

The 2002/2003 radionuclide concentration in the marine environment at various distances from the Barsebäck nuclear power plant

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ABSTRACT The activity concentration of ¹³⁷Cs, ¹³¹I, ⁶⁵Zn, ⁶⁰Co, ⁵⁸Co, ⁵⁴Mn, and ⁴⁰K were determined in samples of brown seaweed (*Fucus*) and some other marine plants using low background high-resolution gamma-spectrometry. The algae were mainly sampled in the bay just north of the Barsebäck NPP (55.4 N, 12.6 E) in the south of Sweden to study the contamination levels in the nearest shallow waters. One aim of the study was to investigate whether the levels were high enough to expect environmental effects. Some samples were also taken at longer distances up to 130 km from the Barsebäck NPP. Measurable levels of the neutron activation products ⁶⁵Zn (up to 17 Bq/kg dw), ⁶⁰Co (100–600 Bq/kg dw), ⁵⁸Co (1–160 Bq/kg dw) and ⁵⁴Mn (12–90 Bq/kg dw) were found in the algae samples within a distance of 5 km from the plant. The decrease in activity concentration with distance from the plant could be described by a power function with an exponent ranging from 1.4 to 2.4. This was in fair agreement with the value for a true two-dimensional dispersion model. The present-day concentrations were found to be considerably lower than in earlier studies made in the late 1970s, especially for ⁶⁵Zn and ⁵⁸Co. The activity concentration of gamma emitting radionuclides in *Fucus vesiculosus* from the bay just north of Barsebäck in the period 2002-2003 was dominated by (in order of decreasing concentration): natural ⁴⁰K, ⁶⁰Co from the plant, ¹³⁷Cs mainly from the Chernobyl debris, ⁵⁴Mn and ⁵⁸Co from the plant. It is not likely that any effects from the very marginal absorbed dose contribution from the Barsebäck NPP releases can be found even in the nearest environment. The study has also shown that the eelgrass *Zostera marina* may be a bioindicator to use in further studies of the radiation environment in shallow water, especially for ⁶⁰Co and ⁵⁴Mn.

Keywords: Barsebäck NPP / *fucus* / activation products / marine environment

RÉSUMÉ La concentration de radionucléides – en 2002/2003 – dans l'environnement marin, à diverses distances de la centrale électronucléaire de Barsebäck.

On a utilisé la spectrométrie gamma de haute résolution et à bas bruit de fond, pour déterminer les concentrations de l'activité de ¹³⁷Cs, ¹³¹I, ⁶⁵Zn, ⁶⁰Co, ⁵⁸Co, ⁵⁴Mn et ⁴⁰K dans des échantillons de goémon brun (*Fucus*) et de plusieurs autres plantes marines. On prélevait les algues surtout dans la baie située juste au nord de la centrale électronucléaire de Barsebäck (55,4 N, 12,6 E), au sud de la Suède, pour étudier les niveaux de contamination des hauts fonds les plus proches. On a aussi

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collecté quelques échantillons à des distances plus grandes, allant jusqu'à 130 km de la centrale électronucléaire de Barsebäck. Pour des distances à la centrale inférieures à 5 km, on a trouvé, dans les échantillons d'algues, des niveaux mesurables de produits d'activation neutronique, ^{65}Zn (jusqu'à 17 Bq/kg dw), ^{60}Co (100–600 Bq/kg dw), ^{58}Co (1–160 Bq/kg dw) et ^{54}Mn (12–90 Bq/kg dw). La décroissance de la concentration de l'activité en fonction de la distance x à la centrale, peut se représenter par une fonction $\alpha x^{-\beta}$, β étant compris entre 1,4 et 2,4. Ce résultat est en honnête accord avec la valeur déduite d'un modèle valable de dispersion à deux dimensions. Les concentrations actuelles sont considérablement plus faibles que celles obtenues dans des études antérieures, effectuées à la fin des années 70, particulièrement en ce qui concerne ^{65}Zn et ^{58}Co . Dans la période 2002–2003, la concentration de l'activité de radionucléides émetteurs gamma dans le goémon (*Fucus vesiculosus*) venant de la baie au nord de Barsebäck était dominée (dans l'ordre des concentrations décroissantes) par ^{40}K naturel, ^{60}Co venant de la centrale, ^{137}Cs venant principalement des retombées de Chernobyl, ^{54}Mn et ^{58}Co venant de la centrale. Il n'est pas probable que l'on puisse trouver, même dans l'environnement le plus proche, quelque effet que ce soit venant de la contribution très marginale à la dose absorbée des rejets de la centrale électronucléaire de Barsebäck. Cette étude a également montré que la zostère *Zostera marina* peut s'avérer un bioindicateur à employer dans les études ultérieures de l'environnement radioactif sur les hauts fonds, en particulier en ce qui concerne ^{60}Co et ^{54}Mn .

1. Introduction

During normal operation of a nuclear power plant (NPP), small controlled releases mainly of neutron activation products are common and occur through the cooling water outlet. Until November 1999, the Barsebäck nuclear power plant (Barsebäck NPP) consisted of two reactors in operation, Barsebäck-1 (between 1976 and 1999) and Barsebäck-2 (since 1977), both of boiling water type, having an electrical power of 600 MW, or 1700 MW thermal power each. Barsebäck-2 has been shut down since the year 2005.

The radiation environment in the vicinity of the plant is relatively well known due to a long-term monitoring programme of the radionuclide concentration in a number of species, both terrestrial and aquatic (Wijk and Luning, 2001). There have also been several research programmes since the start of the operation of the reactors (Mattsson *et al.*, 1980a). Among the most frequently found radionuclides in the vicinity of the NPP are ^{65}Zn , ^{60}Co , ^{58}Co , and ^{54}Mn . There is normally also ^{137}Cs , which originates from various sources. Environmental contamination by ^{137}Cs is of particular public health interest because of the various sources of fallout originating from nuclear weapons, radiological source disruptions, and the Chernobyl disaster. This dispersion may lead to a chronic ecosystem contamination and subsequent ingestion of contaminated foodstuffs.

This study investigates how the various radionuclides are distributed in the vicinity of the plant. Even though the discharged radionuclides from the Barsebäck NPP are dispersed into large volumes of water, one of the bio-indicator used here,

the brown seaweed (*Fucus*), has the ability to accumulate several of the radionuclides of interest into its biomass in measurable quantities (Mattsson *et al.*, 1980a; Nilsson *et al.*, 1980). The results are compared with those of a similar research performed in 1977 and from other studies in the area (Mattsson *et al.*, 1980b; Holm, 1994).

Another aim of the study is to investigate whether the activity concentration in some of the organisms that are commonly found in the Öresund is high enough to give any radiation effects.

2. Material and methods

Samples of *Fucus vesiculosus* were collected along the Swedish shore of the Öresund and Kattegat at different distances from the Barsebäck NPP (Fig. 1). The samples were collected between March 2002 and March 2003. The depth of water at the sampling sites varied between 0.5 and 1 m. All the seaweed samples (whole

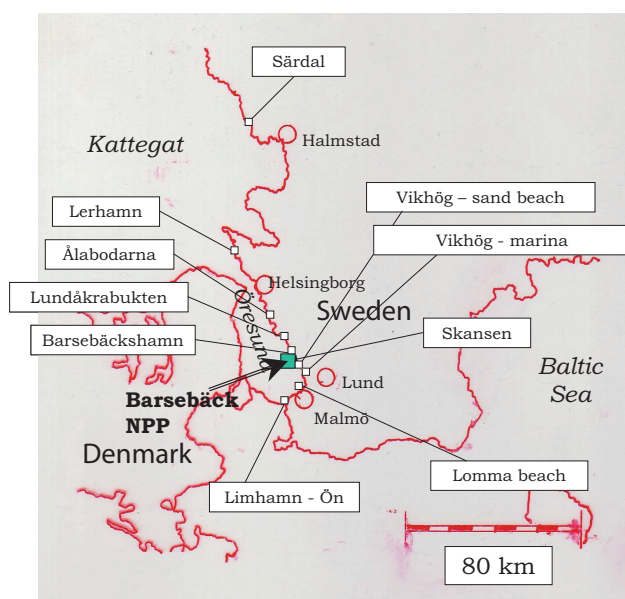


Figure 1 – Sampling points of *Fucus vesiculosus* (and in some cases additional sampling of *Fucus serratus*, *Enteromorpha intestinalis*, *Zostera marina*, *Mytilus edulis*) in the Swedish coast of the Öresund and Kattegat.

Points de prélèvement de Fucus vesiculosus (avec, dans certains cas, des échantillons additionnels de Fucus serratus, Enteromorpha intestinalis, Zostera marina, Mytilus edulis) sur la côte suédoise de l'Öresund et du Kattegat.

Fucus plant) collected were firmly rooted to the bottom of the waters, most often on stones. At collection, the samples were put into plastic bags. All the samples were dried at room temperature (~20 °C) for a period of five days. Afterwards, the samples were grounded using a laboratory mill. Furthermore, the grounded samples were put in 150 ml pre-weighed plastic beakers and the net weight was recorded.

From one sampling spot (Lundåkrabukten), around 6 km north of the Barsebäck NPP, samples of *Fucus vesiculosus*, eelgrass (*Zostera marina*), light green algae (*Enteromorpha intestinalis*), mussels (*Mytilus edulis*) clean from any micro algae, as well as mussels with micro algae on their surface were sampled on August 2nd, 2002. The eelgrass, light green algae, as well as mussel samples were dried at 110 °C for 24 hours while the *Fucus vesiculosus* was dried at room temperature for 5 days.

After the sample preparation was completed, the activities of ^{137}Cs , ^{131}I , ^{65}Zn , ^{60}Co , ^{58}Co , ^{54}Mn and ^{40}K were determined by gamma-ray spectrometry with acquisition times between 100,000 and 150,000 seconds (22 and 33 hours). The measurements were performed using an HPGe (EG&G Ortec) detector of 36% efficiency, and energy resolution of 1.8 keV (FWHM) at 1.33 MeV.

3. Results and discussion

3.1. Variation in radionuclide activity concentration with distance from the Barsebäck NPP

The activity concentrations of ^{137}Cs , ^{60}Co , ^{58}Co , ^{54}Mn and ^{40}K in the *Fucus* samples in relation to the distance from the outlet of Barsebäck are given in Figure 2 and Table I. Some results concerning ^{65}Zn , where the concentration was lower, are given in Table II. Moreover, ^{131}I was found in March 2002.

The results (Fig. 2) show that the activity concentrations of ^{60}Co , ^{58}Co , and ^{54}Mn decrease with distance from the point of release and that the distance dependence can be described by a power function, $C(x) = \alpha \cdot x^{-\beta}$ as in 1977 (Mattsson *et al.*, 1980a, 1980b). From the curve fits of Figure 2 it can be deduced that the concentration in the *Fucus* sampled to the south of Barsebäck is about 1/4 of the concentration at the corresponding distance north of the plant. This distribution agrees roughly with the distribution of the surface water current direction in the Öresund; 30% southward, 60% northward and 10% quiescent (SMHI, 1981). The content of ^{137}Cs found in the *Fucus* samples are most likely originating from Chernobyl fallout over the Baltic Sea and its run off areas.

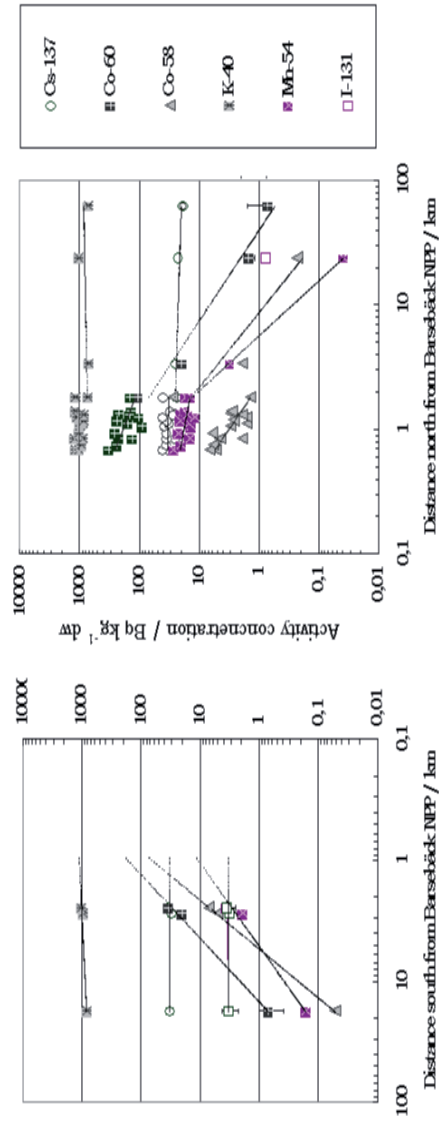


Figure 2 – Activity concentration in *Fucus vesiculosus* in relation to the distance south and north from the cooling water outlet of Barsebäck NPP in August 2002. Power functions were fitted to the observed data. Bars indicate the uncertainty due to pulse statistics (1 SD).

Concentration de l'activité dans *Fucus vesiculosus* en fonction de la distance, au sud ou au nord, au point de sortie de l'eau de refroidissement de la centrale de Barsebäck, en août 2002. Les fonctions $\alpha \cdot x^\beta$ sont ajustées sur les données observées. Les barres indiquent l'incertitude résultant des statistiques des impulsions.

TABLE I

Comparison of the power exponent and their Pearson's determination coefficient (Pearson's R^2) at short (<2 km) and intermediate (>2–50 km) distances north and south of the Barsebäck NPP.

Comparaison de l'exposant de puissance (β) et de leur coefficient de Pearson déterminé (R^2 de Pearson) à faibles (< 2 km) et moyennes (> 2–50 km) distances au nord et au sud de Barsebäck.

Radionuclide	Power exponent					
	Short range North	R^2 North	Intermediate range North	R^2 North	Intermediate range South	R^2 South
^{137}Cs	-0.05	0.01	-0.06	0.74	-0.01	0.01
^{60}Co	-0.79	0.33	-1.37	0.97	-1.9	0.99
^{58}Co	-1.41	0.57	-1.66	0.87	-2.5	0.99
^{54}Mn	-0.38	0.20	-2.26	0.99	-1.46	0.98
^{40}K	-0.05	0.01	-0.03	0.12	0.1	0.90

Table II

Variation of activity concentration levels of radionuclides at Skansen sampling station (1100 m north of Barsebäck NPP cooling water discharge pipe) in June 1977, March, May, August, September, October 2002 as well as January 2003.

Variation des niveaux de concentration de l'activité de radionucléides à la station de prélèvements de Skansen (1100 m au nord de la canalisation de rejet de l'eau de refroidissement de la centrale de Barsebäck, en juin 1977, mars, mai, août, septembre, octobre 2002 ainsi qu'en janvier 2003.

Radionuclide	Physical half-life (y)	June 12, 1977 (Bq/kg dw)	March 25, 2002 (Bq/kg dw)	May 13, 2002 (Bq/kg dw)	August 2, 2002 (Bq/kg dw)	September 3, 2002 (Bq/kg dw)	October 30, 2002 (Bq/kg dw)	January 20, 2003 (Bq/kg dw)
^{65}Zn	0.668	150	2	1.8	0	10	17	5
^{60}Co	5.27	1000	319	154	121	389	686	273
^{58}Co	0.194	500	5	2	28	93	130	18
^{54}Mn	0.854	100	26	15	14	60	90	14
^{40}K	1.28×10^9	N/A	1164	1161	725	746	1182	911

In the samples from March 2002, significant concentrations of ^{131}I were also found, mainly at the points south to the Barsebäck NPP. However, there were no reports on releases of ^{131}I through the cooling water discharge of the Barsebäck NPP to the Öresund marine environment during that period (Goldstrand, 2002). The most likely source of ^{131}I originates from excreta from patients treated with ^{131}I , which is released via local wastewater plants into the Öresund.

A relationship between the concentration of radionuclides and the distance from the discharge pipe of the Barsebäck NPP is found for neutron activation products such as ^{60}Co , ^{58}Co , ^{54}Mn . This distance dependence appears to be similar for the three radionuclides, with the slope of the log-log plots ranging from -1.37 to -2.26 (Tab. I). However, there is no similar distance dependence for the activity concentration of ^{137}Cs . This indicates that the majority of the ^{137}Cs contents found in the *Fucus* samples outside the Barsebäck NPP are residual products of the releases from other sources such as the Chernobyl accidents and from Western European reprocessing plants (Mattsson and Erlandsson, 1991).

The different slopes for the nearest bay and the more distant water at intermediate ranges are explained by the different dispersion patterns of the source term. The bay north of the Barsebäck NPP, lies outside the release water plume, and the radionuclide inventory in the bay is mainly governed by currents intersecting the plume and translocation fractions of the plume towards the shore of the bay (Larsson, 2002). At intermediate distances, the dispersion of the radionuclides more closely resembles a true two-dimensional dispersion.

3.2. Variation over the year in the radionuclide concentration levels in *Fucus* samples

For ^{60}Co and ^{58}Co , a marked difference in the concentration levels between seasons in 2002 is observed (Tab. II) with the highest concentration levels in autumn. The controlled releases of ^{60}Co , ^{58}Co , and ^{54}Mn from the Barsebäck NPP exhibit a typical maximum value during maintenance periods of the NPP, which are performed twice a year, in the late summer as well as in early autumn in September. There is also a variation in the ^{137}Cs concentration over the year (Figs. 3 and 4).

Other factors, such as the salinity of the waters, the physicochemical concentration of the element and the seasonal biological changes in the organisms, also play an important role in the uptake of certain radionuclides by *Fucus* (Carlson and Erlandsson, 1991). The salinity in the sampling areas fluctuates, both in time and space, from about the average of 13‰ at Skansen up to 25‰ at Särödal in the near shore water. Differences in the uptake of radionuclides in *Fucus vesiculosus* from localities with different salinity are more pronounced for ^{137}Cs , with increasing activity concentrations with decreasing salinity.

3.3. Comparison with previous results

The average activity concentrations of neutron activation products in the *Fucus* samples in the Swedish shores of Öresund are compared with the results that were

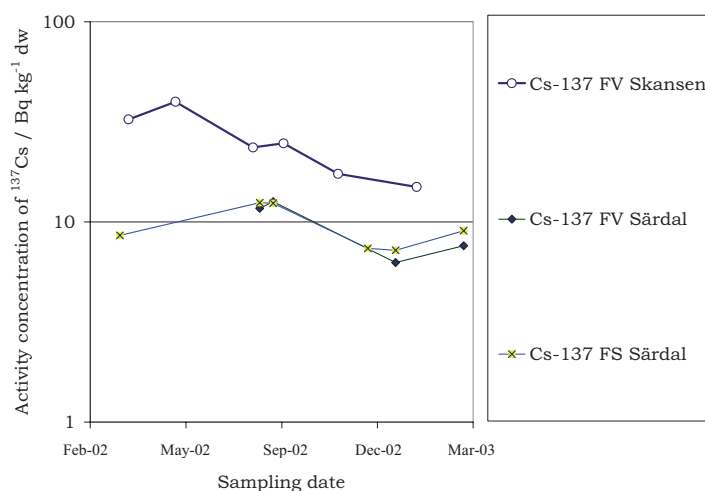


Figure 3 – Activity concentration of ¹³⁷Cs in Fucus over the year 2002 and January and March 2003 at Skansen (1.1 km north of the Barsebäck NPP) and at SärDAL (130 km north of the Barsebäck NPP). FV = Fucus vesiculosus; FS = Fucus serratus.

Concentration de l'activité de ¹³⁷Cs dans Fucus durant l'année 2002, janvier et mars 2003, à Skansen (1,1 km au nord de la centrale de Barkebäck), et à SärDAL (130 km au nord de Barkebäck). FV = Fucus vesiculosus ; FS = Fucus serratus.

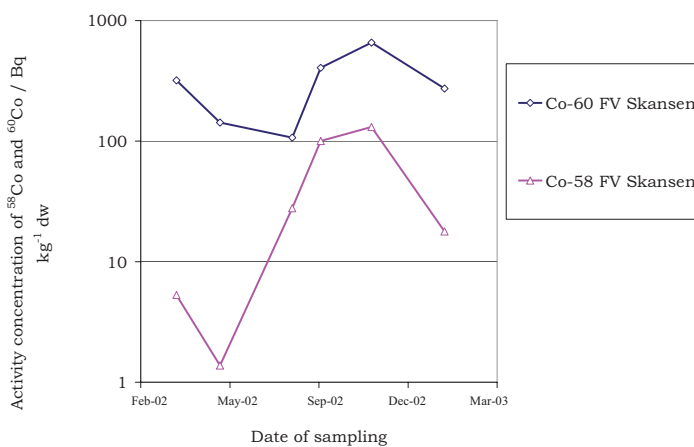


Figure 4 – Activity concentration of ⁶⁰Co and ⁵⁸Co in Fucus vesiculosus over the year 2002 and January 2003 at Skansen.

Concentration de l'activité de ⁶⁰Co et ⁵⁸Co dans Fucus vesiculosus durant l'année 2002 et janvier 2003 à Skansen.

Table III
Activity concentration of radionuclides in various species sampled at Lundåkrabukten (around 6 km north of the Barsebäck NPP) in August 2nd, 2002.
Concentration de l'activité de radionucléides dans diverses espèces prélevées à Lundåkrabukten (à 6 km environ au nord de la centrale de Barsebäck), le 2 août 2002.

Radionuclide	<i>Fucus vesiculosus</i> (Bq/kg dw)	<i>Enteromorpha intestinalis</i> (Bq/kg dw)	<i>Zostera marina</i> (Bq/kg dw)	<i>Mytilus edulis</i> with microalgae on their surface (Bq/kg dw)	<i>Mytilus edulis</i> clean from microalgae on their surface (Bq/kg dw)
¹³⁷ Cs	24	15	24	4	4
⁶⁰ Co	3	2	11	0.3	<MDA
⁵⁴ Mn	<MDA	1	2	2	1
⁴⁰ K	745	804	1137	92	70

obtained in 1977 (Tab. II). It is evident that all activity concentration levels in 2002 were significantly lower than those in 1977, especially for ⁶⁵Zn. Though it can not be the only explanation for such a decrease in all activity concentration, one could expect some decrease as in 2002/2003 only one of the two units had been operating.

3.4. Concentrations of radionuclides in various marine species

Of all the species sampled at Lundåkrabukten, eelgrass, *Zostera marina*, exhibited the highest concentration of ⁶⁰Co and ⁵⁴Mn. Eelgrass has also the possibility to accumulate ¹³⁷Cs at similar concentration as *Fucus vesiculosus* (Tab. III). Within the marine ecology eelgrass is considered as a useful bioindicator of the general ecological status of the shallow waters and estuaries (*e.g.*; PTMSC, 2003). The results found here indicate that eelgrass may also have potential as a sensitive bioindicator of radiation level in marine environments.

4. Conclusions

The activity concentration of gamma emitting radionuclides in *Fucus* from the bay just north of Barsebäck was in 2002/2003 dominated by (in order of decreasing concentration): natural ⁴⁰K, ⁶⁰Co from the plant, ¹³⁷Cs mainly from the Chernobyl releases in 1986 and from western reprocessing plants, ⁵⁴Mn and ⁵⁸Co from the plant. The concentration of Barsebäck-produced radionuclides is lower per generated MW_e than in 1977.

At intermediate ranges, the activity concentration of various gamma-emitting radionuclides in *Fucus* decreases as the distance increases from the Barsebäck NPP in a fashion that is predicted by the theory of two-dimensional dispersion.

The concentration levels of various radionuclides are higher in *Fucus* during the late spring and summer times, while they reach their minimum by the end of winter season. *Zostera marina* appears to be a good bio-indicator for indicating the releases of neutron activation products of radionuclides from nuclear power plants to the marine waters, especially for ^{60}Co and ^{54}Mn .

It is not likely that any effects from the very marginal absorbed dose contribution from the Barsebäck NPP releases can be found even in the nearest environment.

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