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Bio accumulation of radioactive caesium in marine mammals in the Baltic Sea – Reconstruction of a historical time series

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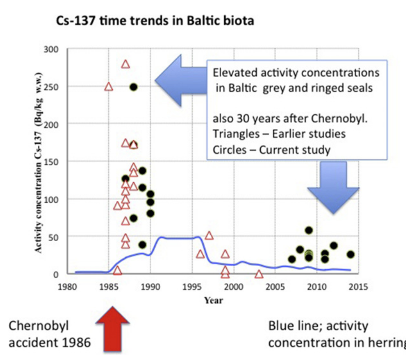
HIGHLIGHTS

- 30 years after the Chernobyl accident activity concentrations of radionuclides in Baltic marine mammals are still elevated.
- Mean activity concentrations of Cs-137 in ringed seals (*Phoca hispida*) declined from 75 Bq/kg w.w. to 30 Bq/kg w.w.
- Mean activity concentrations of Cs-137 in grey seals (*Halichoerus grypus*) has declined from 165 Bq/kg w.w. to about 30 Bq/kg w.w.
- Cs-137 biomagnifies in the Baltic Sea, seals have 3.5-9 times the activity concentrations in their main prey, herring.
- Activity concentrations of Cs-137 in Baltic marine mammals are still elevated compared to before the Chernobyl accident.

GRAPHICAL ABSTRACT



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ABSTRACT

Radionuclides from the Chernobyl accident in 1986 still circulate in the Baltic marine ecosystem and activity levels in water, sediments and fish species such as herring and perch are monitored annually. However, the activity levels of radionuclides in marine mammals have only been sporadically reported. Tissue samples from a museum collection were analysed in two species of seals, and the trends over time in activity level of radioactive caesium (Cs-137) after the Chernobyl accident were reconstructed. We also performed a literature review summarizing activity levels in marine mammals world-wide. We found activity concentrations of Cs-137 in Baltic ringed seals and grey seals to be elevated also in the most recent samples, and during the entire study period measurements ranged between 19 and 248 Bq/kg wet weight. A declining trend in time over the last 30 years follow the general trend of decline in activity levels in other Baltic biota. Accumulation was found to be species specific in the two seal species studied, with 9 times higher activity concentration in grey seals compared to herring, and 3.5 times higher in ringed seals compared to herring. We discuss potential paths and rates of bioaccumulation of radioactive caesium in the Baltic Sea including species specific prey choice of the two seal species and estimate life time exposure. The study contributes one important piece of information to predictive models in risk assessments for nuclear accidents.

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1. Introduction

1.1. Global perspective on emission of Cs-137

The radioactive isotope caesium-137 (Cs-137) in the environment originates solely from artificial sources. Due to the physical half-life of 30.2 years it is bound to disappear as production and emission ceases. Current distribution of Cs-137 in the environment is spatially patchy and can be linked to point emissions from known sources, such as collapsed nuclear power plants and fallout from atmospheric nuclear weapons tests. Currently about 450 nuclear power reactors are in operation and about 60 under construction (IAEA, 2017), so there is a theoretical risk of future emissions. Several research networks and international organizations such as the European Radioecology Alliance and the IAEA have stressed the need to investigate radionuclide routes of transfer in the environment, and the need to predict associated risks to wildlife and humans as the first strategic research challenge to focus on (IAEA, 2010; Hinton et al., 2014).

1.2. Baltic Sea and Cs-137

The Baltic Sea is the most polluted sea region in the world when it comes to Cs-137 (IAEA, 2005) and provides a full scale experiment on how radionuclides are transported in a marine environment. The input of Cs-137 to the Baltic Sea originates to about 82% from the Chernobyl accident in 1986, about 14% from nuclear weapons tests and about 4% from discharges from reprocessing facilities in Sellafield (England) and La Hague (France) (Nielsen et al., 2010). Local nuclear power plants around the Baltic contribute only 0.04% during normal operation (Iosjpe et al., 2014). Regional patterns of Cs-137 activity concentrations within the Baltic Sea show that the Bothnian Sea and Gulf of Finland are the most contaminated regions, while the central and southern Baltic Sea is less polluted. The regional variation results from different fallout patterns from the atmosphere directly after the Chernobyl accident, and different water exchanges and sedimentation processes within the main basins of the Baltic Sea (Ilus et al., 1993; Ilus, 2007).

1.3. Bio accumulation processes

Low trophic organisms are responsible for initial and direct removal of Cs-137 from water, after which bioaccumulation occurs in higher trophic level organisms through the food chain (Bossemeyer et al., 1989; Avery et al., 1991; Avery and Tobin, 1993; Perkins and Gadd, 1993; Avery, 1996; Konovalenko et al., 2017). Caesium follows the pathways of potassium ions (K^+) in living cells, and is transported into the cytoplasm by ion pumps to create an electric potential over cell membranes. This leads to higher concentrations of caesium in excitable tissues such as nerves and muscles (Watson et al., 1999; Mattsson and Thomas, 2006). A new transfer model for radionuclides in an aquatic environment, based on an ecosystem model with rates parameterized for carbon flux has been proposed by Konovalenko et al. (2017). They focus on the first steps in the food chain while the present study contributes to the understanding of processes in the highest trophic level.

1.4. Activity levels in Baltic water masses and biota

The Swedish monitoring of radioactive substances after the Chernobyl accident in 1986 has been targeting surface water, sediments, mussels and different fish species (HELCOM, 2013). Right after the accident, the activity concentration of Cs-137 in surface seawater increased from well below 15 Bq/m^3 to 100–800 Bq/m ^{3} in the different sea regions of the Baltic Sea, and remained above 100 Bq/m^3 for five years in most of the Baltic where after it started to decline. The central and eastern sea regions, the Bothnian Sea and the Gulf of Finland, were the most heavily polluted with peak concentrations over 500 Bq/m^3 (HELCOM, 2015). At present (last data point from 2014) the regional differences are wiped out and levels in sea water are about 20 Bq/m^3, just above the target level which is the pre-Chernobyl values of 15 Bq/m^3 (HELCOM, 2013; HELCOM, 2015; Ilus, 2007). It has been estimated that the activity concentrations of Cs-137 in the Baltic Sea will return to pre-Chernobyl levels by the year 2020 (HELCOM, 2013).

In mussels (*Mytilus edulis* and *Macoma balthica*), activity concentrations of Cs-137 varied between 0.6 and 9.8 Bq/kg dry weight (d.w.) in the years 2009–2011 (Zalewska and Suplinska, 2013; Iosjpe et al., 2014). Freshwater fish species such as pike, perch and roach all contained high activity concentrations that peaked 2–3 years after the accident, and values ranged between 100 and 300 Bq/kg (Ilus, 2007). The cod population in the central region of the Baltic Sea (The Archipelago Sea) had concentrations over 100 Bq/kg for five years, while activity levels in cod from other areas remained below 20 Bq/kg also in the peak years (HELCOM, 2015). In 2010, activity concentrations in marine fish (cod, herring, whiting plaice, flounder, dab) ranged between 1 and 7 Bq/kg wet weight (w.w.), and in freshwater fish (pike) it was estimated to be up to 15 Bq/kg w.w. (HELCOM, 2013; Zalewska and Suplinska, 2013; Iosjpe et al., 2014). The Cs-137 activity concentrations in herring, flatfish and surface waters have declined from peak values in 1987–1988, but are still above pre-Chernobyl levels (HELCOM, 2013).

1.5. Activity levels of Cs-137 in marine mammals

Seals have long life spans (up to 40 years) and are top predators in the Baltic Sea. Consequently, they have a high potential to accumulate radioactive compounds (Carroll et al., 2002). Despite this, very few studies have sampled seal tissue. A recent study from the Polish part of the Baltic Sea on two Grey seals (collected in 1996 and 1999) revealed levels of Cs-137 to be high (27 Bq/kg w.w.) in comparison with available data from marine mammals in Arctic, Asian and European coastal waters (Ciesielski et al., 2015), which are commonly below 1 Bq/kg w.w. (Yoshitome et al., 2003; Ilus, 2007; Ciesielski et al., 2015). A rare exception is grey seals from outside Sellafield that had about 17 Bq/kg w.w. during the peak years. (Anderson et al., 1990).

The highest Cs-137 activity concentration in muscle from seals in the Baltic Sea (280 Bq/kg w.w.) was reported by Ilus (2007) for grey seals from the SW coast of Finland (collected 1987–1988), and Ringed seals (120 Bq/kg w.w.) from Gulf of Finland (collected 1986–1988). Furthermore, a study by Holm et al. (2005) based on 22 grey seals collected between 1950 and 2002 from the Swedish east coast reports Cs-137 activity concentrations of 75–250 Bq/kg d.w. (corresponding to about 22 to 75 Bq/kg w.w., assuming a d.w. to w.w. ratio of 0.3) These early studies from Baltic marine mammals provide important information that points to the need to further investigate bioaccumulation. The present study aims at investigating and summarizing all data and to contribute new measurements and understanding about the trend in levels of Cs-137 over time of seals in the Baltic Sea.

2. Methods

Seals are exposed to caesium mainly through consumption of prey although a minor part could also be ingested with sea water (activity concentrations in sea water is however much lower compared to fish and considered negligible in our analysis). About 70% of caesium intake in seals accumulates in the muscle and the remaining 30% is taken up by other organs (Anderson et al., 1990). In seals, the lowest levels of caesium are found in bones and fat (Carroll et al., 2002). The caesium is subsequently removed from the body by metabolic processes in the kidneys.

2.1. Sample selection

We obtained twenty-two muscle tissue samples of Baltic grey seals (*Halichoerus grypus*) and Baltic ringed seals (*Phoca hispida*) from the environmental tissue bank at the Museum of Natural History (NRM) in