

# Variations in the $^{137}\text{Cs}$ concentration in surface coastal water at the Swedish west coast during 40 years as indicated by *Fucus*

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## INTRODUCTION

Since the middle of last century, the marine environment has been polluted with anthropogenic radionuclides of various origins. At the Swedish west coast the dominating sources have originated from global fallout from atmospheric nuclear weapons tests, fallout from the Chernobyl accident and liquid discharges from European nuclear reprocessing plants for spent nuclear fuel. There are also minor contributions from operative discharges from nuclear power reactors and from hospitals and patients having undergone diagnosis or treatments using radionuclides. A radionuclide of specific interest is the fission product  $^{137}\text{Cs}$ . Even though the concentration factor of the radionuclide from seawater to *Fucus* is lower than for most other radionuclides of interest (IAEA, 1985), it is possible to use *Fucus* as an indicator for the presence of  $^{137}\text{Cs}$  in coastal waters. Depending on sampling location, the variation in the  $^{137}\text{Cs}$  concentration in *Fucus* over time can be used to indicate local and global transportation of radionuclides in the marine environment.

To follow the long-term trends of radionuclide concentration in the marine environment off the Swedish west coast, sampling and gamma spectrometry of the brown algae *Fucus* have been carried out since 1967 (Mattsson, 1984; Mattsson and Erlandsson, 1991) and are still in progress. The aim of this paper is to summarise the long-term variations of the  $^{137}\text{Cs}$ -concentration in *Fucus* and to discuss the possible contributing sources during a 40 years period.

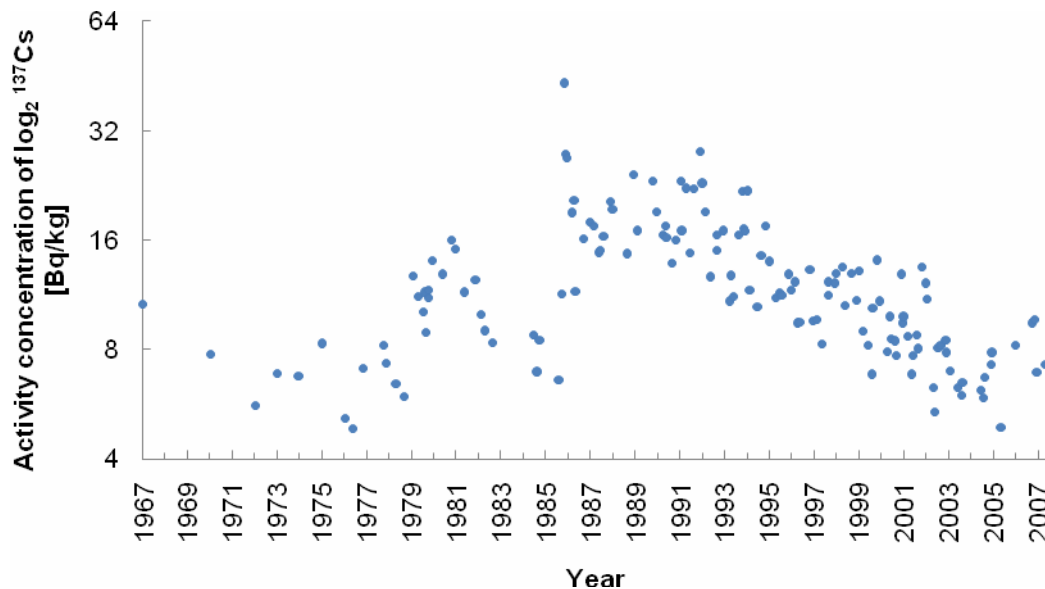
## MATERIAL AND METHODS

From 1967 and to date, samples of the brown algae *Fucus serratus* and *Fucus vesiculosus* have been collected at a specific location at Särö on the Swedish west coast. The samples have been collected within a 10 m radius at the same spot by the same person (SM), at a water depth of 0.5-1 m. Due to seasonal variations in  $^{137}\text{Cs}$  concentration in *Fucus* (compare also Carlsson and Erlandsson, 1991), up to 7 samplings were carried out each year, with an increased sampling frequency during the years after the Chernobyl accident. Up to 1983 the *Fucus* samples consisted mainly of *Fucus serratus*. Thereafter, *F. serratus* and *F. vesiculosus* were sampled and analysed separately. Several plants (normally 10-30), all firmly rooted to rocks under the water surface were taken during each sampling. Within a few days after the collection the samples were dried in air and finally at 75°C for 1 – 3 days. Thereafter the samples were ground and after careful mixing put into standard 180 ml plastic beakers. High resolution gamma spectrometry was performed using Ge(Li) and later HPGe gamma spectrometers. Reproducibility and constancy in detector response over the years were ensured by own samples of known  $^{137}\text{Cs}$  activity and by participating in a number of national

and international intercalibrations of radionuclide contents in environmental samples (e.g. Nielsen *et al.* 1996; Fogh *et al.* 2000).

## RESULTS AND CONCLUSIONS

The variation in the activity concentration of  $^{137}\text{Cs}$  in *Fucus* between 1967 and 2008 is shown in Figure 1. There are essentially two significant increases during the period, one in the late 1970s and one in 1986. Before and after these maxima there are other factors that influence the variation in  $^{137}\text{Cs}$  concentration.

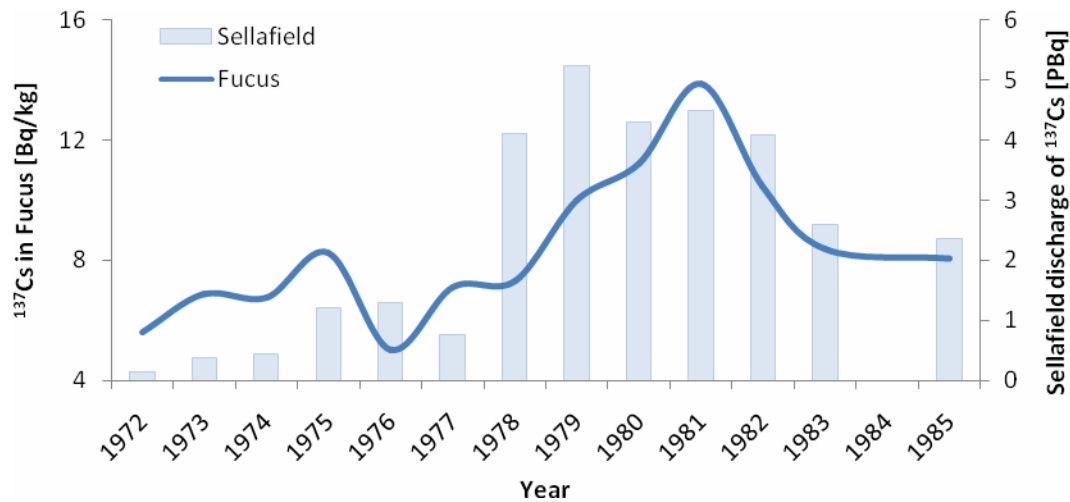


**Figure 1.** Activity concentration of  $^{137}\text{Cs}$  in *F. serratus* after  $\log_2$ -transformation, between 1967–2008.

At the beginning of the time series, the detected  $^{137}\text{Cs}$  in the *Fucus* originates from atmospheric nuclear weapons tests carried out by the super-powers, most frequently in the periods 1956-58 and 1961-62 and to some minor extent from the Sellafield reprocessing plant in UK (in operation since 1952). During the period up to 1970, the discharges from the Sellafield reprocessing plant were relatively constant (about 2% of the value reported in 1974) and the associated activity concentration in *Fucus* very small ( $< 0.8$  Bq/kg), compared with the contribution of the global fallout. In 1963, the Partial Test Ban Treaty (PTBT) was signed, which virtually ended the era of atmospheric and underwater nuclear detonations. Hence, most part ( $> 90\%$ ) of the detected  $^{137}\text{Cs}$  before 1972 is assumed to originate from the nuclear weapons test and after the PTBT there were just a limited number of low-yield Chinese tests that might have added new bomb  $^{137}\text{Cs}$ . The effective half-time of the decrease of  $^{137}\text{Cs}$  concentration in *Fucus* in the period 1967-1972 was determined to  $T_{1/2} = 5.7$  years and in 1972 the concentration was down to 5.6 Bq/kg.

Later there is an increase in the  $^{137}\text{Cs}$  concentration (up to 2.5 times the 1972 level) and it is possible to discern two peaks, the first in 1975 and the second occurring in 1979-82. The elevated levels of  $^{137}\text{Cs}$  in the 1970s and beginning of the 1980s in this part of Kattegat have earlier been associated with discharges from the Sellafield plant (Mattsson, 1984). A simple regression test for the two peaks in the *Fucus* data, when matched to the time patterns of reported Sellafield discharges of  $^{137}\text{Cs}$  (UNSCEAR, 2000), was used to calculate an approximate transportation time between release point and sampling site. The best matching

in terms of correlation coefficient (Pearson  $r$ ) yields a transportation time of about 5 years between the Irish Sea and Särödal, which is in fair agreement with the 4 years estimated in the 1980s (Mattsson, 1984; Mattsson and Erlandsson, 1991) and with later studies of  $^{99}\text{Tc}$  in *Fucus* from the same place (Lindahl *et al.*, 2003). In Figure 2 the activity concentration in *Fucus* between 1972-1985 is compared with the annual discharge from the Sellafield nuclear fuel reprocessing plant (UNSCEAR, 2000) 5 years prior to the *Fucus* collection.



**Figure 2.** The annual liquid discharge from Sellafield (bars) 5 years prior to the observed activity concentration in *Fucus* from Särödal on the Swedish west coast (solid line).

After 1981 there is a fast decrease in the activity concentration up to 1983, illustrating the effect of dilution when water from the North Sea and the Baltic are mixed. In 1983 the remaining  $^{137}\text{Cs}$  from the global fallout was estimated to 20% of the total  $^{137}\text{Cs}$  concentration in the *Fucus*. From 1983 and up to the beginning of 1986, the concentration level is relatively constant and in the end of March 1986, the  $^{137}\text{Cs}$  activity concentration in *F. serratus* was down to 6.6 Bq/kg.

Soon after the Chernobyl accident, radioactive fallout in the area came as dry deposition, but the main Chernobyl contribution at Särödal came with rain on 8 May (Erlandsson and Mattsson, 1988) and the  $^{137}\text{Cs}$  activity concentration increased temporarily to 4 times its pre-Chernobyl value. After rapid mixing, the concentration in *Fucus* stabilized at about 18 Bq/kg for both the species and the equilibrium sustained up to 1993. This period of stability is most likely achieved by the transportation of “new” Chernobyl  $^{137}\text{Cs}$  from contaminated areas of the Baltic through the Sound to Kattegat which balanced the inflow of “cleaner” water from the North Sea. This balance is interrupted after 1993 when the activity concentration in *Fucus* starts to decrease with an effective half-time of 14 years. This decrease is however much lower than after the Sellafield discharges, indicating the sustained inflow of Chernobyl  $^{137}\text{Cs}$  from the Baltic.

*Fucus* has shown a fast response to airborne fallout (both wet and dry deposition). Liquid discharges directly into the sea are traceable with a delay, dependent on the distance between the source and sampling locations and the water currents along the way. In time, physical decay and dilution with uncontaminated water as well as sedimentation will lower the activity concentration. During 2007 the  $^{137}\text{Cs}$  activity concentration was down to the pre-Chernobyl values.

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