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Differential concentration of Technetium-99 (^{99}Tc) in common intertidal molluscs with different food habits

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ABSTRACT

Concentration of ^{99}Tc has been measured in fucoids and molluscs, sampled in a sheltered intertidal at the southwest coast of Norway from February to November 2006. The concentrations of ^{99}Tc in molluscs differed significantly between species. The filtering bivalve *Mytilus edulis* had the lowest concentrations with averages of 2.3–5.9 Bq kg^{-1} d.w., while the herbivorous gastropods *Littorina littorina*, *Littorina obtusata* and *Patella vulgata* had higher concentrations. *P. vulgata* and *L. obtusata* had the highest concentrations, 40–47 and 26–30 Bq kg^{-1} d.w., respectively. *L. obtusata* has a specialized habit of living, and prefers to feed on fucoids. *P. vulgata* can graze extensively on the fucoid *Ascophyllum nodosum* when available. Fucoids are known to have very high uptake of ^{99}Tc, and this was also found in the present study. The high ^{99}Tc-concentrations of *L. obtusata* and *P. vulgata* are most likely due to their habit of feeding on fucoids.

1. Introduction

Technetium-99 (^{99}Tc) is a long-lived ($t_{1/2} = 2.13 \times 10^5$ y), man-made radionuclide produced in the fission of uranium-235 (^{235}U) or plutonium-239 (^{239}Pu), of which the largest source to the marine environment in Northern Europe is discharges from the nuclear fuel reprocessing plants Sellafield (UK) and La Hague (France). Following the commissioning of the Enhanced Actinide Removal Plant (EARP) in 1994, the effluents from the former contained elevated levels of ^{99}Tc in the period 1994–2004 (e.g. Kershaw et al., 2004; NRPA, 2009).

Technetium exists as the highly soluble pertechnetate anion (TcO_4^-) in oxic seawater (e.g. Beasley and Lorz, 1986). As a result of the main oceanic current patterns of the North Sea (described by e.g. Otto et al., 1990), ^{99}Tc is transported from both Sellafield and La Hague to Norwegian coastal areas. The elevated discharges from Sellafield led to increased concentrations of ^{99}Tc in seawater and biota along the Norwegian coast. This received much attention and strong criticism from Norwegian Ministers, media, NGOs and local community groups around the turn of the millennium (Osborne and Huston, 2009).

Concentration of ^{99}Tc in marine biota differs between groups of organisms. Of the primary producers the large brown algae belonging to the order Fucales show the highest concentrations, whereas red and green benthic algae show relatively low concentrations (Jeanmarie et al., 1981; Beasley and Lorz, 1986). Also phytoplankton normally shows low concentrations of ^{99}Tc (Fisher, 1982; Beasley and Lorz, 1986). A limited number of studies exist on uptake and concentration of ^{99}Tc in marine invertebrates and fishes. Most examined taxa show a low uptake of ^{99}Tc from seawater (Beasley and Lorz, 1986) but in a few taxa – common lobster (*Homarus gammarus*), Norway lobster (*Nephrops norvegicus*), abalone (*Haliotis* spp.) and some Polychaeta – a relatively high uptake has been found, with Concentration factors (CFs) of 10^2–10^3 (Beasley and Lorz, 1986; Masson et al., 1989; Swift, 2001). CF is defined as concentration per unit mass of organism (Bq kg^{-1} wet weight) divided by the concentration per unit volume of seawater (Bq L^{-1}) (IAEA, 2004).

Considerable assimilation of ^{99}Tc through feeding has been observed for some crustaceans and gastropods (Fowler et al., 1980; Beasley et al., 1982; Beasley and Lorz, 1986). For the intertidal herbivorous gastropod *Littorina littorea* Swift (1989) found a CF of 241 (shell excluded) due to uptake of ^{99}Tc from seawater alone, whereas estimates of CFs from field samples ranged between 1,450 and 29,000 (Table 7 in Swift (1989) and references therein). This strongly suggests that the main way of uptake of ^{99}Tc may be through feeding. Dissection studies have shown that the main part of ^{99}Tc is found in internal organs or hepatopancreas in mussels and crustaceans (Fowler et al., 1980) and in the digestive-gland-kidney complex in *L. littorea* (Swift, 1989). Elimination of ^{99}Tc is shown to follow a biphasic pattern in most studied cases, with an initial rapid phase followed by a slower phase (Masson et al., 1989).Whereas no indication of retention of Technetium through binding in metallothioneins has been found in the mussel *Mytilus*

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sp. (Verthé et al., 1986), binding of Technetium by proteins has been observed in the seastar *Marthasterias glacialis*, and this has also been suggested for the gastropod *Patella caerula* (Goudard et al., 1985; Verthé et al., 1986).

In the coastal zone not only phytoplankton, but also benthic algae contribute greatly to the primary production. Various fucoid species inhabit intertidal rocky temperate shores in the North Atlantic, and have a primary production rate which typically is within the range 300–600 g C m^{-2} y^{-1} (Brinkhuis, 1977; Cousens, 1981). This production provides food for intertidal herbivorous invertebrates and marine filter and detritus feeders of the coastal zone. Given that food can be the dominant source of ^{99}Tc in some marine invertebrates (Swift, 1989), one should expect a close relationship between the concentration of ^{99}Tc in the food and the accumulation level in the feeding organism. The fact that several of the gastropods which show high accumulation of ^{99}Tc (e.g. *P. vulgata* (Jeanmarie et al., 1981) and *L. littorea* (Swift, 1985)) are known to be grazers on seaweeds (Little and Kitching, 1996), which in turn may have high concentrations of ^{99}Tc, seems to support this.

This study was carried out in order to examine if molluscs with different food habits, collected in a sheltered intertidal dominated by fucoids, also have differential concentrations of ^{99}Tc. A number of common mollusc species of a sheltered intertidal on the southwestern coast of Norway were chosen, of which all are secondary producers and relatively long-lived. *Mytilus edulis* is a filter-feeding bivalve. *P. vulgata*, *L. littorea* and *L. obtusata* are all herbivorous gastropods, commonly occurring in sheltered intertidal. All three feed on algae or microorganisms by using a radular tongue. *L. obtusata* has the most specialized habit of the three, and is most frequently found on the fucoids *Fucus* spp. and *Ascophyllum nodosum* (Reid, 1996). Samples of the selected molluscs were collected at different seasons and ^{99}Tc-concentrations were measured. In order to give an overview of the dominating ^{99}Tc sources at the sampling site, the ^{99}Tc-concentrations of the dominating macroalgae and the seawater at the site were included.

2. Material and methods

2.1. Sampling area

The sampling of fucoids, molluscs and seawater was carried out in a sheltered intertidal about 20 km south of Bergen on the southwest coast of Norway, in a fjord system (60°16′N, 05°14′E). The site was chosen due to an abundant intertidal cover of fucoids. The zonation pattern of fucoids found at the site is typical for a sheltered intertidal in southwest Norway (Jorde, 1966) The mean difference between high and low tide is 90 cm in this area (Norwegian Hydrographical Service: http://vannstand.statkart.no/Engelsk/main.php).

In order to investigate if seasonal variations in ^{99}Tc-concentration were present in the sampled species and the seawater, the sampling was carried out at regular intervals between February and November 2006 (Tables 1 and 2). The sampling was carried out at low tide along a 20–30 m long horizontal stretch. The seawater sampling and results of ^{99}Tc-concentrations of sea water at the site are described in Heldal and Sjøtun (2010).

2.2. Sampling and sample treatment of fucoids and molluscs

The fucoids *A. nodosum* and *Fucus vesiculosus* were collected on five different sampling dates, and *Pelvetia canaliculata* and *Fucus serratus* on three sampling dates. Sample procedure and results of ^{99}Tc distribution in *A. nodosum* are described and reported in Heldal and Sjøtun (2010). For the other fucoids, bulk samples of between 0.3 and 1 kg wet weight were collected during each sampling. The samples were placed in clean plastic bags, transported to the Institute of Marine Research (IMR) and frozen. Prior to analysis, the samples were first ground using a Braun kitchen machine, then dried in a laboratory freeze dryer (Christ Alpha 1–4 or Christ Beta 1–8) and finally homogenized using a Braun kitchen machine.

The molluscs were collected mainly in the middle and lowermost part of the intertidal. *L. obtusata* and *P. vulgata* were collected on three different sampling dates, and *M. edulis* and *L. littorea* on four sampling dates. In order to test if ^{99}Tc concentration would increase with size, samples of small and large individuals were obtained on two or three sampling dates for each species. The limit between small and large *M. edulis* and *P. vulgata* was set to 45 mm bivalve length and anterior–posterior shell length, respectively. The limit between small and large *L. littorina* and *L. obtusata* was set to 17 and 12 mm, respectively, measured between the outermost part of the shell lip to the apex of the coil. All size limits were based on the first collections of the molluscs. Based on earlier experience, around 10 g dry weight of biota is needed for analysis of the content of ^{99}Tc. A large number of individuals were therefore needed for the smallest species. The numbers of individuals per sample varied from eight in *P. vulgata* to 188 in *L. obtusata*, which has the smallest size of the examined molluscs.

The samples were either kept in a refrigerator and processed the following day, or frozen and processed later. The hard parts of the

Table 1
Concentrations of ^{99}Tc (Bq kg^{-1} dry weight (d.w.) and Bq kg^{-1} wet weight (w.w.)) in fucoids, concentrations of ^{99}Tc in seawater and Concentration factors (CFs). Bq kg^{-1} w.w. is determined by multiplying (Bq kg^{-1} d.w.) with dry weight/wet weight ratios of 0.24, 0.16 and 0.23 for *Fucus vesiculosus*, *Fucus serratus* and *Pelvetia canaliculata*, respectively. These are averages of ratios calculated for each species at each sample preparation. Concentration factors are calculated from average tissue concentrations of ^{99}Tc (Bq kg^{-1} w.w.) divided by ambient concentration of ^{99}Tc (Bq L^{-1}) in seawater. The errors include the statistical counting errors and uncertainties in the analytical procedure. The analytical error covers the collective uncertainties of the gravimetric procedure, including calibration of the detector, weighing of the final precipitate and the precise physical characteristics of the source as presented for counting.

Collection date	Species	^{99}Tc (Bq kg^{-1} d.w.)	^{99}Tc (Bq kg^{-1} w.w.)	^{99}Tc (Bq m^{-3})[a]	Concentration factors (Bq kg^{-1} w.w.)/(Bq m^{-3})
22.02.2006	*Fucus vesiculosus*	125 ± 7	30 ± 2	0.75	4.0×10^4
20.04.2006	*Fucus vesiculosus*	91 ± 5	22 ± 2	0.67	3.3×10^4
19.06.2006	*Fucus vesiculosus*	122 ± 7	29 ± 2	0.67	4.4×10^4
31.08.2006	*Fucus vesiculosus*	95 ± 5	23 ± 2	0.54	4.2×10^4
16.11.2006	*Fucus vesiculosus*	96 ± 5	23 ± 2	0.62	3.7×10^4
22.02.2006	*Fucus serratus*	142 ± 7	23 ± 2	0.75	3.0×10^4
20.04.2006	*Fucus serratus*	113 ± 6	18 ± 1	0.67	2.7×10^4
19.06.2006	*Fucus serratus*	55 ± 3	9 ± 1	0.67	1.3×10^4
20.04.2006	*Pelvetia canaliculata*	192 ± 10	44 ± 3	0.67	6.6×10^4
31.08.2006	*Pelvetia canaliculata*	107 ± 5	25 ± 2	0.54	4.6×10^4
16.11.2006	*Pelvetia canaliculata*	128 ± 6	29 ± 2	0.62	4.7×10^4

[a] Reported in Heldal and Sjøtun (2010).