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On seasonal variations of radiocesium speciation in the surface sediments of Lake Juodis, Lithuania

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Abstract

Data on radiocesium speciation measured seasonally in black silt surface sediments in the shallow terrace and in the deepest southern part of Lake Juodis (Lithuania) in 2004–2005 are presented. It is shown that seasonal variations of radiocesium exchange-able fractions in the sediments in the shallow terrace are mainly related to its redistribution due to the vital cycle of green algae covering bottom surfaces – processes of their growth and decomposition. A seasonal course of those fractions in surface sediments of the deepest bottom areas of the lake follows a distinct pattern and is annually recycling. It is maximum in winter and declines due to stagnant bottom water zone disruption in summer. It is shown that surface sediments in the deepest bottom areas of the lake are not a radiocesium source during the formation of the anaerobic zone in bottom water in winter, and act in that case as its acceptor. The radiocesium remobilization to the bottom water is seasonal and site-specific. It is suggested to be due to three main processes: its diffusion from deeper and more active sediments, its redistribution during decomposition of organics freshly accumulated in surface sediments and a radiocesium flux induced by the mechanism of the seasonal anaerobic zone formation in near-bottom water. © 2007 Elsevier Ltd. All rights reserved.

Keywords: Radiocesium; Lake; Sediments; Speciation; Seasonal variations; Remobilization

1. Introduction

Speciation of radiocesium and other trace metals in soils and sediments is widely used to estimate their behavior, mobility and bioavailability as well as their sources (Alberts et al., 1989; Martinez-Aguirre et al., 1994; Cundy and Croudace, 1995; Askbrant et al., 1996; Bunzl et al., 1998; Forsberg et al., 2001; Hlavay et al., 2004; Korfali and Davies, 2005; Deryagin et al., 2006). Physico-chemical form separation techniques are mainly based on a sequential chemical extraction method of Tessier et al. (1979). It is known (Kheboian and Bauer, 1987; Schultz et al., 1998) that due to a large number of serious drawbacks, this method is only qualitative and operationally defined. Nevertheless, a sequential extraction method is accepted as a valuable tool analyzing trace metal complexes with the environmental sample fractions of different stability (Hirner, 1992; Schultz et al., 1998). An assessment of radiocesium mobility in aquatic systems is related to the determination of its exchangeable (mobile) and potentially mobile fractions (associated with carbonates, iron and manganese oxides and organics) in surface sediments (Forsberg et al., 2001; Lucey

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et al., 2007). Usually these investigations are irrespective of possible seasonal changes in sediment samples under study. However, this approach is insular. For example, the radiocesium exchangeable fraction in surface sediments of the Nemunas river accumulation zone in a small bay at the beginning of the Kaunas man-made basin was found to be minimum during warm season phytoplankton blooms in the water column and maximum in winter (Tarasiuk et al., 1999). Korfali and Davies (2005) have shown that a study of seasonal variations of trace metal speciation in river-bed sediments may be useful in the assessment of its self-purification.

The aim of the present work is to show that seasonal variations of radiocesium speciation in surface sediments of Lake Juodis are typical of this eutrophic lacustrine system and are site-specific. They are shown to be related to seasonal changes in standard water variables of the lake and to be very indicative of processes of radiocesium remobilization from the bottom sediments as well as lake self-cleaning.

2. Materials and methods

2.1. Object of the study

Lake Juodis (54° 46' 49" N, 25° 26' 29" E) is located 17 km to the northeast from Vilnius city in a wooden region (Fig. 1). It is a running shallow eutrophic lake in a lake chain connected by a brook. The inflows reach their maximum values ($\sim 80 \text{ L s}^{-1}$) in spring, in summer after rains they vary in the range of $20-30 \text{ L s}^{-1}$, and during dry periods they may decrease down to $7-10 \text{ L s}^{-1}$. The lake has no groundwater feeding sources. Its banks are rush-grown, with large marshy zones formed at the brook inflow and outflow areas. The Lake Juodis basin is of the glacier origin (groove type) and consists of two parts: the southern part of the lake is wider and deeper (up to 3.5-m deep) and the northern one is a shallow bottom terrace (depth $\sim 1.0-1.6$ m). The lake is very calm and screened from wind by the pine forest. The surface area of the lake is equal to 0.1 km^2 , drainage basin -3.5 km^2 , and hydraulic retention time is ~ 0.3 year. The water level of the lake is controlled by a Beaver Dam (Fig. 1). The typical amount of dry materials in the surface layer of the sapropelic type sediments in this lake varies in the range of $23-30 \text{ g L}^{-1}$. The organics content of sediments was assessed to be very high (~68%) (Tarasiuk et al., 2002). The thickness of the sediment layer in the northern shallow part of the lake was evaluated to be about 5-7 m (Koviazina et al., 2003). The sedimentation rate in the lake was shown to be related to the vital cycle and possibilities to grow of green algae covering all open bottom surfaces. Its maximum values were measured on the terrace (up to $\sim 6.6 \text{ mm y}^{-1}$) (Tarasiuk et al., 2007). Photosynthetic activity of green algae in the shallow bottom areas oxygenated all over the year induced the formation of carbonate deposits. These deposits were shown to be acting as a radiocesium barrier preventing its remobilization to the bottom water (Tarasiuk et al., 2007). The mineral content of the surface water is maximum in winter ($\sim 174 \text{ mg L}^{-1}$).

Radioactive pollution in the lake was caused by fallout of nuclear weapons testing and the Chernobyl accident in 1986. Processes of the lake self-cleaning from radiocesium have been studied since 2000. The mean value of ¹³⁷Cs enrichment coefficient (ratio of water-soluble ¹³⁷Cs activity concentrations in the brook outflowing from the lake to that of inflowing one) was determined to be equal to 2.1 for the period of 2000–2001. The annual course of seasonal values of this coefficient showed them to be maximum in autumn (~3.1) and minimum in winter (~1.5) (Tarasiuk et al., 2002).

Water-soluble radiocesium activity concentrations measured seasonally in the outflowing brook varied in the range of $2.1-6.8 \text{ Bg m}^{-3}$ in 2000–2001. Maximum values (4.7–6.8 Bg m⁻³) were reached in autumn.

2.2. Water and sediment sampling and preparative procedures

Ten-litre water samples (34 - at all) were collected seasonally in 2004–2005 at two stations: in the southern deepest part of the lake (station 1) and in the shallow bottom terrace in the areas of black silt sediments (station 2), over the carbonate barrier zone (Tarasiuk et al., 2007) of the bottom terrace (fairway), before the old Beaver Dam and in the inflowing and outflowing brooks (Fig. 1). Additional water sampling was carried out on 2 June 2006. In the temperature-stratified water column (in winter and spring) in the southern part of the lake, water samples were taken from different depths using the Molchanov type bathometer. In this case, parameters of water samples were averaged over the 40-cm depth interval of the sampler. The bottom water sample on the shallow terrace was taken only in winter over the area of black silt sediments. In summer and autumn under good mixing conditions of the water column, only surface water samples were collected.

After delivery to the laboratory all water samples (aerobic and anaerobic) were passed through the Filtrak 391 type filters using a vacuum pump system. This procedure aimed to avoid sediment particles possibly involved to the water samples. Further, water aliquots were evaporated on a water bath to get dry deposits that were analyzed for the radiocesium content.

Sediment cores were taken seasonally beside station 1 (depth interval -3.0-3.4 m) and beside station 2 (depth interval -1.5-1.6 m) in 2004–2005 using the Ekman–Birge type sampler. It was a steel tubing (20-cm height with a square cross-section of 15×15 cm) and a manually operated spring bottom shutter. The sampling was carried out with the weight compensation, where

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