



## Radiocaesium ( $^{137}\text{Cs}$ ) in marine mammals from Svalbard, the Barents Sea and the North Greenland Sea

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Received 8 February 2005; accepted 27 June 2005

Available online 9 September 2005

### Abstract

Specific activities of the anthropogenic radionuclide,  $^{137}\text{Cs}$ , were determined in marine mammals from Svalbard and the Barents and North Greenland Seas. Muscle samples were collected from 12 polar bears, 15 ringed seals, 10 hooded seals, 7 bearded seals, 14 harp seals, one walrus, one white whale and one blue whale in the period 2000–2003. The mean concentrations ( $\pm$  SD) of  $^{137}\text{Cs}$  were:  $0.72 \pm 0.62$  Bq/kg wet weight (w.w.) for polar bears;  $0.49 \pm 0.07$  Bq/kg w.w. for ringed seals;  $0.25 \pm 0.10$  Bq/kg w.w. for hooded seals;  $0.22 \pm 0.11$  Bq/kg w.w. for bearded seals;  $0.36 \pm 0.13$  Bq/kg w.w. for harp seals;  $0.67$  Bq/kg w.w. for the white whale sample;  $0.24$  Bq/kg w.w. for the blue whale; and below detection limit for the walrus. Significant differences in  $^{137}\text{Cs}$  specific activities between some of the species were found. Ringed seals had higher specific activities than the other seal species in the study. Bearded seals and hooded seals had similar values, which were both significantly lower than the harp seal values.

The results in the present study are consistent with previous reported results, indicating low specific activities of  $^{137}\text{Cs}$  in Arctic marine mammals in the Barents Sea and Greenland Sea region during the last 20 years. The species specific differences found may be explained by varying diet or movement and distribution patterns between species. No age related patterns were found in specific activities for the two species (polar bears and hooded seals) for which sufficient data was available.

Concentration factors (CF) of  $^{137}\text{Cs}$  from seawater were determined for polar bears, ringed, bearded, harp and hooded seals. Mean CF values ranged from  $79 \pm 32$  (SD) for bearded seals sampled in 2002 to  $244 \pm 36$  (SD) for ringed seals sampled in 2003 these CF values are higher than those reported for fish and benthic organisms in the literature, suggesting bioaccumulation of  $^{137}\text{Cs}$  in the marine ecosystem.

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**Keywords:** Radionuclides; Caesium-137; European Arctic; Marine mammals

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## 1. Introduction

During the last few decades, anthropogenic pollution has become an important issue for research and management authorities dealing with the Arctic. A range of pollutants have been, and continue to be, transported to the Arctic from industrialised areas in temperate regions via atmospheric and oceanic circulation (Oehme, 1991; Barrie et al., 1992; AMAP, 1998). Additionally, local sources of pollutants are found in the Arctic region. All the main contamination groups (persistent organic pollutants, heavy metals, petroleum hydrocarbons, and radionuclides) are present. The characteristics of Arctic ecosystems, such as low annual productivity, lack of species diversity and short food chains increases the vulnerability of these ecosystems to the impacts of pollution (AMAP, 1998). Additionally, the tendency for Arctic marine food chains to be dependent on benthic and sea ice associated systems provides an efficient mechanism for the biomagnification of contaminants, while the longevity of marine mammals in these food chains allows for the potential accumulation of contaminants over long periods of time in these top consumers. Certain marine biota exhibit high uptake rates of radionuclides (e.g., Pentreath et al., 1982; Aarkrog et al., 1997; Brown et al., 1999), while radiocaesium ( $^{137}\text{Cs}$ ) has been shown to biomagnify through marine food chains and therefore be found in high concentrations in marine mammals (Calmet et al., 1992; Kasamatsu and Ishikawa, 1997; Watson et al., 1999; Heldal et al., 2003). Together, these observations may have important consequences for Arctic marine ecosystems if significant levels of contamination occur.

Cs-137 contamination of the Arctic marine environment takes place through global fallout from atmospheric weapon testing, discharges from European reprocessing facilities and fallout from the Chernobyl accident in 1986. Of the 543 atmospheric weapons tests conducted globally, 91 were carried out in the Arctic region by the former Soviet Union at Novaya Zemlya with a total yield of 239.6 Mt (UNSCEAR, 2000). Aarkrog (1993) estimated a level of fallout in the Arctic region of 30 PBq of  $^{137}\text{Cs}$  from 87 of these tests alone. Another important source of  $^{137}\text{Cs}$  to the Arctic marine environment has been discharges from the major nuclear fuel reprocessing facilities in Europe. These facilities include Sellafield in the United

Kingdom, Dounreay in Scotland, and Cap la Hague in France. Sellafield's discharges in the late sixties through to the mid eighties, have dominated the supply of  $^{137}\text{Cs}$  to the Arctic with an estimated 14 PBq passing into the Barents Sea and through the Fram Strait (Kershaw and Baxter, 1995). Due to stronger regulatory controls and plant improvements that have been implemented since this time, releases of many of the main radionuclides, including  $^{137}\text{Cs}$  have declined markedly in recent years. Discharges from the European reprocessing facilities are transported into the Arctic via the Norwegian Coastal Current, North Cape Current and West Spitsbergen Current with an estimated transport time of 6 to 10 years (Kautsky, 1987; Dahlgaard, 1995). It is estimated that as a result of the Chernobyl accident a total 131 PBq of radio-caesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) were released to the environment (AMAP, 1998), and some of this was deposited directly in the Arctic. In addition to direct fallout from the atmosphere, the Arctic marine environment continues to be contaminated by the oceanic transport of Chernobyl derived  $^{137}\text{Cs}$  from the North Sea and the Baltic Sea, the catchments of which both received considerably greater fallout from Chernobyl than Arctic regions. Calculations based on  $^{134}\text{Cs}/^{137}\text{Cs}$  ratios in the Kara Sea in 1992, suggested that some 30% of the  $^{137}\text{Cs}$  inventory in the Kara Sea was derived from the Chernobyl accident (Strand et al., 1994).

The monitoring of radioactivity in Arctic marine mammals is important for a number of reasons. Information on current levels of contamination is required to understand the impacts and behaviour of radionuclides in Arctic ecosystems and the potential consequences of any future contamination. Additionally, due to the subsistence harvesting of some marine mammals by arctic indigenous peoples there is a need for information on current radionuclide burdens, so that accurate assessments can be made concerning potential doses to man. Cooper et al. (2000) reported radionuclide contaminant burdens for several marine mammals in northern Alaska and Canada to be low. Similar species-specific data in the literature for marine mammals in the European Arctic is scarce. Given the potential risk for future sources of contamination to the Arctic, the purpose of the present study is to establish baseline data on specific activity of  $^{137}\text{Cs}$  in a selection of species and age-groups of marine mam-

mals from the European Arctic for reference for future monitoring.

## 2. Methods

### 2.1. Field sampling

Muscle samples (~1 kg wet weight (w.w.)) were collected from 12 polar bears (*Ursus maritimus*), 15 ringed seals (*Phoca hispida*), 10 hooded seals (*Cystophora cristata*), 7 bearded seals (*Erignathus barbatus*), 14 harp seals (*Pagophilus groenlandicus*), one blue whale (*Balaenoptera musculus*), one white whale (*Delphinapterus leucas*), and one walrus (*Odobenus rosmarus*) (Table 1). The polar bears sampled were problem bears shot throughout the Svalbard archipelago, while samples of ringed and bearded seals were collected from hunted animals on Spitsbergen, the main island in Svalbard. The white whale was an accidental netting mortality. The harp seal samples were from animals drowned in fishing nets during normal commercial fishing operations in the southern Barents Sea, while the hooded seals were collected in the northern parts of the North Greenland Sea during a scientific expedition in 2002. Samples of blue whale and walrus were collected from stranded animals on Jan Mayen. Sex and reproductive class (juvenile, sub-adult or adult) were available for most individuals (Table 1). All muscle samples were stored frozen at  $-20\text{ }^{\circ}\text{C}$  from the time of collection until analysis.

### 2.2. Analytical methods

All samples were thawed and allowed to drain for 24 h to remove excess blood. Samples were then dried

at  $105\text{ }^{\circ}\text{C}$  in a fan-assisted oven until they were at a constant weight, and then were homogenized in a stainless steel laboratory blender. The resulting material was packed into plastic containers of various sizes that are used as analytical geometries. Typical analytical sample masses were between 50–300 g. Once prepared in this way, all samples were counted on an HPGe gamma spectrometer (Canberra Industries). The samples were counted for periods between 24–48 h and corrected for background signal using the peak subtraction method.  $^{137}\text{Cs}$  was determined via its emission at 661 keV. Minimum detectable activity (MDA) limits ( $L_D$ ) were calculated according to Currie (1968). The expressed error in the gamma results (which is given at the 95% confidence level) is a standard accumulation of uncertainty sources for radiometric measurements. It can be broken down into uncertainty in calibration, weighing error, counting statistics and analyst bias. For samples with specific activities below  $L_D$ , half the detection limit was used for calculation of mean values.

Analysis of variance (ANOVA) (Kruskal–Wallis Test) and  $t$ -tests were performed to evaluate any statistical differences between species. Due to small sample sizes, age specific radionuclide values were calculated only for polar bears and hooded seals. Because of the low and variable sample sizes in each age group, statistical tests were not run to evaluate differences; however, non-overlapping 95% confidence intervals were considered to indicate statistically significant differences ( $p \leq 0.05$ ). The SAS statistical package (SAS Institute Inc. Cary, NC, USA, 1989) was used for analyses of data, and all specific activities presented are in Bq/kg wet weight (w.w.) with standard deviation (SD) being the chosen measure of variance.

Table 1  
Age, sex (F=females, M=males, U=Unknown), sampling year and location of marine mammal radionuclide samples

Species	Juveniles	Sub-adults	Adults	Total #	Year	Location
Polar bear	4 (F: 2, M: 2)	3 (F: 1, M: 2)	4 (F: 1, M: 3)	12 <sup>a</sup>	2000–2003	Spitsbergen
Ringed seal		1 (M)	14 (F: 1, M: 13)	15	2003	Spitsbergen
Hooded seal	2 (M)	3 (F: 1, M: 2)	5 (F: 4, M: 1)	10	2002	North Greenland Sea
Bearded seal			7 (M)	7	2000–2002	Spitsbergen
Harp seal			14 (F)	14	2003	Southern Barents Sea
Blue whale			1 (U)	1	2001	Jan Mayen
White whale	1 (U)			1	2000	Spitsbergen
Walrus			1 (M)	1	2000	Jan Mayen

Table 2

Mean concentrations of  $^{137}\text{Cs}$  (Bq/kg w. w.)  $\pm$  SD, ranges and analysis errors for marine mammal radionuclide samples from Svalbard and the Greenland and Barents Seas

Species	<i>N</i>	$^{137}\text{Cs}$ (mean $\pm$ SD)	$^{137}\text{Cs}$ range	$^{137}\text{Cs}$ error mean (Range)
Polar bear	12	0.72 $\pm$ 0.62	0.20–2.25	0.08 (0.03–0.09)
Ringed seal	15	0.49 $\pm$ 0.07	0.40–0.61	0.04 (0.03–0.06)
Hooded seal	10	0.25 <sup>a</sup> $\pm$ 0.11	<0.20 <sup>b</sup> –0.42	0.03 (0.03–0.06)
Bearded seal	7	0.22 $\pm$ 0.12	0.06–0.42	0.04 (0.03–0.06)
Harp seal	14	0.36 <sup>a</sup> $\pm$ 0.13	<0.20 <sup>b</sup> –0.53	0.04 (0.02–0.08)
Blue whale	1	0.24		0.04
White whale	1	0.67		0.06
Walrus	1	<0.20 <sup>b</sup>		

<sup>a</sup> When calculating mean specific activities, values below  $L_D$  were included, but these were set to half the  $L_D$ -value.

<sup>b</sup> For values below  $L_D$ , measurement error cannot be calculated.

To study the transfer of radionuclides in the marine environment concentration factors are commonly used. Concentration factors can be determined on the basis of the specific activity ratio of the radionuclide between the organism of interest and the surrounding seawater:

$$\text{Concentration factor} = \frac{\text{Concentration in biota (Bq/kg w.w.)}}{\text{Concentration in seawater (Bq/l)}}$$

Seawater concentrations employed were taken from the literature or from unpublished data from sites close to the marine mammal sampling location both in time and space. Concentration factors were determined for polar bears, ringed, bearded, harp and hooded seals and the white whale sample. Concentration factors were only calculated for polar bears where evidence of seal foraging (and thus marine foraging) was available from stomach contents. For other polar bears, either the foraging history was not known or there was suspicion of fasting or terrestrial foraging due to the time of the year during which the bears were shot.

### 3. Results

Specific activities of  $^{137}\text{Cs}$  in the muscle tissues of all marine mammals examined were generally low; many were below the limits of detection. Mean specific activities ranged between 0.22–0.71 Bq/kg w.w., with a maximum of 2.25 Bq/kg w.w. in a sample from a polar bear (Table 2). Specific activity of  $^{137}\text{Cs}$  differed significantly between species (Kruskal–Wallis Test,

$p < 0.0001$ ), with higher values in ringed seals than the other species in the study, for which sufficient data were available ( $t$ -test,  $p \leq 0.015$ ). Specific activities of  $^{137}\text{Cs}$  were similar in bearded seals and hooded seals, but values in both of these species were significantly lower than those for harp seals ( $t$ -test,  $p \leq 0.047$ ). Polar bears contained highly variable  $^{137}\text{Cs}$  specific activities, which overlapped those of the other species.

The polar bear and hooded seal samples consisted of material from various age groups, but a separation of the samples into three different age categories revealed no age-related pattern in the data (Fig. 1).

Mean  $^{137}\text{Cs}$  concentration factors for seal species ranged from  $79 \pm 32$  (SD) for bearded seals to  $244 \pm 36$  (SD) for ringed seals (Table 3). For other marine mammals,  $^{137}\text{Cs}$  concentration factors of 223 and 237

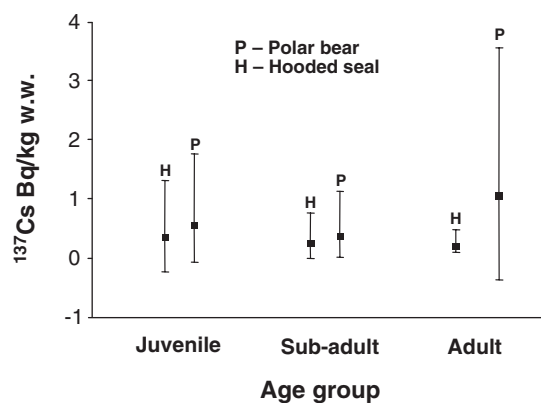


Fig. 1. Levels of  $^{137}\text{Cs}$  for sub-adult and adult polar bears and hooded seals from the European Arctic. Values presented as means with 95% confidence intervals.

Table 3

Concentration factors (CF) of  $^{137}\text{Cs}$  in muscle in marine mammals from Svalbard and the Greenland and Barents Seas

Species	<i>N</i>	Year	CF (mean $\pm$ SD)	CF range	Seawater concentration (SWC) ( $\text{Bq}/\text{m}^3$ )	SWC reference
Polar bear	2 <sup>a</sup>	2003	230	223–237	2.0	NRPA unpublished data
Ringed seal	15	2003	244 $\pm$ 36	201–305	2.0	NRPA unpublished data
Harp seal	14	2003	116 $\pm$ 28	73–155	3.4	Svaeren I. pers.comm. <sup>b</sup>
Hooded seal	8	2002	104 $\pm$ 45	40–167	2.5 <sup>c</sup>	NRPA, 2004
Hooded seal	2	2002	99	71–127	2.1 <sup>c</sup>	NRPA, 2004
Bearded seal	2	2000	167	153–181	2.3	Gärfvert et al., 2003
Bearded seal	5	2002	79 $\pm$ 32	30–117	2.0	NRPA, 2004
White Whale	1	2000	196		2.4	Povinec et al., 2003

When calculating mean concentration factors, values below  $L_D$  were included, but these were set to half the  $L_D$ -value.

<sup>a</sup> Calculated from two animals assumed to be feeding on seals.

<sup>b</sup> Svaeren I. Institute of Marine Research, Bergen, Norway.

<sup>c</sup> Seawater concentration of  $2.5 \text{ Bq}/\text{m}^3$  used to calculate CF for eight samples and  $2.1 \text{ Bq}/\text{m}^3$  used to calculate CF for two samples, due to different sampling locations.

were calculated for two polar bears and 196 for the white whale sample (Table 3).

#### 4. Discussion

Within the European Arctic region, there has been a concerted effort to study radionuclide contamination of sea water (for review, see: AMAP, 1998). However, marine organisms and especially higher trophic level animals have received little attention in this region compared with other areas in the Arctic. One reason for this might be that a wider variety of marine mammal species are hunted by indigenous people for human consumption in other localities in the Arctic, while most of these species are protected from hunting in the European Arctic.

The low  $^{137}\text{Cs}$  activities observed in the marine mammals in the present study reflect the current low  $^{137}\text{Cs}$  activities in sea water in the European Arctic, following the reduction in discharges from the reprocessing facilities at Sellafield, UK in the mid 1970s. Recently reported  $^{137}\text{Cs}$  activities in sea water from the study areas ranged from 2.0 to  $3.4 \text{ Bq}/\text{m}^3$  (See Table 3 for references) compared to peak values of 20 to  $45 \text{ Bq}/\text{m}^3$  for the Svalbard area and Barents Sea in the 1980s (Hallstadius et al., 1982; Kershaw and Baxter, 1995; Strand et al., 2002). Although a large number of potential local sources of radionuclide contamination are known in the region (dump sites of nuclear reactors and radioactive waste, atmospheric

nuclear bomb testing sites on Novaya Zemlya). However, it would appear from our data that these potential sources currently have little impact on marine mammals in the European Arctic.

Cooper et al., (2000) reported average  $^{137}\text{Cs}$  activities in muscle samples for a variety of marine mammal species from Alaska and Canada, including polar bears, ringed seals and bearded seals that were similar to those seen in this study and other studies done in the European Arctic. The global distribution of radionuclide contamination in marine mammals shows that values are generally higher in the northern hemisphere than in the southern. The highest values of  $^{137}\text{Cs}$  worldwide have been reported in grey seals (*Halichoerus grypus*) along the UK coast (3 muscle samples of 11.08, 14.3 and  $27.5 \text{ Bq}/\text{kg}$  w.w.; Anderson et al., 1999). From these areas harbour porpoises (*Phocoena phocoena*) had a mean  $^{137}\text{Cs}$  activity in muscle tissue of  $6.9 \text{ Bq}/\text{kg}$  w.w. ( $N=19$ , range from undetectable to  $66.6 \text{ Bq}/\text{kg}$  w.w.) (Watson et al., 1999). The second highest values of  $^{137}\text{Cs}$  reported are from Baikal seals (*Phoca sibirica*) with a mean muscle value of  $14.0 \pm 2.0$  (SD)  $\text{Bq}/\text{kg}$  w.w. ( $N=5$ , range from 12.0–17.0  $\text{Bq}/\text{kg}$  w.w.) (Yoshitome et al., 2003). Values of  $^{137}\text{Cs}$ , comparable to those reported by Watson et al. (1999) and Yoshitome et al. (2003) were also reported from harbour porpoises collected in the Black Sea in 1993 ( $N=5$ , mean= $9.0 \pm 2.1$  (SD)  $\text{Bq}/\text{kg}$  w.w.) (Kanivets et al., 1999). In the Eastern Tropical Pacific specific activities in three dolphin species were above detection limits in samples from

the late 1970s and early 1980s. These were collected near nuclear weapons test sites (Calmet et al., 1992). The general decrease in values towards the south is confirmed by analyses of spinner dolphin (*Stenella longirostris*) samples from the Indian Ocean collected in 1990–1991 and a Weddell seal (*Leptonychotes weddellii*) sample from Antarctica collected in 1981 where no  $^{137}\text{Cs}$  was detected (Yoshitome et al., 2003).

Cs-137 specific activities in minke whales (*Balaenoptera acutorostrata*) from 1998 ranged from 0.298 to 0.655 Bq/kg w.w. (Born et al., 2002), which is comparable to the values for the single samples of blue whale (baleen whale) and white whale (toothed whale) in this study. Previous data for  $^{137}\text{Cs}$  in polar bears in the European Arctic is limited to an average  $^{137}\text{Cs}$  activity muscle concentration from two animals from eastern Svalbard in 1980 of  $1.27 \pm 0.06$  Bq/kg (w.w.) (Holm et al., 1983; IAEA, 2004). This value is within the range observed for this species in this study, despite a 3 to 5 fold difference in  $^{137}\text{Cs}$  sea water concentrations between the 1980 sampling site (Holm et al., 1983; IAEA, 2004) and contemporary sea water concentrations. However, as mentioned previously, the bioaccumulation of  $^{137}\text{Cs}$  by marine mammals may be dependent on prey availability and feeding rates. These factors may be particularly important in the bioaccumulation of  $^{137}\text{Cs}$  in polar bears, which can display episodic feeding behaviour. This may account for the wider range of observed  $^{137}\text{Cs}$  specific activities in polar bears in this study compared to any of the seal species. In addition  $^{137}\text{Cs}$  specific activities in polar bears killed due to their proximity to man (i.e., 'problem' animals) may differ to those in animals living on the open sea-ice.

The specific activities of  $^{137}\text{Cs}$  in marine mammals are principally dependent on the concentrations within prey species and so differences in diet, especially when considering differences between seal species (Gjertz and Lydersen, 1986; Hjelset et al., 1999; Wathne et al., 2000; Haug et al., 2004), may account for some of the observed variation in  $^{137}\text{Cs}$  bioaccumulation. Yoshitome et al. (2003) reported that marine mammals feeding predominantly on fish generally showed higher degrees of  $^{137}\text{Cs}$  bioaccumulation than those feeding predominantly on cephalopods. Concentration factors for  $^{137}\text{Cs}$  recommended by the IAEA (2004) for fish ( $1 \times 10^2$ ) are one order of magnitude higher than for cephalopods ( $1 \times 10^1$ ). Addi-

tionally, differences in  $^{137}\text{Cs}$  assimilation efficiencies between species, prey availability, feeding rates and migration patterns (Folkow et al., 1996; Haug et al., 1998; Gjertz et al., 2000a; Gjertz et al., 2000b; Lydersen et al., 2004) may all have impact on the observed  $^{137}\text{Cs}$  specific activity within the muscle of a given marine mammal. In regard to other contaminants, several studies have focused on geographic variation in organic pollutants in arctic marine mammals and spatial trends have been seen within this group of compounds (e.g., Muir et al., 2000; Andersen et al., 2001). Concentration factors for all seal species in the current study ranged from  $3 \times 10^1$  to  $3 \times 10^2$ , which is lower than the recommended IAEA (2004) value for a generic seal species of  $4 \times 10^2$ . However, specific activities and concentration factors of  $^{137}\text{Cs}$  for seal species in this study are similar to a mean specific activity of  $0.23 \pm 0.04$  (SD) Bq/kg w.w. and a concentration factor range of 34 to 130 for  $^{137}\text{Cs}$  in muscle of mainly juvenile ringed, bearded and harp seals from Svalbard in 1999 (Carroll et al., 2002). In comparison, Rissanen et al. (1997) reported specific activities of 0.4 to 0.9 Bq/kg (w.w.) with a concentration factor range of 32 to 72 in muscle of harp seals (all juvenile) from the White Sea in 1995 and 1996, while Yoshitome et al. (2003) reported an average  $^{137}\text{Cs}$  specific activity of  $2.0 \pm 0.5$  Bq/kg (w.w.) and a concentration factor range of 320 to 560 for ringed seals (age unknown) from the Kara Sea in 1995.

A number of studies have shown that  $^{137}\text{Cs}$  biomagnifies through marine food chains (e.g., Calmet et al., 1992; Kasamatsu and Ishikawa, 1997; Watson et al., 1999; Heldal et al., 2003). More recently, Brown et al. (2004), utilising a biokinetic modelling approach to the trophic transfer of  $^{137}\text{Cs}$  in marine food chains, demonstrated biomagnification at lower trophic levels but not to the highest level, which was represented by the harp seal in their study. In the present study,  $^{137}\text{Cs}$  concentration factors for seal species, although often similar in magnitude, are typically higher than those reported for lower trophic levels, which suggests that  $^{137}\text{Cs}$  is biomagnified through marine food chains to these consumers. The main prey of polar bears in the study area is ringed, bearded and harp seals (Derocher et al., 2000), and the results from these species compared to data for the polar bears does not suggest biomagnification of  $^{137}\text{Cs}$ ; thus this observation is in agreement with the findings of Brown et al. (2004). It

has also been reported that radionuclide specific activities varies by age or size of the animal (Watson et al., 1999). In the present study no such patterns were found between age and  $^{137}\text{Cs}$  specific activity in polar bears and hooded seals (Fig. 1). This finding is in agreement with Born et al. (2002), who observed positive but non-significant correlations between body length of minke whales from the Northeast Atlantic and  $^{137}\text{Cs}$  specific activities.

This study has shown that  $^{137}\text{Cs}$  contamination of marine mammals in the European Arctic region is low at present. Comparison of concentration factors suggests that  $^{137}\text{Cs}$  is biomagnified through marine food chains to seal species, while the situation with regard to further trophic transfer to polar bears is unclear.

## Acknowledgements

This research was funded by the Norwegian Polar Institute and the Norwegian Radiation Protection Authority. We thank Dr. Tore Haug, Bjørn Krafft, the Governor of Svalbard and the crew of the Jan Mayen Meteorological Station for contributing samples to this study.

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