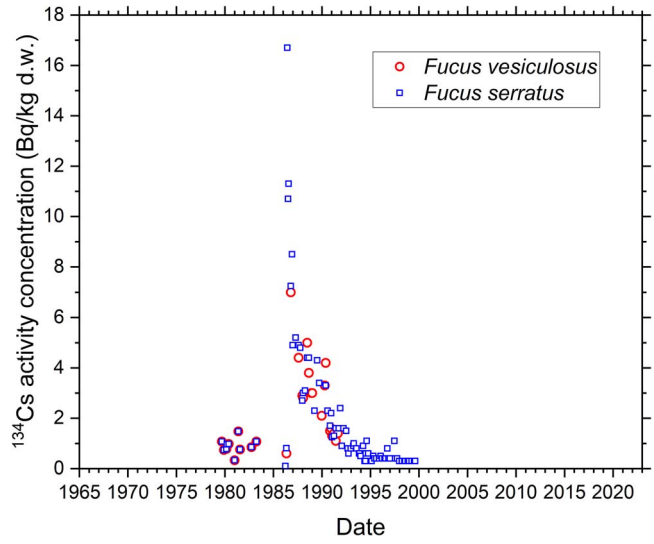

Figure 5. ^{134}Cs-activity concentration (Bq/kg d.w.) in *Fucus serratus and F. vesiculosus* from Särdal (56.76 N, 12.63E) on the Swedish west coast during the period 1967–2020. For long periods, the activity concentration was below the detection limit. The uncertainty (1 SD) in the individual measurements is typically ±0.4 Bq/kg d.w.

of ^{60}Co from Sellafield, Winfrith, La Hague and Barsebäck (Fig. 7) the most likely explanation for the largest contribution to the ^{60}Co peak level seems to be the releases from La Hague around 1984 with a 2-year delay. Contributions from the releases from the Winfrith plant (20 TBq of ^{60}Co in 1980/81 into the English Channel) cannot be excluded [55, 56].

^{131}I. Iodine-131 ($T_{1/2}$ = 8.02 days) has only been occasionally detected in the *Fucus* spp. samples and in very low activity concentrations, typically 0.5–5 Bq/kg d.w. The ^{131}I most likely originates from discharges of excreta from patients after nuclear medicine therapy with ^{131}I. Its short physical half-life in combination with the delayed discharge via sewers imply that only low activities of ^{131}I reach the marine environment.

^{3}H. For the tritiated water extracted from *Fucus* spp. samples collected in 2020 and analysed for ^{3}H by liquid scintillation counting, most values were below the estimated minimum detectable activity concentration (MDA) of 1.65 Bq/L [17]. Only three of 9 *Fucus* samples exceeded this limit with 1.67 ± 0.57, 2.11 ± 0.76 and a maximum of 2.48 ± 0.81 Bq/L. For 11 sea water samples collected during the same period, all except one (with 1.87 ± 0.99 Bq/L) show activity concentrations below the MDA. The observed ^{3}H activity concentrations for *Fucus* spp. and seawater are at the expected natural environmental level.

^{14}C. The results of the analyses of ^{14}C of ∼70 of the *Fucus* spp. samples in the Särdal series are presented as F^{14}C in Fig. 8. Some of the ^{14}C results have been published previously and are thoroughly discussed in the paper by Eriksson Stenström and Mattsson [19].

F^{14}C in the ocean is generally lower than in the atmosphere [19]. As can be seen in Fig. 8, F^{14}C in *Fucus* spp. collected at Särdal is neither well represented by atmospheric data, nor by modelled marine data. The latter is the global average of F^{14}C in ocean water down to 75 m. The habitat of *Fucus* spp. is however close to the water surface. Exchange between CO_2 in the atmosphere and the water (CO_2 becoming dissolved inorganic carbon, DIC, which is taken up by *Fucus* spp.) implies that DIC in very shallow waters may have F^{14}C values between the atmospheric curve and the modelled marine curve. However, Särdal samples collected after ∼1990 show higher F^{14}C values than the atmospheric CO_2 curve. Additionally, there is a sudden further increase starting in 1998 and another one in the mid-2010s.

The main sources of excess ^{14}C in Särdal are believed to be long-range transport of ^{14}C from the spent nuclear fuel reprocessing plants in La Hague and Sellafield, with smaller contributions from Ringhals NPP. There was an attempt [19] of source apportionment by using estimated dilution factors for Sellafield/Särdal (200) and La Hague/Särdal [40] found for other radionuclides at Särdal([19], and references therein). These dilution factors were applied to annual liquid ^{14}C water-borne releases from La Hague and Sellafield [21, 51, 64] (Fig. 9a). The resulting predicted excess F^{14}C values at Särdal were found to be of the same

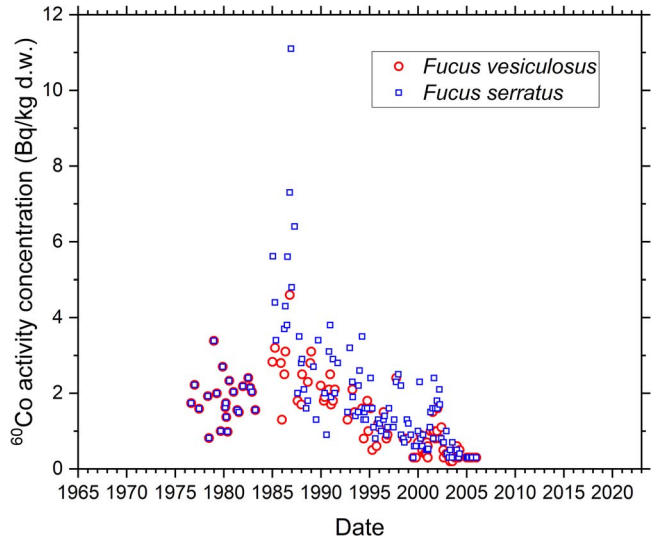

Figure 6. ^{60}Co-activity concentration in *Fucus serratus* and *F. vesiculosus* from Särdal (56.76 N, 12.63E), 1967–2000. First detected on august 21, 1976. Last time for detectable amount December 29, 2005. Barsebäck NPP was in operation from July 1, 1975 to May 30 2005. Minimum detectable activity concentration 0.3 Bq/kg d.w. The uncertainty (1 SD) in the individual measurements is typically ±0.4 Bq/kg d.w.

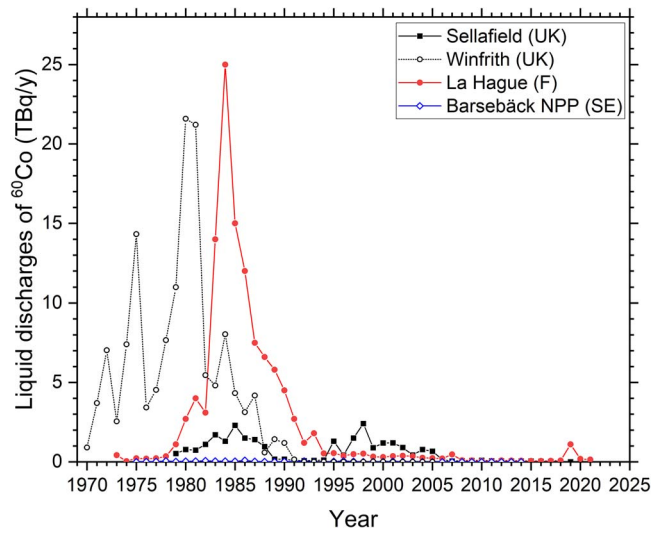

Figure 7. Liquid discharges of ^{60}Co from Sellafield, Winfrith, La Hague and Barsebäck 1973–2022 [21, 26, 55–58].

order of magnitude as the observed increased $F^{14}C$ values observed at Särdal (in comparison to a modelled marine $F^{14}C$ curve for surface water). Applying these dilution factors, as well as an estimated transport time of ~2 years from La Hague to Särdal and 4 years from Sellafield to Särdal[19], and references therein), results in a normalized inflow rate from La Hague and Sellafield of relevance to Särdal (Fig. 9b). The peaks in $F^{14}C$ in Fig. 8 (starting in 1998 and in the mid-2010s) have corresponding peaks in the normalized release data in Fig. 9b, supporting that parts of the ^{14}C found in *Fucus* spp. in Särdal are derived from La Hague and Sellafield. The observation in Eriksson Stenström and Mattsson [19] that $F^{14}C$ increases towards the north of the west coast further confirms the influence from La Hague and Sellafield (see Fig. 10).

As seen in Fig. 10, displaying $F^{14}C$ in *Fucus* spp. collected along the Swedish west coast in spring and autumn 2020 [19], samples collected close to Ringhals NPP are considerably enhanced in ^{14}C. At most sites close to Ringhals NPP, samples collected in the autumn 2020 have higher $F^{14}C$ values than those collected

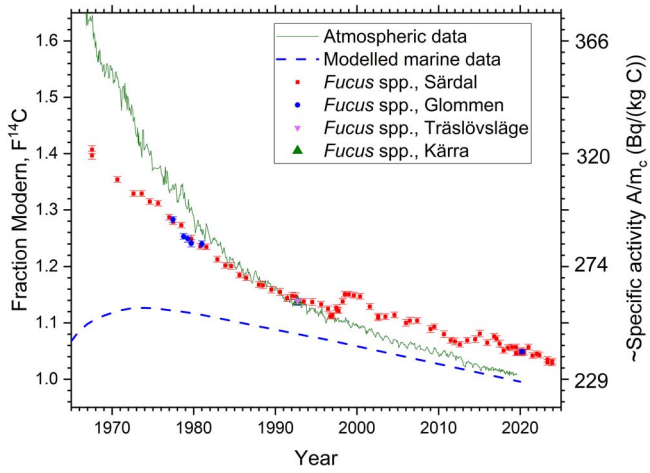

Figure 8. ^{14}C in *Fucus* spp. from Särdal in the period 1967–2023. Additional data from three other places (Glommen (56.93 N, 12.35E), Träslövsläge (57.06 N, 12.27E), and Kärradal (57.19 N, 12.21E), on the west coast are also included. The atmospheric data are representative of CO_2 of the northern hemisphere [59–63]. Prior to the atmospheric nuclear testing $F^{14}C$ in atmospheric CO_2 was ∼1 due to naturally produced ^{14}C. The modelled marine bomb pulse represents the average global oceanic water down to a depth of 75 m (data up to 1996 are from Reimer et al [63]; values from 1996 to 2020 are based on linear regression of the data from 1987 to 1996).

earlier the same year. Samples of *F. vesiculosus* (analysed part of the *Fucus* individuals: vesicles and above) collected in autumn 2020 showed higher $F^{14}C$ than *F. serratus* (top 10 cm of the sampled individuals were analysed). The difference in $F^{14}C$ between *F. vesiculosus* and *F. serratus* may be attributed to different time spans of growth and temporarily varying $F^{14}C$ in DIC due to discharges of ^{14}C from the NPP. As earlier reported [19] $F^{14}C$ in samples from the west coast are not only higher than *Fucus* spp. samples from the Baltic Sea, but also elevated above the terrestrial level (site 30, Skillinge, in Fig. 10, see also Eriksson Stenström *et al*. [65]).

^{99}Tc. The discharge of ^{99}Tc from the reprocessing facility at Sellafield increased sharply in 1970-ies as well as in 1990-ties (Fig. 11). In 2004, the implementation of a new treatment process for spent nuclear fuel led to a reduction in the discharges of 90% [66]. Previously published results for ^{99}Tc in *F. serratus* from Särdal for the period 1967–2000 [14] have indicated 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. Similar delays were observed along the Norwegian [67] and Danish [27] coasts in algae and sea water samples. Additional measurements for the period 2000–2010 carried out for *F. serratus* as well as for *F. vesiculosus* from Särdal show further decreasing concentrations. An interesting observation is the significant difference in ^{99}Tc concentration between *F. serratus* and *F. vesiculosus* with an average 68% (19–125%) higher concentration in *F. vesiculosus* (Fig. 12). This series was recently complemented by the analysis of samples for the period 2011–2024 that confirmed the decrease and the current low level of ^{99}Tc on the Swedish west coast.

^{129}I. Brown algae are known to accumulate iodine from seawater to a remarkably high degree. The ^{129}I concentration in brown algae relative to that in water has been estimated to a factor of 10,000([68] and references therein). Studies of ^{129}I in water, sediments and seaweed have provided important information on water circulation and water exchange procedures [69–72].

The first results regarding ^{129}I from this project were reported at the 5th International Conference on Environmental Radioactivity – ENVIRA 2019 in Prague, Czech Republic [16]. Since then, more samples have been analysed and the results so far are summarized in Fig. 13. The results have to be seen in the light of the reported releases from La Hague and Sellafield (Fig. 14) indicating that the main reason for the increase in ^{129}I levels at the Swedish west coast is an effect of the increased releases from La Hague. 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 ^{129}I in *F. 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 ^{129}I in the coastal

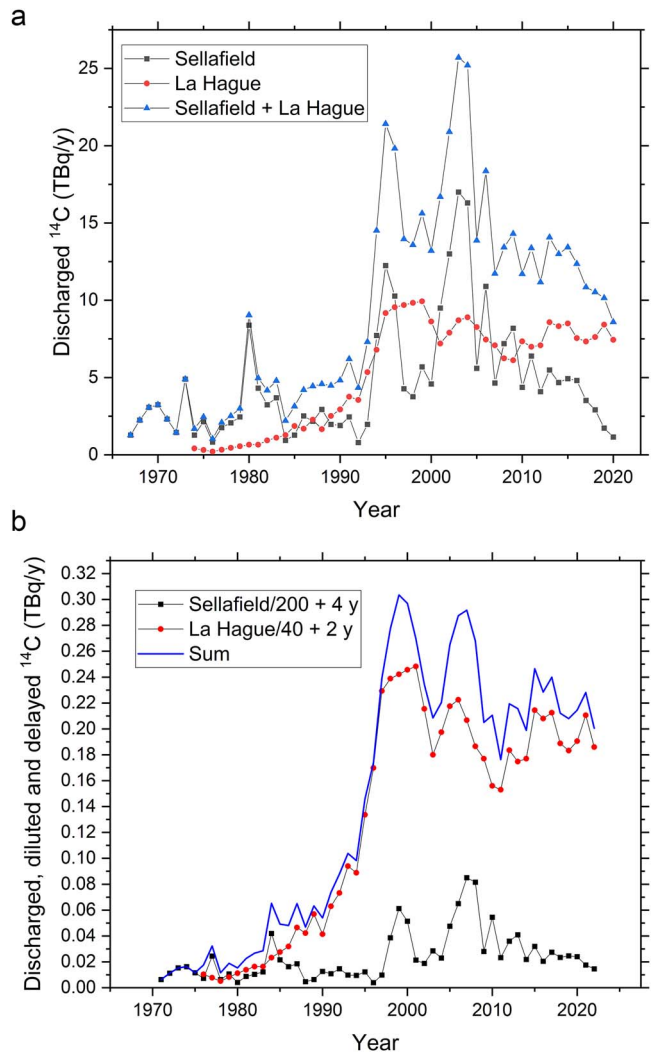

Figure 9. **a.** Annual liquid ^{14}C water-borne discharges from La Hague and Sellafield [21, 51, 64]. **b.** Annual liquid ^{14}C water-borne discharges from La Hague and Sellafield [21, 51, 64], with dilution factors and delay times according to Eriksson Stenström and Mattsson [19].

waters. Further analyses of the correlation between releases and activity concentration in *Fucus* spp. should be done.

$^{239+240}$Pu. Samples of *Fucus* spp. from the period 1967–1992 were earlier [4, 5, 13] analysed and the results are shown in Fig. 15 together with the results of the new measurements. To obtain the ratio of ^{239}Pu ($T_{1/2}$ = 24,100 years) and ^{240}Pu ($T_{1/2}$ = 6560 years), AMS measurements were made on a number of samples at Centro Nacional de Aceleradores (CNA), University of Seville, Spain.

The ^{240}Pu/^{239}Pu atom ratios due to nuclear weapons testing have been estimated to 0.178 ± 0.023 [73], from Chernobyl disaster 0.38 ± 0.07 [74] and from reprocessing plants 0.20–0.33 [75]. A first glance, the ^{240}Pu/^{239}Pu activity ratio (average from Table 1 ± 1 SD: 0.184 ± 0.014) as well as the time trend could indicate that the majority of Pu in *Fucus* spp. from the Swedish west coast originates from the testing of nuclear weapons in the atmosphere. However, the measured trend for $^{239+240}$Pu in *Fucus* spp. from Särdal is in agreement with the estimated releases of ^{236}U from Sellafield nuclear reprocessing plant according to shell data from Castrillejo *et al.* [23]. During the whole time period, releases from Sellafield dominate over the releases from La Hague [68] with maximum emissions between 1970 and 1980 and 4 years

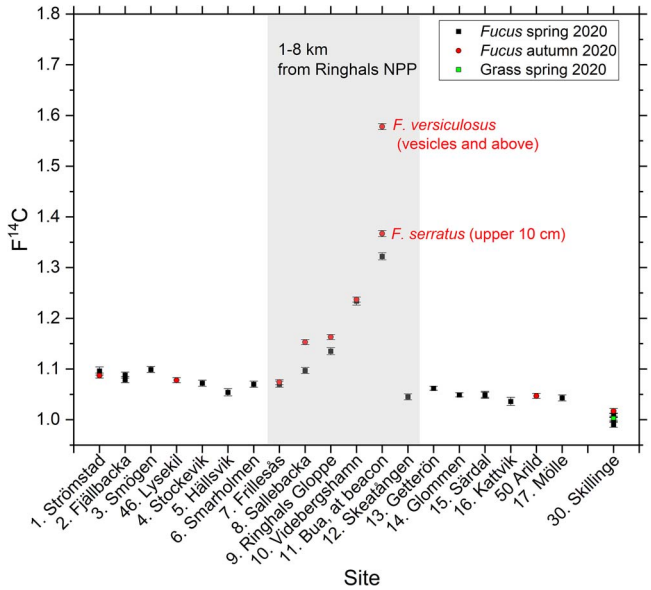

Figure 10. $F^{14}C$ values in samples of *Fucus* spp. collected along the Swedish coast in spring 2020 (11 March to 13 May) and autumn 2020 (24 September to 15 October). Site 1 (Strömstad) is the northernmost site at the Swedish west coast, located close to the border of Norway, while Site 17 (Mölle) is located in southernmost Kattegat. Site 30 (Skillinge) is a terrestrial and marine reference site located in the south east of Sweden, at the Baltic Sea [65]. At Site 11 (Bua, at beacon) *Fucus vesiculosus* as well as *F. Serratus* were sampled in autumn 2020.

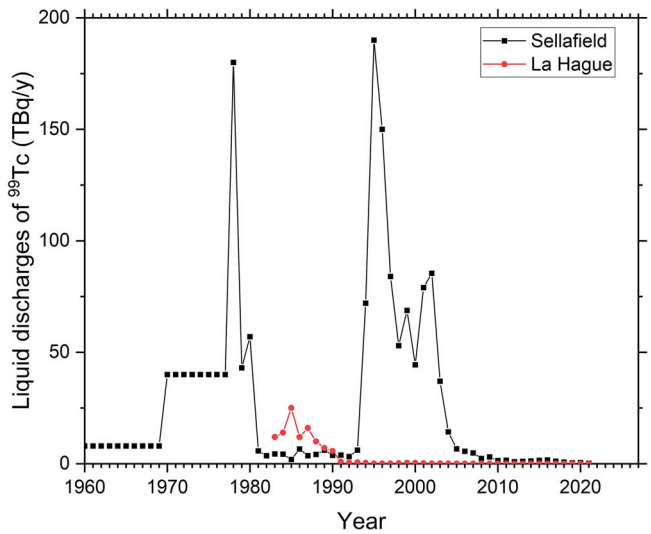

Figure 11. Reported releases of ^{99}Tc from the reprocessing facilities at Sellafield [42, 55] and La Hague [51, 66].

transport time (Fig. 16). Moreover, as is shown by Lindahl *et al.* [74], during the maximum emission peak (1970–1980), the $^{240}Pu/^{239}Pu$ ratio was between 0.15 and 0.20 and thus, could not be distinguished from global fallout. Given that only a small fraction of the Sellafield-derived Pu may have been transported in soluble form, and that this fraction was mixed with Pu from global fallout, the fact that the ratios are not higher does not contradict the hypothesis that the Pu seen in the algae from Särdal is mainly from Sellafield releases.

^{236}U. A total amount of 35 kg of mobile ^{236}U is estimated to come from natural sources. Global fallout from atmospheric nuclear weapons tests contributed to the total inventory with ~900–2100 kg [24, 76, 77]. Finally, the nuclear fuel reprocessing plants at Sellafield and La Hague have carried out significant

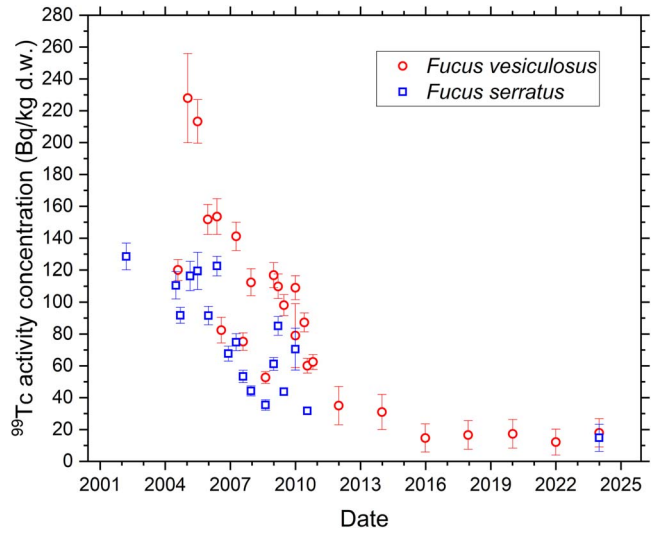

Figure 12. The ^{99}Tc activity concentration in *Fucus serratus* and *F. vesiculosus* from Särdal during the period 2001–2024.

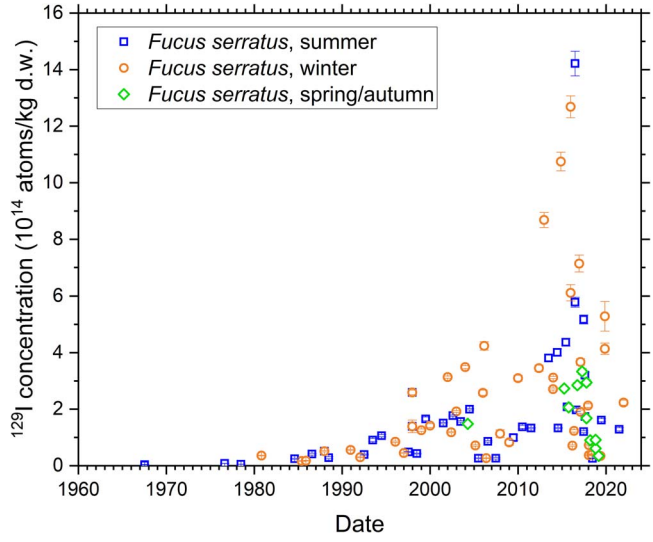

Figure 13. ^{129}I concentration (atoms/kg d.w.) in *Fucus serratus* from Särdal (1 atom = $1.40 \cdot 10^{-15}$ Bq).

liquid discharges to the sea. Uranium-236 contributions from these sources have been estimated to be from ~95 to over 260 kg [76, 78]. More recently, measurement of another anthropogenic uranium isotope, ^{233}U ($T_{1/2}$ = 0.16 $\cdot 10^6$ years), in environmental samples by AMS provided a new possibility to discriminate ^{236}U sources between global fallout and nuclear reactors [79, 80]. The estimated overall global fallout ^{233}U inventory is 14 kg [81]. Thus, being extremely minor, its analysis is very challenging. No ^{233}U measurements have been made so far on the current *Fucus* spp. samples.

The ^{236}U concentration in *Fucus* spp. is given in Table 2 and in Fig. 17. The time course of the discharges from Sellafield, reconstructed from ^{236}U data from shells of *Cerastoderma edule* [23], match with the concentrations trend seen in the algae from Särdal, and thus confirms the finding of the significant underestimation of the reported releases from Sellafield during the 1970-ies.

Table 2 shows the measured ^{236}U concentrations as well as the ^{236}U/^{238}U atomic ratio (also in Fig. 18) in *F. serratus* from Särdal during the period 1976–2021. The table also contains the ^{129}I concentrations in some

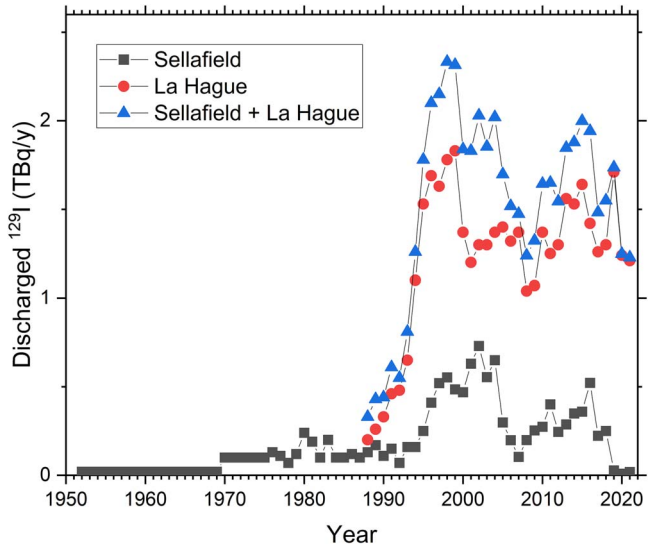

Figure 14. Reported releases of ^{129}I from the reprocessing facilities at Sellafield and La Hague [21, 22, 42, 46, 47].

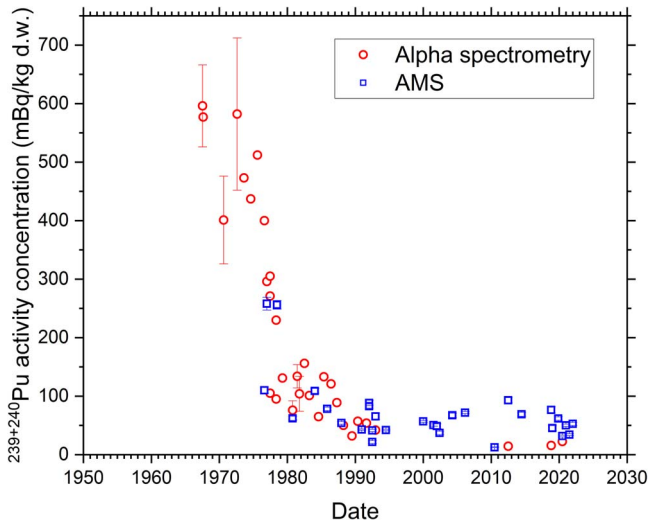

Figure 15. The activity concentration (mBq/kg d.w.) of $^{239+240}$Pu in *Fucus* spp. collected at Särdal in the period 1967 to 2021.

of the samples as well as the ^{129}I/^{236}U -atomic ratios for these samples.

Uranium-236 with its conservative behaviour would provide an additional transient signal of water masses. In combination with other anthropogenic radionuclide tracers (e.g. ^{3}H, ^{14}C, ^{99}Tc, ^{134}Cs, ^{137}Cs, and ^{129}I) tracer applications of ^{236}U in the world oceans have provided a clearer picture of water mass movements, circulation and mixing patterns of different water flows [78]. Further measurement and analysis of the *Fucus* samples from various parts of the inter Nordic waters might give similar information.

In the period 1976–2021, the ^{236}U/^{238}U atomic ratios in *Fucus* spp. from Särdal were 12–80 times higher than the estimated global fallout value of $1{\cdot}10^{-9}$ indicating significant contributions from other sources, most likely the reprocessing plants in Sellafield and La Hague. The decreasing trend of the ^{236}U/^{238}U reflect the decreasing releases of ^{236}U in both Sellafield and La Hague [82, 83]. The results agree well with previously published results for surface water from the Kattegat [84]. Measurements in 2013 gave ^{236}U/^{238}U atomic ratios of $(20.9 \pm 7.0) \cdot 10^{-9}$ and in 2014 $(17.2 \pm 6.6) \cdot 10^{-9}$ in surface water from Kattegat

Table 1. Activity concentration of ^{239}Pu and ^{240}Pu in *Fucus serratus* from Särdal (56.76 N, 12.63E).

Sampling date	^{239}Pu	^{240}Pu	$^{239+240}$Pu	^{240}Pu/^{239}Pu	^{240}Pu/^{239}Pu
YYYY-MM-DD	mBq/kg d.w.	mBq/kg d.w.	mBq/kg d.w.	Activity ratio	Atom ratio
1976-08-21	61.9 ± 2.5	48.3 ± 2.2	110 ± 4.7	0.780 ± 0.067	0.212 ± 0.005
1976-12-31	158.8 ± 6.4	98.7 ± 4.1	258 ± 11	0.622 ± 0.051	0.169 ± 0.014
1978-06-26	152.4 ± 4.4	103.7 ± 3.0	256 ± 7.4	0.680 ± 0.039	0.185 ± 0.011
1980-10-26	38.1 ± 1.4	24.1 ± 2.3	62.2 ± 3.7	0.633 ± 0.084	0.172 ± 0.023
1984-01-22	65.8 ± 2.6	42.9 ± 1.6	109 ± 4.2	0.652 ± 0.050	0.178 ± 0.014
1985–11–17	45.8 ± 1.5	32.5 ± 1.5	78.3 ± 3.0	0.710 ± 0.055	0.193 ± 0.015
1987-12-24	33.5 ± 1.1	20.7 ± 1.3	54.2 ± 2.4	0.618 ± 0.041	0.168 ± 0.011
1990-12-15	25,3 ± 0.5	17.6 ± 0.6	42.9 ± 1.1	0.696 ± 0.037	0.189 ± 0.010
1992-01-19	53.3 ± 2.5	35.1 ± 2.0	88.4 ± 4.5	0.659 ± 0.069	0.179 ± 0.019
-"-	47.8 ± 1.8	35.0 ± 1.7	82.8 ± 3.5	0.732 ± 0.063	0.199 ± 0.017
1992-07-04	25.2 ± 1.1	16.1 ± 0.9	41.3 ± 2.0	0.639 ± 0.064	0.174 ± 0.017
-"-	13.5 ± 1.0	8.1 ± 1.4	21.6 ± 2.4	0.60 ± 0.088	0.163 ± 0.024
1994-07-09	38.8 ± 2.2	26.6 ± 3.2	65.4 ± 5.4	0.686 ± 0.121	0.187 ± 0.033
1997-12-21	24.5 ± 0.5	17.5 ± 0.8	42.0 ± 1.3	0.714 ± 0.047	0.194 ± 0.027
1999-12-31	33.4 ± 0.8	23.5 ± 0.9	56.9 ± 1.7	0.623 ± 0.039	0.170 ± 0.011
2001-07-03	30.4 ± 1.2	20.2 ± 0.8	50.6 ± 2.0	0.664 ± 0.053	0.180 ± 0.014
2001-12-31	28.5 ± 2.0	20.3 ± 2.0	48.8 ± 4.0	0.712 ± 0.120	0.194 ± 0.033
2002-05-26	23.3 ± 1.4	13.8 ± 1.6	37.1 ± 3.0	0.592 ± 0.104	0.161 ± 0.028
2004-04-22	41.4 ± 1.2	25.8 ± 2.5	67.2 ± 3.7	0.623 ± 0.078	0.170 ± 0.021
2006-02-25	41.3 ± 0.9	30.5 ± 0.9	71.8 ± 1.8	0.738 ± 0.038	0.195 ± 0.010
2010-07-03	7.6 ± 0.3	5.0 ± 0.4	12.6 ± 0.7	0.658 ± 0.076	0.179 ± 0.021
2014-06-20	54.1 ± 1.6	38.9 ± 2.8	93.0 ± 4.4	0.719 ± 0.073	0.196 ± 0.020
2018-06-23	39.3 ± 1.7	29.6 ± 1.7	69.0 ± 3.4	0.753 ± 0.077	0.204 ± 0.027
2018-12-31	45.8 ± 2.8	30.6 ± 2.4	76.5 ± 5.2	0.668 ± 0.093	0.182 ± 0.025
2019-08-29	28.5 ± 2.3	16.8 ± 2.0	45.4 ± 4.3	0.589 ± 0.117	0.160 ± 0.032
2019-11-15	36.3 ± 1.8	25.4 ± 1.1	61.7 ± 2.9	0.700 ± 0.065	0.190 ± 0.018
2020-06-23	19.1 ± 0.8	12.7 ± 0.8	31.8 ± 1.6	0.665 ± 0.070	0.180 ± 0.019
2021-01-01	29.7 ± 1.2	20.5 ± 0.9	50.2 ± 2.1	0.684 ± 0.058	0.188 ± 0.016
2021-07-04	19.3 ± 0.8	14.8 ± 0.8	34.1 ± 1.6	0.767 ± 0.073	0.209 ± 0.020
2021-12-31	30.0 ± 1.6	21.6 ± 1.1	51.6 ± 2.7	0.720 ± 0.075	0.196 ± 0.020

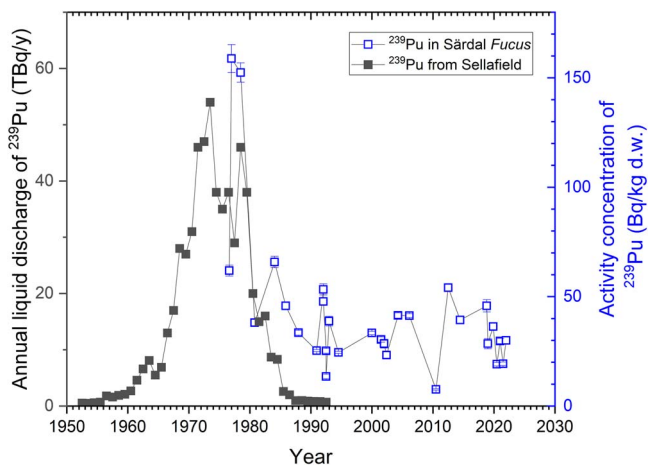

Figure 16. Estimated yearly ^{239}Pu releases (TBq) [23] from Sellafield and measured ^{239}Pu activity concentration in *Fucus serratus* from Särdal. To compare the time course of releases and activity concentration in the algae, the transport time from Sellafield to Särdal must be taken into account, which for ^{137}Cs and ^{99}Tc was previously determined to be 4 years [2, 12, 14, 15].

[84]. Measurements of further *Fucus* samples from Särdal, especially in the time interval 1967–2000 will be done.

The ^{236}U-results from *Fucus* spp. from Särdal agree also well with recently publisher data by Qiao et al. [85] from the southern part of Kattegat (Klint, 53.14 N,

Table 2. ^{236}U-concentration as well as ^{236}U/^{238}U- and ^{129}I/^{236}U-atom ratio in *Fucus serratus* from Särdal (56.76 N, 12.63E).

Sampling date YYYY-MM-DD	^{236}U (atoms/kg d.w.) 10^9	^{236}U/ ^{238}U (atom ratio) 10^{-9}	^{129}I/^{236}U (atom ratio)
1976-12-31	44.2 ± 2.3	38.8 ± 1.7	0.14 ± 0.06
1978-06-01	78.0 ± 7.6		
1984-01-22	36.1 ± 2.1	73.8 ± 3.6	0.83 ± 0.20
1985-11-01	197.0 ± 17.0		
1990-11-01	49.3 ± 4.1		
1992-01-19	41.0 ± 1.3	80.2 ± 7.2	0.75 ± 0.14
1992-01-19	39.8 ± 5.6		
1992-07-04	53.6 ± 1.8	30.6 ± 0.9	0.73 ± 0.06
1992-07-04	54.1 ± 2.2		
1994-07-09	53.9 ± 2.2	28.0 ± 1.7	1.96 ± 0.16
2001-07-03	41.6 ± 1.9	30.2 ± 1.2	3.63 ± 0.22
2001-12-31	29.8 ± 1.2	23.2 ± 0.9	10.5 ± 0.6
2002-05-26	27.1 ± 1.9	23.7 ± 1.4	4.35 ± 0.36
2004-04-12	18.8 ± 4.1		
2010-07-03	5.8 ± 0.3		
2018-06-23	22.4 ± 1.4	11.4 ± 1.4	3.25 ± 0.28
2018-12-31	13.1 ± 0.45	9.6 ± 0.5	5.82 ± 0.41
2019-06-23	16.3 ± 0.56	10.6 ± 0.7	9.89 ± 0.70
2019-11-15	12.4 ± 3.3		
2020-06-23	15.0 ± 0.8	12.7 ± 0.7	10.7±
2021-01-01	13.7 ± 0.6	12.6 ± 0.5	16.3 ± 1.3
2021-07-04	10.0 ± 0.8		
2021-12-21	9.0 ± 1.1		

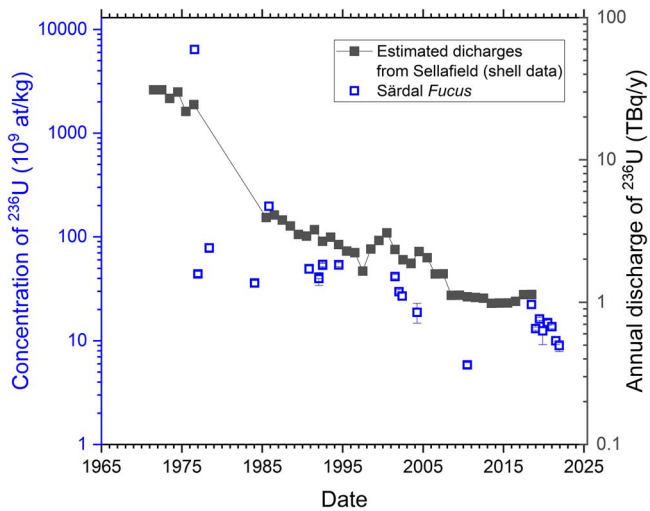

Figure 17. ^{236}U concentration (at/kg) in *Fucus* spp. from Särdal and the estimated releases of ^{236}U from Sellafield nuclear reprocessing plant according to shell data by Castrillejo *et al*. [64]. Note the logarithmic scale on the y-axis. During the whole time period releases from Sellafield seems to dominate over the releases from La Hague [22]. To compare the time course of releases and activity concentration in the algae, the transport time from Sellafield to Särdal must be taken into account, which for ^{137}Cs and ^{99}Tc was previously determined to be 4 years [2, 12, 14, 15].

4.34E) both for the concentrations and the ^{236}U/^{238}U atom ratios in *F. serratus* collected outside Gothenburg in 2020 and 202 [38].

The exponential increase of the ^{129}I concentration by time and the likewise exponential decrease of the ^{236}U concentration result in an exponential increase of the ^{129}I/^{236}U atomic ratio as shown in Fig. 19.

The releases from La Hague are characterized by increasing ^{129}I/^{236}U-ratios, while the releases from Sellafield show equal ^{129}I/^{236}U-ratios year by year [79].

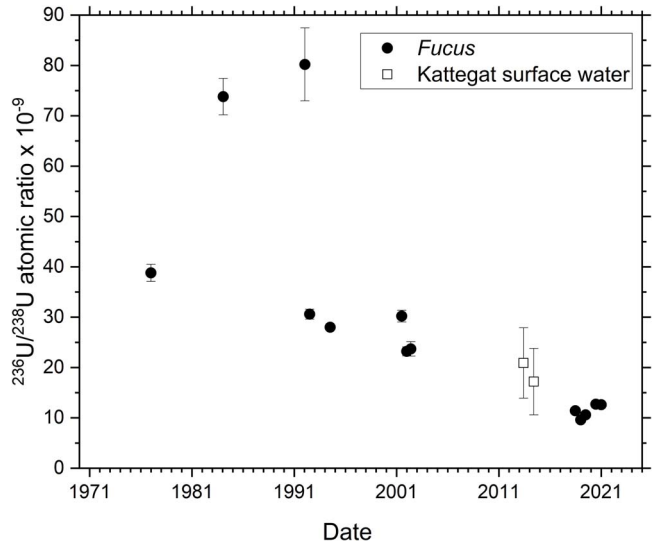

Figure 18. The $^{236}U/^{238}U$ atomic ratio in *Fucus serratus* from Särdal at various times between 1976 and 2021 and the same ratio in surface water from the Danish straits in 2013 and 2014 [84].

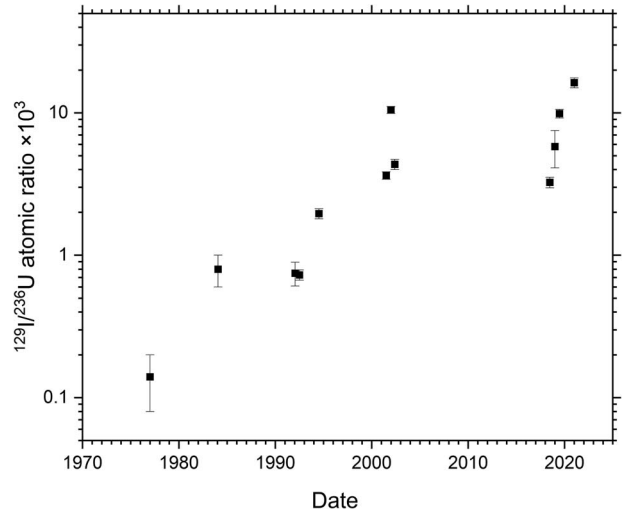

Figure 19. $^{129}I/^{236}U$ atomic ratio in *Fucus serratus* from Särdal at various times between 1976 and 2021.

The increasing $^{129}I/^{236}U$ ratio in *Fucus* spp. from Särdal is an indication that an increasing proportion of the ^{129}I that reaches the Swedish west coast comes from La Hague, at least until ~2005.

Summary and conclusions

The high uptake of different radionuclides from seawater in the brown algae *F. serratus* and *F. vesiculosus* has been used to map the temporal and geographical variations of the presence of various anthropomorphic radionuclides in the marine environment at locations on the Swedish coast. By studying the radionuclide composition in different places and over time, it is possible to gain an increased understanding of the amount, transport routes and mixing conditions for radionuclide emissions. This is important for today's and tomorrow's risk assessments, and not least in connection with radiological and nuclear emergencies. Then background levels of relevant radionuclides are needed. The results have also implications on the understanding of transport, mixing and dilution of nutrients and toxic substances in the marine environment. The studies at Särdal show increasing levels of ^{129}I and a decrease in the concentration of ^{137}Cs, ^{99}Tc, ^{14}C, ^{236}U, and $^{239+240}Pu$ over time. The

influx of water from the North Sea, with radionuclides released from French and British spent nuclear fuel reprocessing facilities into the waters of the Swedish west coast, is clear and particularly visible in the ^{137}Cs, ^{14}C, ^{129}I, and ^{236}U data. The studies of ^{14}C in Swedish water in 2020 also show a contribution from the Ringhals nuclear power plant, which is significant in the nuclear power plant's immediate environment. The ^{3}H activity concentrations for *Fucus* spp. and seawater were at the expected natural environmental level. Chernobyl-derived ^{137}Cs, stored in the Baltic Sea since 1986, still act as a dominant anthropogenic radionuclide in the coastal waters of the Swedish west coast.

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Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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