Radiocarbon distribution in sediments of the cooling pond of the RBMK type nuclear power plant

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Introduction

Radiocarbon is currently one of the main radionuclides discharged by nuclear industry. ^{14}C produced in a reactor can be released directly to the environment from the coolant and moderator in a gaseous form or in much smaller quantities as liquid effluents [1]. The Ignalina Nuclear Power Plant (INPP), situated in the northeast of Lithuania, operated two RBMK-1500 units (design electric power 1500 MW_e): Unit 1 started operation in December 1983 and was shut down on December 31, 2004, whereas Unit 2 started operation in August 1987 and was shut down on December 31, 2009.

In this work we investigated the cycle of radiocarbon from a nuclear power plant in a cooling pond - Lake Drūkšiai (Fig. 1). ^{14}C-specific activity measurements were performed in the pelagic fish scales and the bottom sediment core layers. A collection of pelagic fish, the planktivorous vendace (it feeds exclusively on zooplankton, so the pathway for ^{14}C accumulation in this fish is expected to be dissolved inorganic carbon → phytoplankton → zooplankton → planktivorous vendace) was collected throughout the lifetime of the nuclear power plant. We also combined data from previous studies, i.e., the distribution of ^{14}C in tree rings from the vicinity of the nuclear power plant indicating $^{14}CO_2$ discharges to the atmosphere [2] and ^{14}C measurements in dissolved inorganic carbon form from the surface water [3].

Nuclear power plants are usually built along rivers or the sea shores, water residence time is very short and sediment layers are disturbed. This complicates the investigation of radionuclide distribution in the aquatic ecosystems. The Ignalina Nuclear Power Plant is a rare case when lake water is used for cooling. Thus, Lake Drūkšiai is an exceptional case to assess the impact of a nuclear facility on the aquatic ecosystem with a sufficiently high temporal resolution.

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Materials and methods

Sediment chronology for the last 66 years was determined using the ^{210}Pb technique supported by anthropogenic ^{137}Cs as a chronostratigraphic marker.

Alkali-soluble and alkali-insoluble organic fractions in sediment samples were extracted using the acid-base-acid (ABA) method, including sequential washes with 1 M HCl (overnight), 0.2 M NaOH (for 1 h), and 1 M HCl (for 1 h) to yield the alkali-insoluble fraction [4]. The planktivorous vendace scales, obtained from sets of 5 mature fishes (with ages of 2+ y) caught in summer in 1980–1999 and 2005–2012, were decalcified by immersing them into 1.2 N HCl for 2 min [5]. All samples were graphitized using Automated Graphitization Equipment AGE-3 (IonPlus AG) prior measurements with a 250 kV single stage accelerator mass spectrometer (SSAMS, NEC, USA) at the Center for Physical Sciences and Technology in Vilnius. The background of measurements was estimated to be 2.45×10^{-3} f_M using phthalic acid. The IAEA-C3 standard was used as a reference material (the percent of a modern carbon (pMC) value of 129.41). The $^{14}C/^{12}C$ ratio was measured with an accuracy better than 0.3%. All ^{14}C-specific activity measurements are reported in units of pMC [6, 7]:

$$\mathrm{pMC} = \frac{A_{SN}}{A_{ON}} \times 100\%,$$

where A_{SN} is the specific activity of the sample A_S, normalized to $\delta^{13}C = -25‰$; A_{ON} is the normalized activity of the standard.

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[5] Perga, M. E. et al. Using the $\delta^{13}C$ and $\delta^{15}N$ of Whitefish Scales for Retrospective Ecological Studies: Changes in Isotope Signatures during the Restoration of Lake Geneva, 1980-2001. *J. Fish Biol.* 2003, *63* (5), 1197–1207. https://doi.org/10.1046/j.1095-8649.2003.00239.x.
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[7] Donahue, D. J. Et al. Isotope-Ratio and Background Corrections for Accelerator Mass Spectrometry Radiocarbon Measurements. *Radiocarbon* 1990, *32* (2), 135–142. https://doi.org/10.1017/S0033822200040121.

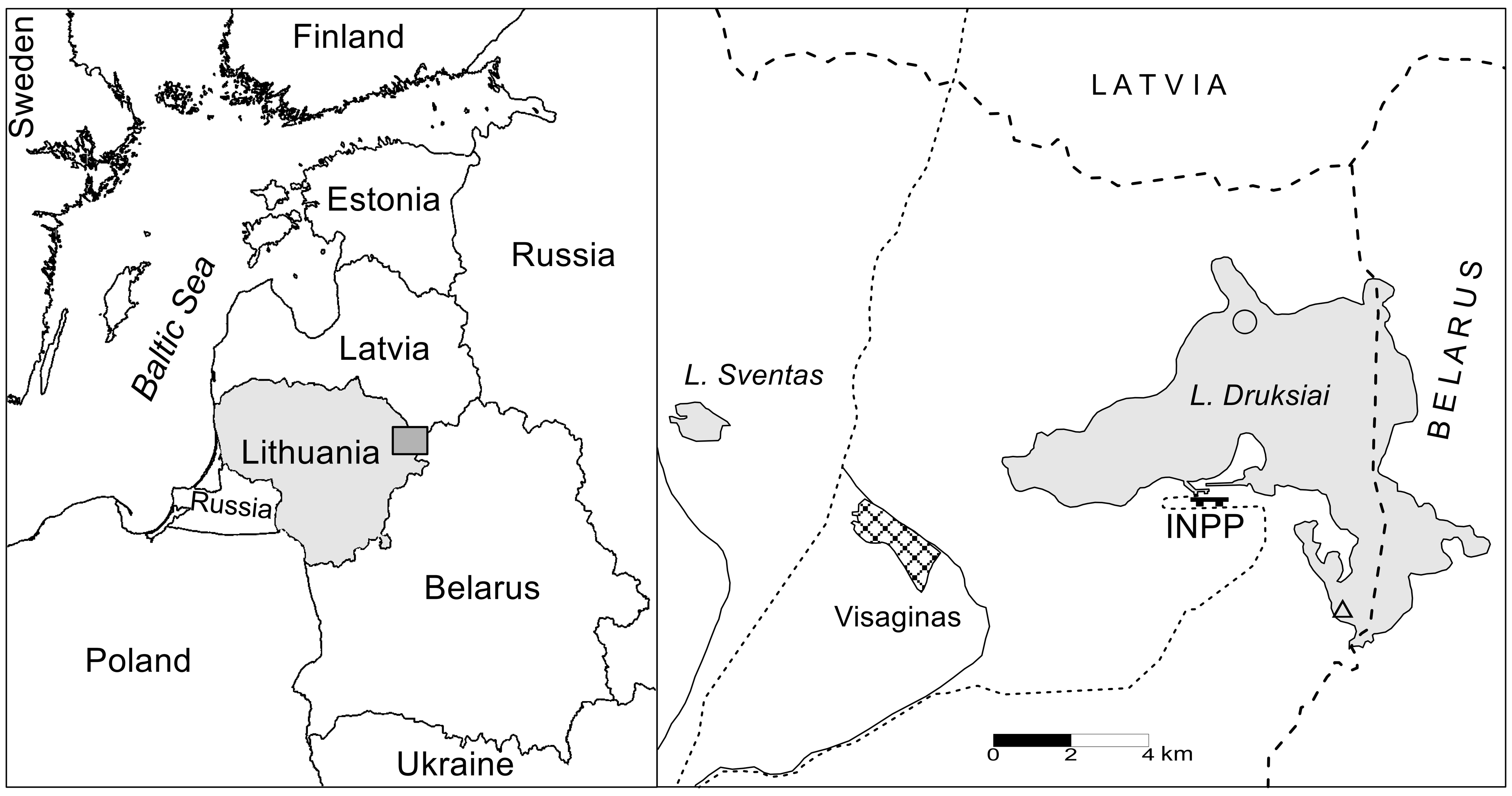

Fig 1. Location of the study area with sampling sites in Lake Drūkšiai. The sediment core site and water sampling site are marked by a circle and a triangle, respectively.

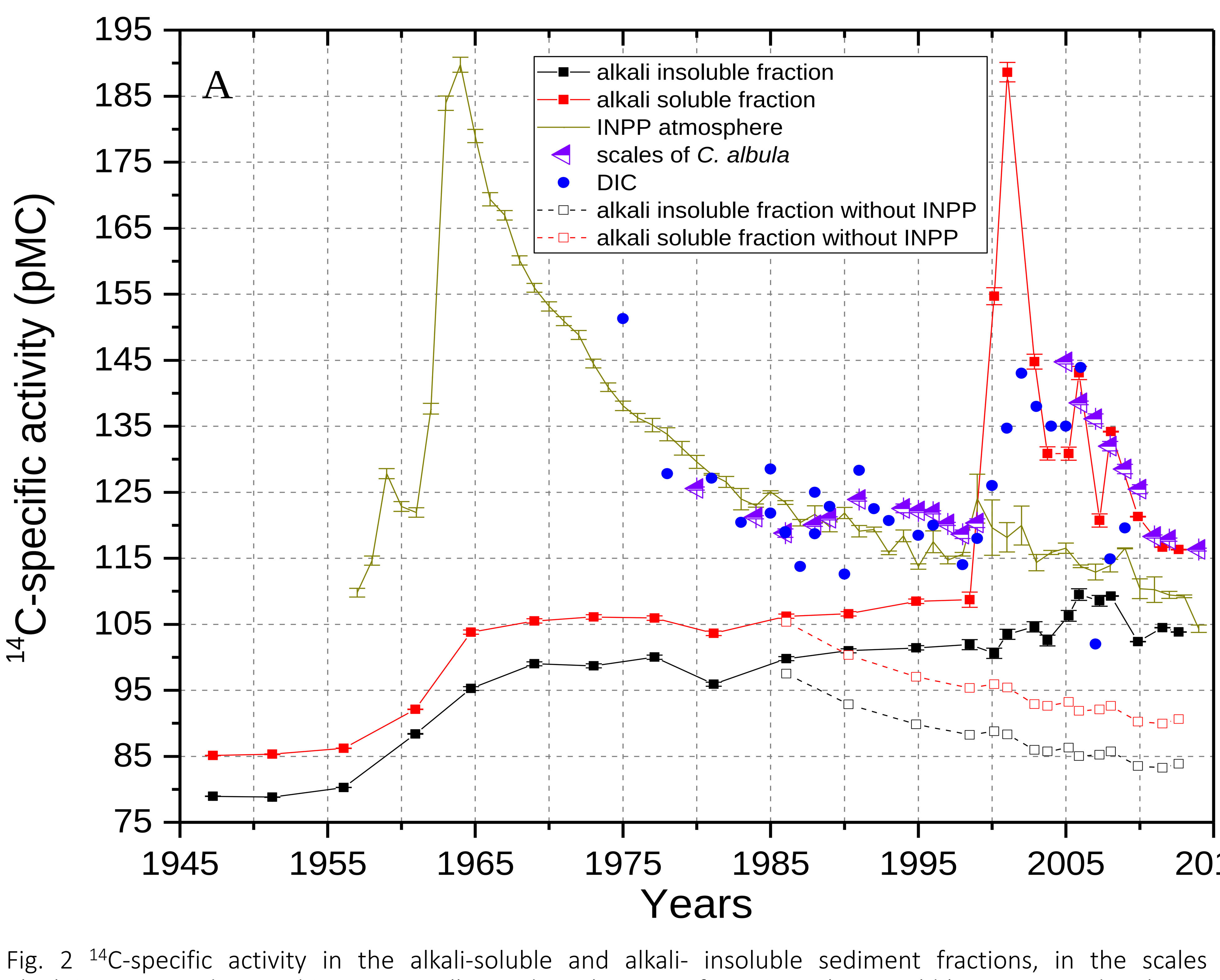

Fig. 2 ^{14}C-specific activity in the alkali-soluble and alkali- insoluble sediment fractions, in the scales of planktivorous vendace and in DIC as well as radiocarbon specific activity that would have occurred without any INPP impact.

Conclusions

C/N measurements in sediments show that the main contribution (~ 86-88 %) of organic fraction in sediments throughout the study period is of autochthonous origin. Thus, both before the commissioning of the INPP and during the first 15 years of its operation with minor maintenance activities, the radiocarbon-specific activity in both fractions of bottom sediments exhibits the parallel course, and undisturbed chain *dissolved inorganic carbon → aquatic primary producers → sediments* governed migration of radiocarbon in Lake Drūkšiai sediments (Fig 2). ^{14}C redistribution observed in the last 11 years of operation of the INPP in both organic sediment fractions indicates that another ^{14}C fractionation pathway has taken place. This might be explained by the new processes of waste management and treatment implemented at the INPP in late operational period before the final shutdown of Unit 1 (end of 2004) and Unit 2 (end of 2009).

The gaseous discharges from the INPP with RBMK type reactors are mainly carbon dioxide containing ^{14}C. Although systematic monitoring of ^{14}C airborne releases started in 2008 only, it can be relatively well reproduced by ^{14}C measurements in tree rings near the INPP [8]. Routine monitoring of radiocarbon in liquid releases was not performed, but it is expected that radiocarbon releases from RBMK type reactors is in dissolved inorganic carbon form. However, complex investigations of the radiocarbon activity in alkali-soluble and -insoluble fractions of sediment core layers indicating changes in ^{14}C redistributions between sediment fractions can retrospectively reveal facts of occurrence of elevated liquid radiocarbon releases, not only in DIC but also in dissolved organic compounds which might have been used in processes of maintenance and decontamination of technological circuits of the plant.

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