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^{90}Sr in *Zostera marina* from the Gulf of Gdańsk (southern Baltic Sea)

by

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Abstract

^{90}Sr activity was determined in *Zostera marina* collected in the Gulf of Gdańsk in 2008-2013. ^{90}Sr activity in *Z. marina* from Jama Kuźnicka and Klif Orłowski range from 0.83 Bq kg^{-1} d.w. to 3.78 Bq kg^{-1} d.w. and from 0.68 Bq kg^{-1} d.w. to 4.95 Bq kg^{-1} d.w., respectively. The plants collected in spring-summer showed significant differences between ^{90}Sr content in blades – 1.55 Bq kg^{-1} d.w. in 2011 and 2.18 Bq kg^{-1} d.w. in 2013, and in roots – 7.75 Bq kg^{-1} d.w. and 11.44 Bq kg^{-1} d.w., respectively. The reduction in ^{90}Sr concentrations in *Z. marina* roots to 1-2 Bq kg^{-1} d.w. in summer resulted from the transport of this element to the young parts of the plant. In autumn and winter, the ^{90}Sr content in the blades of *Z. marina* was increasing and reached the maximum of 3.77 Bq kg^{-1} d.w. This could be explained by the process opposite to dilution, related to the biomass reduction and strontium concentration in the plant tissues. Changes in ^{90}Sr concentration in *Z. marina* tissues are affected by isotope concentration in seawater as well as by salinity, which affects the concentration of Ca^{2+} – for which Sr^{2+} is a chemical analogue.

Key words: ^{90}Sr, *Zostera marina*, bioaccumulation, Gulf of Gdańsk, Baltic Sea

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Introduction

The Baltic Sea is one of the most polluted marine areas with radioactive isotope ^{90}Sr. Nuclear weapons testing in the atmosphere, carried out in the 1950s and 1960s, were the main source of ^{90}Sr in the marine environment (HELCOM 2013). At present, the major input pathways changed into surface run-off and atmospheric fallout (Saniewski, Zalewska 2016). Because of a relatively long half-life of 28.8 years and chemical similarity to calcium, ^{90}Sr is considered one of the most dangerous radioactive isotopes of anthropogenic origin (Kryshev 2006). Despite the fact that ^{90}Sr is currently becoming the most important element in the radioactivity field of the Baltic Sea (Saniewski, Zalewska 2016), information on its distribution in various elements of the marine environment is scarce. It is, however, well established that ^{90}Sr is bioaccumulated by marine flora and fauna (Solecki, Chibowski 2002; Zheleznov et al. 2002; Outola et al. 2009; Starichenko 2011).

This article presents an attempt to investigate the bioaccumulation efficiency of ^{90}Sr in the eelgrass (*Zostera marina*) tissues. *Zostera marina* is a common vascular plant in the southern Baltic Sea region (Kruk-Dowgiałło 1998; Pliński, Jóźwiak 2004). It occurs in large number also in other seas, e.g. the North Sea and along the Atlantic from the northern Norwegian coasts down to Spain (Barum et al. 2004). Interestingly, the plant plays the role of an environmental engineer – seagrasses provide a substrate for the attachment of epiphytic organisms, stabilize the sediment and facilitate the process of organic matter accumulation in the sediment. As a result they form a habitat for a large set of organisms (Gacia et al. 2003; Barum et al. 2004; Bouma et al. 2005). The abundance and diversity of the fauna and flora living in seagrass meadows are consistently higher than those of adjacent unvegetated areas. A number of animals feed on *Zostera marina* – from invertebrates to mute swan and mallard (Barum et al. 2004). It is estimated that in the previous century, the global loss of seagrass from the direct and indirect human impact amounted to 18% of the documented bottom area covered with seagrass (Barum et al. 2004). In 1957-1988, dense seagrass meadows in the Puck Lagoon (the southern Baltic Sea) were nearly completely replaced by other vascular plants e.g. *Zanichellia palustris* and filamentous brown algae species like *Pylaiella litoralis* and *Ectocarpus siliculosus* (Kruk-Dowgiallo 1991). Currently, a growing tendency of eelgrass recovery is observed and its expanding distribution at many sites in the Gulf of Gdańsk (Jankowska et al. 2014).

Taking into account the ecological importance of *Zostera marina* for the functioning of the Gulf of Gdańsk ecosystem and relatively high concentrations of ^{90}Sr in the marine environment of the southern Baltic Sea, the study was conducted with the objective to determine the radioactivity concentration of this isotope in the eelgrass, with a special focus on its distribution in various parts of the plant, like roots and blades, as well as to determine spatial and temporal differences in ^{90}Sr content in plant tissues.

Materials and methods

Sampling

Zostera marina was collected at two locations in the Gulf of Gdańsk in the southern Baltic Sea: at Klif Orłowski (KO), at a depth of 3 meters, and in Jama Kuźnicka (JK), at a depth of 2 meters (Fig. 1). The sampling location in Jama Kuźnicka was situated in the very shallow and sheltered internal Puck Lagoon while the sampling site close to Klif Orłowski (outer Puck Bay) is characterized by more intense water dynamics. Samples of plants at different stages and condition were collected by a scuba diver from the seafloor at a definite depth of 2-3 m in the period of 2008-2013. The mean contribution of *Z. marina* in the total macrophyte biomass collected at the definite depth reached ca. 10% in Jama Kuźnicka and 15% at Klif Orłowski. Samples placed in the field into plastic bags were transported to a land laboratory for further processing and analyses.

Analysis

Prior to analysis, *Z. marina* samples were ashed at a temperature of 450°C in a muffle-type furnace. ^{90}Sr activity was determined by the radiochemical method. The ashed samples were digested with concentrated nitric acid on a hotplate to extract strontium to the liquid phase. After digestion, the residue was separated on a hard filter paper and discarded. The filtrate was diluted with distilled water to 150 ml. Next, 100 ml of 8% oxalic acid, 20 mg of natural strontium, and ammonia (to raise pH to 4.0-4.5) were added to the diluted filtrate. The solution was heated to 80°C to complete the precipitation of strontium oxalate. The precipitate was collected on a hard filter paper and allowed to dry at ambient conditions. The oxalate was then converted to carbonate at 650°C in the muffle furnace. Next, strontium carbonate was isolated from calcium carbonate with 65% HNO_3. Radium was removed from the samples by precipitation with $BaCrO_4$ in the presence of a buffering agent (pH = 5.5).

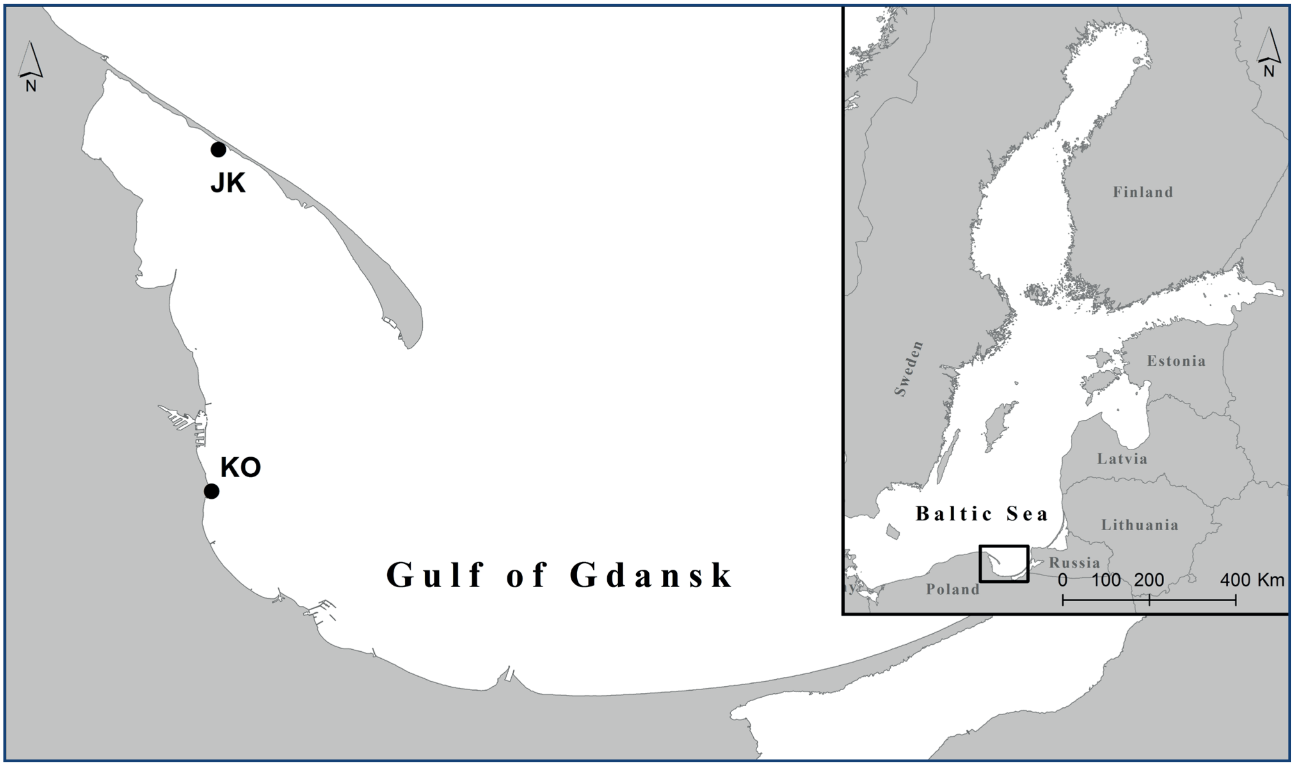

Figure 1

Sampling sites for macrophytobenthos

At this stage, 20 mg of stable yttrium was added, and the samples were allowed to stay for 21 days to reach the equilibrium between ^{90}Y and ^{90}Sr (Volchock et al. 1957). Yttrium was then precipitated as hydroxide, converted to oxalate, and collected on a pre-weighted filter. Beta activity of the samples on filters was measured using Low-Level Beta Counter FHT 7700T (ESM Eberline) with the background count rate of 0.01 counts s^{-1} and the lowest detectable activity of 3 mBq per sample. The measurement time of each sample was 21 600 seconds.

The reliability and accuracy of the applied method of ^{90}Sr determination were verified by HELCOM-IAEA-446 Proficiency Test on the Determination of Radionuclides in the Marine *Fucus* Sample (Laboratory no 7) (Table 1) (IAEA 2013; Pham et al. 2014).

Result and discussion

In the period of 2008-2012, the mean concentration activity of ^{90}Sr in *Z. marina* from Jama Kuźnicka reached 1.69 Bq kg^{-1} d.w. in the whole plant (blades and root). The minimum activity of 0.83 Bq kg^{-1} d.w. was measured in June 2010, while the maximum activity of 3.78 Bq kg^{-1} d.w. was recorded in September of the same year (Fig. 2).

In the Klif Orłowski area, the mean ^{90}Sr activity concentration was slightly higher and reached 2.40 Bq kg^{-1} d.w., however, the overall range of activity concentrations was nearly twice as high compared to plants from Jama Kuźnicka; this difference was not statistically significant (test U-W, p = 0.34). In the Klif Orłowski area, the minimum concentration activity of ^{90}Sr, 0.68 Bq kg^{-1} d.w. in the whole plant, was measured

Table 1

Evaluation results for ^{90}Sr in HELCOM-IAEA-446 Proficiency Test on the Determination of Radionuclides in Marine *Fucus* Sample – Laboratory No7. Institute of Meteorology and Water Management, Gdynia, Poland (IAEA 2013)

Analyte	IAEA Value (Bq kg^{-1} d.w.)	IAEA Unc. (Bq kg^{-1} d.w.)	Lab. Value (Bq kg^{-1} d.w.)	Lab. Unc. (Bq kg^{-1} d.w.)	Lab. Unc. (%)	Rel. Bias (%)	Accuracy	P (%)	Precision	Final Score
^{90}Sr	5.1	0.2	5.07	0.31	6.1	0.6	Passed	7.3	Passed	Accepted

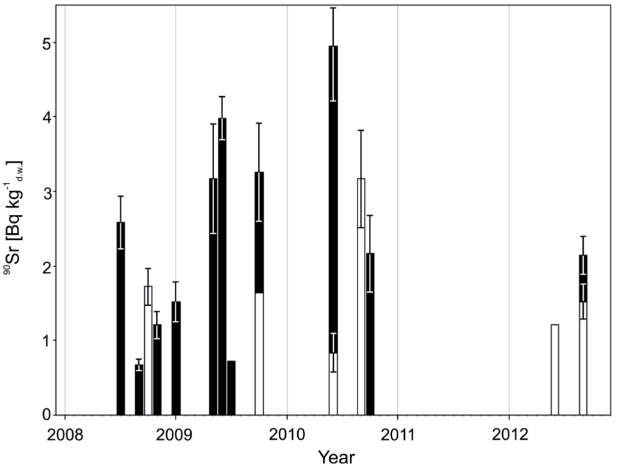

Figure 2

^{90}Sr activity in *Zostera marina* at the site KO (black bars) and JK (white bars); whiskers represent standard deviation

in September 2008, and the maximum one – 4.95 Bq kg^{-1} d.w. – in June 2010 (Fig. 2). The levels of radioactive activities of ^{90}Sr in *Z. marina* at both sampling locations were somewhat lower or comparable to those observed in other vascular plants from the Gulf of Gdańsk, e.g. *Potamogeton* sp. – 4.56 Bq kg^{-1}d.w. in June and 6.27 Bq kg^{-1} d.w. in September, *Zannichellia palustris* – 3.92 Bq kg^{-1} d.w. (September) (Zalewska 2015). Analogically, the mean ^{90}Sr concentrations in *Z. marina* were not significantly different compared to the concentrations observed in macroalgae species growing in the same areas, e.g. the mean value (range) – *Ectocarpus siliculosus* – 3.67 Bq kg^{-1} d.w. (2.02 Bq kg^{-1}d.w. – 8.71 Bq kg^{-1} d.w.), *Furcellaria lumbricalis* – 2.76 Bq kg^{-1} d.w. (0.62 Bq kg^{-1} d.w. – 5.93 Bq kg^{-1} d.w.), *Polysiphonia fucoides* 3.66 Bq kg^{-1} d.w. (0.81 Bq kg^{-1} d.w. – 15.24 Bq kg^{-1} d.w.) and *Cladophora glomerata* 1.36 Bq kg^{-1} d.w. (0.36 Bq kg^{-1} d.w. – 2.61 Bq kg^{-1} d.w.) (Zalewska 2015).

^{90}Sr activity concentrations in *Z. marina* showed a statistically significant correlation ($r = 0.63$, $p = 0.009$) with the isotope content in seawater in the study period, despite the data pool being not very abundant ($n = 16$). Higher activity concentrations of ^{90}Sr in *Z. marina* measured in 2010 are most probably related to the enhanced strontium activity in surface water of the Gulf of Gdańsk, reaching 8.3 mBq dm^{-3} as compared to 6.4 mBq dm^{-3} – a mean in the other years of the study period.

Salinity is considered another important factor in ^{90}Sr bioaccumulation in *Z. marina*. Despite the meagre data set (n=16), a statistically significant correlation was determined between ^{90}Sr activity concentrations in the whole *Z. marina* plant and salinity: $r = -0.56$, $p < 0.02$. The inverse proportionality seems to result directly from the concentrations of Ca^{2+} cations, amounting to ca. 1.2% of the total ion mass in seawater. Although Sr is apparently not a plant micronutrient, it is absorbed following the plant's metabolic requirements for Ca. Interactions between Sr and Ca are complex, and the elements can compete with each other. Multiple experiments demonstrated strong antagonistic interactions between ^{90}Sr bioaccumulation and calcium ions concentration in water (Kabata-Pendias, Pendias 2001; Kabata-Pendias, Mukherjee 2007; Smith et al. 2009), supporting the assumption that a salinity increase results in the decline of strontium phytoavailability.

In 2011 and 2013, ^{90}Sr activity was determined in particular parts of *Z. marina*: in blades and roots. The highest activities of ^{90}Sr were determined in eelgrass roots collected in June of both years (Fig. 3) – they amounted to 7.75 Bq kg^{-1} d.w. and 11.44 Bq kg^{-1} d.w., respectively. The minimum concentration of 1.09 Bq kg^{-1} d.w. in *Z. marina* roots was measured in October 2011.

As regards the eelgrass leaves, the lowest activity concentration, i.e. 0.68 Bq kg^{-1} d.w., was recorded in October 2011. On the other hand, the maximum activity (3.77 Bq kg^{-1}) was measured at the beginning of December 2011. In vascular plants, assimilation of nutrients is accomplished via the root system as well as via leaves (Barum et al. 2004). However, it is assumed that nutrient uptake by roots prevails over the uptake by leaves (Carignan, Kalff 1980). Taking into account the main transport route of nutrients assimilated from

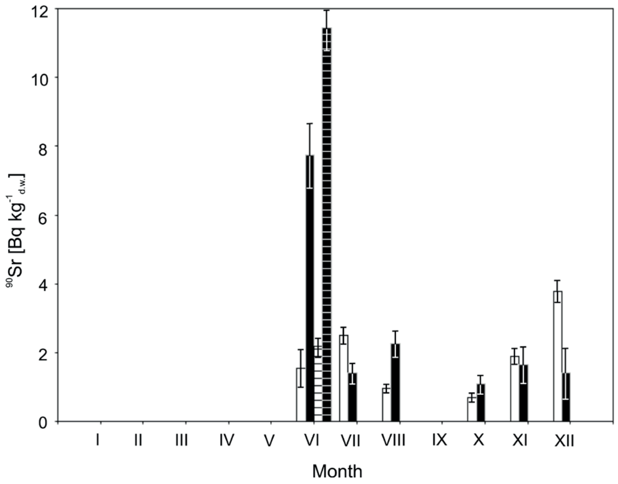

Figure 3

^{90}Sr activity in tissue in 2011: leaf (white bars), root (black bars) and in 2013 leaf (white bars with stripes), root (black bars with stripes); whiskers represent standard deviation the aquatic environment and the huge difference between ^{90}Sr concentrations in seawater (6.4 mBq dm^{-3}) and in the substrate [sand] (376 mBq kg^{-1} d.w.) where the root system is fixed, root uptake may be regarded as the main reason for higher strontium concentrations in the roots than in the *Z. marina* blades. It should be emphasized, however, that such considerable differences were found in June, i.e. they can be related also to the plant seasonal development cycle. In spring and summer, metabolic and growth processes in plants reach the peak intensity, hence the efficiency of nutrient assimilation from the surrounding aquatic environment and the accumulation in plant tissues intensify as well. Already in July, the mean ^{90}Sr activity in the eelgrass roots decreased to only 1.58 Bq kg^{-1} d.w. It is highly likely that the observed decrease was caused by transport both macroelements and metals to newly developed plant tissues. In summer, ^{90}Sr decreased also in blades – to 2.49 Bq kg^{-1} d.w in July and even 0.96 Bq kg^{-1} d.w. in August. Consequently, ^{90}Sr concentration in eelgrass leaves increased from October to December. A similar observation was made for ^{137}Cs bioaccumulation in the whole *Z. marina* plant (Zalewska 2015). The observed final result of the bioaccumulation efficiency is a result of a number of processes, frequently of opposite directions. In summer, the increase in the bioaccumulation rate results from the intensified life processes – the plant biomass increases, and this may cause the so-called dilution effect, manifested as a decrease in the element's concentration in plant tissues. In autumn, when the plant metabolism is still intense, the biomass declines and this may result in the concentration increase (Zalewska 2015).

Despite the fact that ^{90}Sr activities in eelgrass leaves are lower than in the roots, their specific concentration coefficient (CF = concentration in tissue/concentration in water) reached 200 and was much higher than in the roots (CF = 10). This observation proves that the transfer of the isotope occurs from the roots to plant leaves.

Conclusions

The mean activity concentrations of ^{90}Sr in the whole *Z. marina* plant were 1.69 Bq kg^{-1} d.w. in Jama Kuźnicka and 2.40 Bq kg^{-1} d.w. in Klif Orłowski. The measured activities were lower or similar to activity levels observed in other vascular plants and macroalgae occurring in the Gulf of Gdańsk in the study period (2008-2013).

^{90}Sr concentrations in *Z. marina* tissues are affected by the element's availability in the plant habitat (seawater, interstitial water of the substrate), as well as the plant physiological activity (metabolism, growth processes) closely related to the sampling season and external conditions. Bioaccumulation efficiency of *Z. marina* can be affected by salinity and inherent change in Ca^{2+} concentration, as calcium is the natural analogue of strontium.

The maximum activities of radioactive ^{90}Sr isotope were recorded in eelgrass roots in spring time. In summer, strontium activity in plant roots significantly decreased, probably due to the transportation of macroelements to the newly developed shoots.

The maximum concentrations of ^{90}Sr in the blades of *Z. marina* were measured in late autumn and winter, probably because of the plant biomass reduction and the concentration effect. In summer, due to the biomass increase, ^{90}Sr activity in eelgrass leaves decreased as a result of the dilution effect.

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