Трансформация экосистем
Ecosystem Transformation

ISSN 2619-0931 Online
www.ecosysttrans.com

DOI 10.23859/estr-230109
EDN HJOXYS
UDC 574.589+579.26

Article

Biotransformation of butyltin compounds and microbiome diversity in the bottom sediments of the Gulf of Finland, the Baltic Sea

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Abstract. Pollution of the marine environment with organotin compounds is a serious environmental problem due to their extreme toxicity to a wide range of organisms. A high level of contamination with butytin compounds was detected in the bottom sediments of the Koporye Bay, the Gulf of Finland, the Baltic Sea. The concentration of tributyltin and dibutyltin reached up to 35.7 ng(Sn)/g DW and 9.7 ng(Sn)/g DW, respectively. The butyltin compounds transformed in bottom sediments due to the activity of autochthonous sediment microbiota. The degradation rate of tributyltin and dibutyltin was 0.014 day^{-1} and 0.022 day^{-1}, their half-life, 49.5 and 31.5 days, respectively. Metagenomic analysis revealed the changes in taxonomic composition and the decrease in species diversity of the bottom sediment microbiomes during the transformation of organotin compounds. During the biodegradation of butyltin compounds in bottom sediments, the abundance of bacteria of the genera *Acidithiobacillus*, *Halothiobacillus*, *Alicyclobacillus*, *Sulfurospirillum*, *Denitrovibrio*, as well as of methanogenic archaea of the genus *Methanolobus* increases, which may indicate their potential participation in the biodegradation of organotin compounds. These results may be used to identify active tributyltin degraders for developing self-purification methods of bottom sediments contaminated with organotin compounds.

Keywords: tributyltin, dibutyltin, monobutyltin, biotransformation, microbial diversity, bottom sediments

Funding. The work is carried out within the framework of the State Assignment of the Ministry of Science and Higher Education of the Russian Federation (no. 122041100086-5).

Acknowledgements. The authors are grateful to Dr. E.E. Andronov, Head of the Laboratory of Microbiological Monitoring and Soil Bioremediation of the All-Russian Research Institute of Agricultural Microbiology for invaluable advice and assistance in performing molecular analysis.

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To cite this article: Kuzikova, I.L. et al., 2024. Biotransformation of butyltin compounds and microbiome diversity in the bottom sediments of the Gulf of Finland, the Baltic Sea. *Ecosystem Transformation* **7** (2), 126–143. https://doi.org/10.23859/estr-230109

Received: 09.01.2023
Accepted: 31.01.2023

DOI 10.23859/estr-230109
EDN HJOXYS
УДК 574.589+579.26

Научная статья

Биотрансформация соединений бутилолова и видовое разнообразие микробиомов в донных осадках Финского залива

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Аннотация. Загрязнение морской среды оловоорганическими соединениями представляет серьезную экологическую проблему ввиду их чрезвычайной токсичности для широкого круга организмов. В донных осадках Копорской губы Финского залива выявлен высокий уровень загрязнения соединениями бутилолова. Концентрация трибутилолова и дибутилолова составила 35.7 нг(Sn)/г а.с.о. и 9.7 нг(Sn)/г а.с.о. соответственно. Установлено, что трансформация соединений бутилолова в донных осадках происходит за счет автохтонной микробиоты осадков. Скорость убыли трибутилолова и дибутилолова составила 0.014 сут$^{-1}$ и 0.022 сут$^{-1}$, а периоды их полураспада 49.5 и 31.5 суток соответственно. Метагеномный анализ выявил изменения в таксономическом составе и уменьшение видового разнообразия микробиомов донных осадков в процессе трансформации оловоорганических соединений. В ходе биодеградации соединений бутилолова в донных осадках возрастает численность бактерий родов *Acidithiobacillus, Halothiobacillus*, *Alicyclobacillus*, *Sulfurospirillum*, *Denitrovibrio*, а также метаногенные археи рода *Methanolobus*, что может свидетельствовать об их потенциальном участии в биодеградации оловоорганических соединений. Полученные результаты могут быть использованы для выявления видов-активных деструкторов трибутилолова с целью разработки научно-обоснованных методов деконтаминации донных осадков, загрязненных оловоорганическими соединениями.

Ключевые слова: трибутилолово, дибутилолово, монобутилолово, микробное разнообразие

Финансирование. Работа выполнена в рамках государственного задания Министерства науки и высшего образования Российской Федерации (№ 122041100086-5).

Благодарности. Авторы выражают благодарность д.б.н. Е.Е. Андронову, заведующему лабораторией микробиологического мониторинга и биоремедиации почв ФГБНУ «Всероссийский научно-исследовательский институт сельскохозяйственной микробиологии» за консультации и помощь в проведении молекулярного анализа.

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Для цитирования: Кузикова, И.Л. и др., 2024. Биотрансформация соединений бутилолова и видовое разнообразие микробиомов в донных осадках Финского залива. *Трансформация экосистем* **7** (2), 126–143. https://doi.org/10.23859/estr-230109

Поступила в редакцию: 09.01.2023
Принята к печати: 31.01.2023

Introduction

Ecosystems polluted by anthropogenic toxic substances are characterized by low values of microbiome diversity indices and a high dominance index, which indicates impoverishment of their species composition and reduction of overall ecosystem stability (Kuzikova et al., 2022; Medvedeva et al., 2009; Wang et al., 2015).

Organotin compounds are among the common anthropogenic toxicants worldwide. They are used as agricultural pesticides, preservatives for wood treatment, and biocidal preparations. The most common are tributyltin (TBT) compounds, $(C_4H_9)_3Sn_X$, which were widely used as antifouling agents in marine paints in the 1950s–1980s (Du et al., 2014; Finnegan et al., 2018). Butyltin compounds (BTs) are hydrophobic, so they adsorb on suspended particles when once in water and then precipitate in bottom sediments. They are quite stable in sediments, their estimated half-life is about 87 ± 17 years (Viglino et al., 2004). Tributyltin is toxic to a wide range of organisms at all trophic levels, from bacteria to mammals. TBT disrupts the endocrine system by changing hormone content in the pituitary, thyroid and sex glands (Silva et al., 2014). The harmful effect of TBT on the endocrine system of aquatic organisms (e.g., mollusks and fish) is manifested at a concentration as low as 1.0 ng/L in water (or 10 ng/g body weight, when bioaccumulated), while the concentrations an order of magnitude higher cause the death of organisms (Lagadic et al., 2018). In addition, TBT has neuro- hepato-, nephro- and gastrotoxic effects, as well as that on the immune system (Gupta et al., 2011). The decomposition products of TBT, dibutyltin (DBT) and monobutyltin (MBT), also have toxic properties, but to a lesser extent and in the following order: MBT < DBT < TBT. Due to the BTs high overall toxicity to almost all range of living organisms, the International Maritime Organization has adopted the International Convention for the Control of Harmful Anti-Fouling Systems on Ships in 2001[1], proposing a global ban on TBT, which has become effective worldwide in 2008.

Despite the bans imposed, the problem of marine pollution by BTs still exists in many regions as evidenced by monitoring data (Filipkowska and Kowalewska, 2019; Rodríguez-Grimon et al., 2020; Uc-Peraza et al., 2022). For example, extensive studies of bottom sediments in the coastal zone of the southern Baltic Sea revealed the sites where the total concentration of BTs reached 3321 ng(Sn)/g DW, with 80% of the sediment samples studied being heavily contaminated with TBT (Filipkowska and Kowalewska, 2019).

In the bottom sediments, butyltin compounds degrade mainly through biodegradation. Microorganisms play a key role in transformation of these xenobiotics. Due to physiological and genetic features of microbiota representatives, they respond quickly to environmental changes and they are able to adapt quickly to new substrates. Microbiota involve resistance mechanisms to adapt to the

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[1] International Convention on the Control of Harmful Anti-Fouling Systems on Ships, 2001. IMO (International Maritime Organization). Web page. URL: https://www.imo.org/en/About/Conventions/Pages/International-Convention-on-the-Control-of-Harmful-Anti-fouling-Systems-on-Ships-(AFS).aspx (accessed: 23.01.2023).

action of pollutants (McAdams et al., 2004). It is known that some bacteria are TBT-resistant, for example, that of the genera *Escherichia*, *Pseudomonas*, *Proteus*, *Serratia*, *Alcaligenes*, *Aeromonas*, *Klebsiella*, *Enterobacter*, *Staphylococcus*, *Bacillus*, *Mycobacterium*, *Sphingobium*, *Stenotrophomonas*, *Rhizobium,* and some others (Cruz et al., 2007, 2014, 2015; Hassan et al., 2018), as well as some algae, for example, *Skeletonema costatum*, *Chlorella vulgaris*, *Scenedesmus obliquus*, *Dunaliella salina*, *Leptocylindrus danicus*, *Amphidinium carterae* (Reader and Pelletier, 1992; Tam et al, 2003; Xie et al., 2011) and fungi *Cunninghamella elegans*, *Cochliobolus lunatus*, *Metarhizium robertsii* (Bernat et al., 2013; Stolarek et al., 2019). Autochthonous bacteria play a significant role in the transformation of various xenobiotics, including TBT (Cruz et al., 2014; Kuzikova et al., 2022). Bacteria are the dominant group of microorganisms in bottom sediments, accounting for up to 90–95% of the total sediment biomass (Nealson, 1997). According to K. Cybulska et al. (2020), the abundance of mesophilic, psychrophilic and halophilic bacteria may reach up to 2.85×10^7, 4.11×10^7 and 5.60×10^7 CFU/g DW, respectively, in bottom sediments in the Baltic Sea (Cybulska et al., 2020). The studies of microbial communities of bottom sediments contaminated with butyltin compounds and associated changes in the species composition during the process of pollutant transformation are extremely scarce (Cruz et al., 2014; Suehiro et al., 2006). Applying molecular biology methods within such research significantly expands the knowledge about the diversity of microorganisms involved in the transformation of butyltin compounds. This is extremely important for the development of effective biotechnologies for the remediation of contaminated environmental objects.

In this regard, the present study aims (1) to describe the diversity of bottom sediment microbiota of the southern Gulf of Finland, Baltic Sea (Koporye Bay) during the BT transformation and (2) to determine the role of autochthonous microbiota in the dynamics of butyltin compounds in the bottom sediments contaminated with organotin compounds.

Materials and methods

Characteristics of the study area

The Koporye Bay has an area of 255 km² and locates in the southern part of the Gulf of Finland, the Baltic Sea. It receives a heavy anthropogenic load due to the use of large volumes of sea water in the cooling cycle of the operating Leningrad Nuclear Power Plant (LNPP). The heated LNPP water discharges to the eastern part of the Koporye Bay (area ~50 km; average depth 5 m). In addition, the anthropogenic pressure here is even more intense because of the discharge of river water contaminated by household wastewater (Sosnovy Bor town) into the bay, as well as by the operation of wastewater treatment facilities (Kryshev et al., 2021).

The Koporye Bay belongs to the small semi-enclosed water bodies with an extended watershed boundary with the main water area. Water salinity varies as 2–4‰. The bottom is mainly sandy, somewhere silty with random stones and gravel. The average depth is about 12 m. Maximum depths (down to 27 m) are recorded at the border with the main water area. Three rivers (Sista, Kovashi and Voronka) flow into the eastern part of the bay with an average discharge of about 10 m³/s.

Sediment sampling and setting up an experiment

Bottom sediment samples (0–5-cm layer) were collected in the Koporye Bay area in June 2018, N 59.9916° E 28.99839°. Dark brown silt loam (0.5-cm thick) with no sand admixture was the surface sediment layer; below, there was dark greenish-gray clayey sand (0.5-cm thick), followed by dense clayey sand (4–5-cm thick). The sediment pH was 7.57, temperature, 6.5 °C.

Bottom sediments were sampled with a Peterson bottom grab, placed in the sterile glass containers, and transported to the laboratory on the ice (cooled box). The samples were stored in a refrigerator at 4.0 °C. The bottom sediment microcosms were set in the sterile 1000-mL vials containing 200 g of sediment (DW). Control microcosms of sterile sediments were prepared at the same time. For this purpose, the sediment sample was autoclaved at 120 °C (twice for 40 min each). Microcosms of sterile and non-sterile sediments were prepared in three repetitions. Vials with sediments were covered with lids and incubated at room temperature under constant dark conditions for 240 days. After 0, 90, 180 and 240 days, the samples were thoroughly mixed and aseptically collected for chemical and molecular analysis.

Analysis of butyltin compounds in sediments

Butyltin compounds in bottom sediments were analyzed using the ISO 23161:2018 method for the quantification of volatile organotin derivatives in sediments and soil[2] using a QP-2010 gas chromatograph/mass spectrometer (Shimadzu, Japan) in SIM (selective ion monitoring) target mode. The procedure of sample preparation and gas chromatography–mass spectrometry (GC-MS) analysis conditions have been described in detail earlier (Kuzikova et al., 2022).

A first-order kinetic model was used to evaluate the removal efficiency of BTs:

$$\ln(C_{in}/C_t) = kt,$$

where C_t – BTs' concentration at time *t*, ng/g; C_{in} – initial BTs' concentration, ng/g; t – degradation period, day; k – degradation rate constant, day^{-1}.

The BTs half-life ($t_{1/2}$) was calculated according to the equation $t_{1/2} = 0.693/k$ (Rajendran et al., 2017).

Molecular analysis and diversity assessing

The taxonomic composition of the microbiome of bottom sediment microcosms was studied using high-throughput sequencing (Illumina MiSeq, USA). DNA isolation and sequencing of 16S rRNA gene amplicon libraries were performed according to previously described procedures (Ivanova et al., 2020). DNA was isolated from 0.5-g sediment sample using the Power Soil kit (Mobio Laboratories, USA) according to the manufacturer's protocol. The purified DNA was used as a matrix in a PCR with universal primers added for the variable B4 region of the 16S rRNA gene F515 (5′-GTG CCA GCMGCC GCG GTAA-3′) and R806 (5′-GGA CTA CTA CBSGGG TAT CTAAT-3′), along with the oligonucleotide identifiers for each sample and service sequences required for pyrosequencing (Bates et al. 2010).

The applied primers were designed in accordance with the analysis of nucleotide sequences of both bacteria and archaea, allowing amplification of an approximately 300 bp 16S rRNA gene fragment. Sample preparation and sequencing were performed according to Illumina manual. Sequencing of amplicon libraries was performed at the Genomic Technologies, Proteomics and Cell Biology Center (All-Russia Research Institute for Agricultural Microbiology, Russia).

Data were processed in the QIIME 1.8.0 program (Caporaso et al., 2010). Sequences shorter than 200 nucleotides or having a quality score below 25 were excluded from analysis. Chimeric sequences were excluded from amplicon libraries. Amplicon libraries were normalized by the number of sequences of the smallest size.

Sequences characterized by ≥97% similarity were combined into operational taxonomic units (OTUs) using a *de novo* algorithm (based on the *uclust* method). The total diversity of prokaryotic communities (α-diversity) of sediments was estimated by the number of detected taxa (OTUs), Shannon index, Simpson index, and Chao1 index, which estimates theoretically possible species number (Chirak et al., 2013). Bray-Curtis similarity index and principal coordinate analysis (PCoA) were applied to calculate β-diversity.

Statistical analysis

The data were processed statistically in Past 4.0 software. Statistical significance of differences was assessed using one-way ANOVA and Tukey's post-hoc test for data with normal distribution, otherwise using Mann–Whitney U-test; the differences were considered significant at $p < 0.05$. The data are presented as mean ± standard deviation ($m \pm SD$) for three independent replicates of each treatment with triplicated repetitions for each.

Results and discussion

Organotin compounds

According to GC-MS analysis, tributyltin (35.7 ng(Sn)/g DW) and dibutyltin (9.7 ng(Sn)/g DW) were found in the bottom sediments of the Koporye Bay of the Gulf of Finland, the content of monobutyltin was below the detection level (Fig. 1).

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[2] ISO 23161:2018. Soil quality. Determination of selected organotin compounds. Gas-chromatographic method.

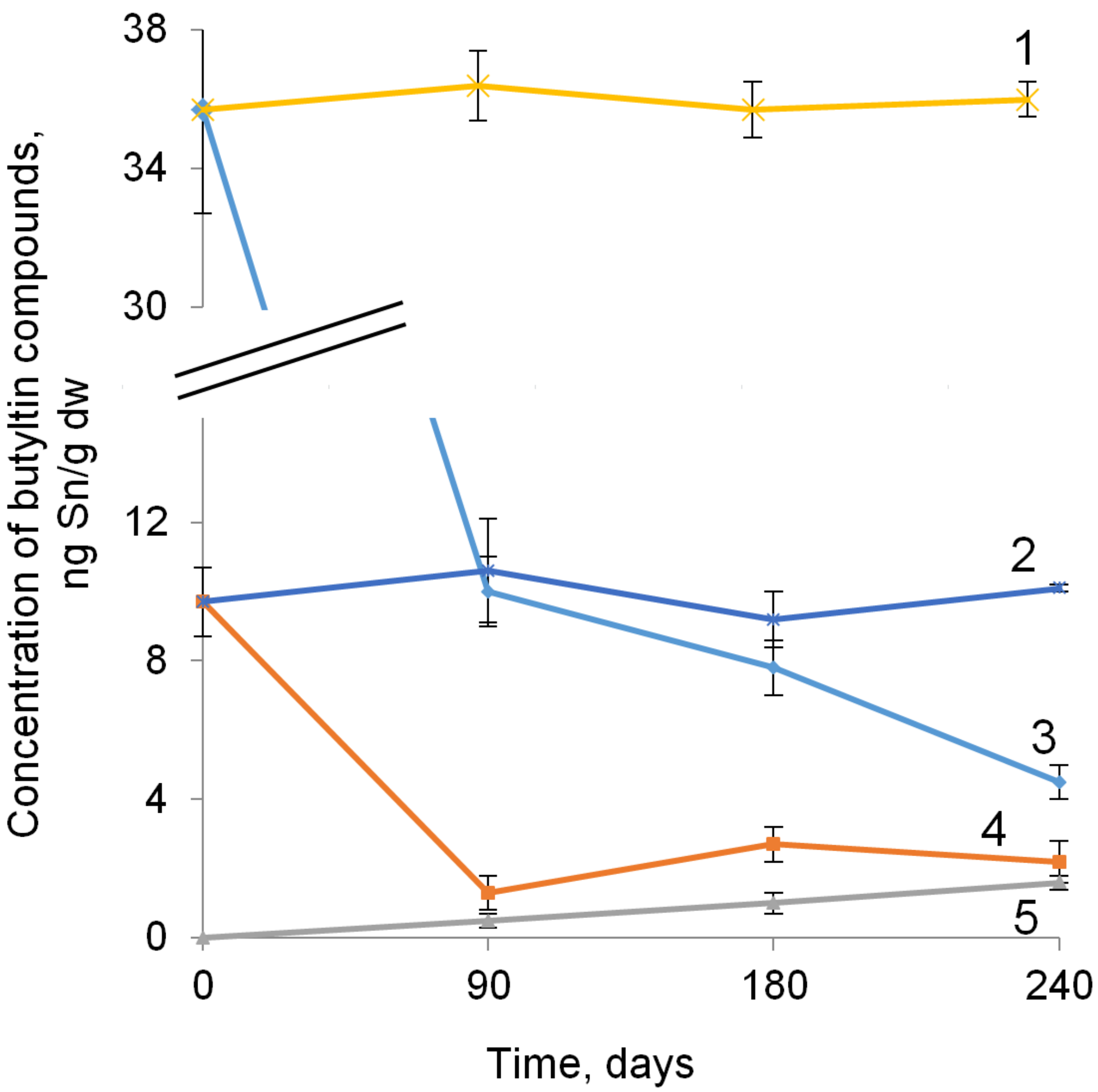

Fig. 1. Content of butyltin compounds in sediment microcosms: 1 – TBT (sterile microcosm); 2 – DBT (sterile microcosm); 3 – TBT (non-sterile microcosm); 4 – DBT (non-sterile microcosm); 5 – MBT (non-sterile microcosm).

These contents of BTs are high in accordance to the Norwegian Environmental Quality Classification System for pollutants in bottom sediments (Bakke et al., 2010), to the classification proposed by P.H. Dowson et al. (1993), and to the HELCOM recommendations (2010), so the studied bottom sediments are highly contaminated. Similar levels of BTs have been previously recorded in the sediments from the Gulf of Gdansk (the Baltic Sea) with total BTs' concentration ranging as 5.7–3321 ng(Sn)/g DW, where tributyltin comprised 80% (Filipkowska and Kowalewska, 2019). A few years earlier, the maximum concentration of TBT in these sediments was even higher and reached 15780 ng(Sn)/g DW (Filipkowska et al., 2011, 2014).

In order to reveal the nature of BTs transformation in bottom sediments (abiotic or biological), the BTs' content was determined in both sterile and non-sterile sediments during the experiment. In sterile sediment microcosms, no loss of TBT and DBT was detected throughout the entire incubation period of 240 days (Fig. 1).

In contrast to sterile microcosms, a statistically significant loss of TBT and DBT ($p < 0.05$) was observed in non-sterile sediments (Fig. 1). In 90 days, TBT content has decreased more than threefold from 35.7 ng(Sn)/g DW down to 10.0 ng(Sn)/g DW, DBT, even stronger, from 9.7 ng(Sn)/g DW down to 1.3 ng(Sn)/g DW. At the end of the incubation period (240 days), a further TBT decrease down to 4.5 ng(Sn)/g DW has been registered, but DBT content remains stable when compared to day 90 ($p < 0.05$). The calculated loss rate constant (K) of TBT and DBT were 0.014 day^{-1} and 0.022 day^{-1}, the half-life ($t_{1/2}$), 49.5 and 31.5 days, respectively. However, when MBT (the least toxic BT) was not detected in the initial samples, its content increased during the incubation period and reached 1.6 ng(Sn)/g DW in 240 days.

According to the published data, the loss dynamics of BTs in bottom sediments and nature of changes of these compounds depend on many factors. Primarily, these are the contamination degree, sedimentation rate, sediment enrichment with organic matter, and microbial activity. For example, there was practically no loss of TBT, DBT and MBT after 150 days of incubation of bottom sediments sampled in the Ria de Aveiro estuarine system (Portugal), contaminated with BTs at a level similar to that in the present study (TBT, 25 ng(Sn)/g; DBT, 7.1 ng(Sn)/g; MBT, 13 ng(Sn)/g) (Cruz et al., 2014). At the same time, the half-life of TBT (1.2–1.3 µg/g DW) in the bottom sediments of the Mekong River (Vietnam), exceeding that of the Ria de Aveiro estuarine system in 50 times, was about 150 days (Suehiro et al., 2006).

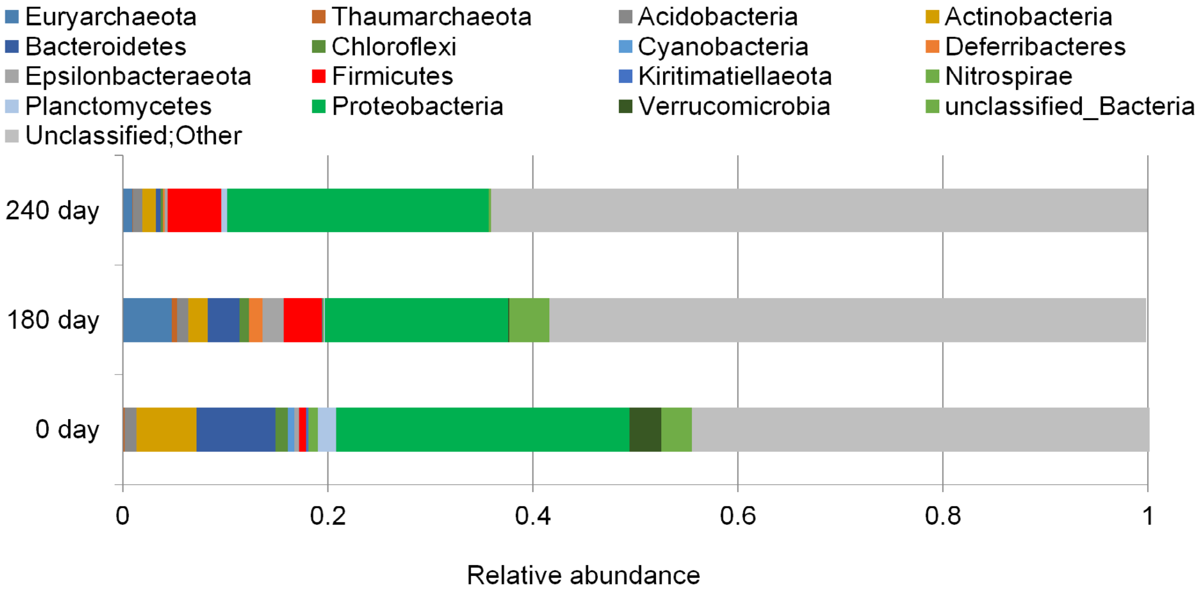

Fig. 2. Taxonomic structure (phylum level) of prokaryotic communities of bottom sediment microcosms during incubation.

Taxonomic structure of prokaryotic communities

The taxonomic composition of the microbial community changed during the transformation of BTs in bottom sediments. According to metagenomic analysis, dominant prokaryotic phyla (> 1%) were Proteobacteria (28.6%), Bacteroidetes (7.7%), Actinobacteria (5.9%), Verrucomicrobia (3.1%), Planctomycetes (1.8%), Chloroflexi (1.2%). Minor phyla (< 1%) were represented by Acidobacteria (0.9%), Nitrospirae (0.9%), Cyanobacteria (0.6%), Firmicutes (0.7%), Epsilonbacteraeota (0.5%), and Kiritimatiellaeota (0.2%); the share of Thaumarchaeota (Archaea) did not exceed 0.2%. The share of unclassified prokaryotes was 3%. Nucleotide sequences not attributed to the domain level accounted for 44.7% due to the incompleteness of the available databases, which was typical of metagenomic studies (Fig. 2).

According to ANOVA, there were significant changes in the quantitative ratios of almost all phyla in the sediment microcosms contaminated with BTs during entire incubation period (240 days). A significant decrease in the relative abundance of bacteria at day 180 and day 240 compared to their initial content was noted practically for all dominant phyla, while the share of some minor phyla (Firmicutes, Epsilonbacteraeota, and Deferribacteres) has increased (Fig. 2). The abundance of dominant phylum Proteobacteria has decreased during sediment incubation due to a decrease of that of Alphaproteobacteria and Deltaproteobacteria (classes within this phylum). On the contrary, the share of Gammaproteobacteria has increased by 1.3 times (Fig. 3). At the same time, although Gammaproteobacteria belonging to order Betaproteobacteria prevailed in the initial sediment microcosm, the increase in the abundance of representatives belonging to the orders Acidithiobacillales and Hallotiobacillales (genera *Acidithiobacillus* and *Halothiobacillus*) was observed after 180 and 240 days of incubation.

After 240 days of incubation, bacteria of the genus *Acidithiobacillus* have increased their abundance and so became dominants in sediment microcosms, their share has changed from 0.1% up to 15.2%. The abundance of bacteria of the genus *Halothiobacillus* increased by an order of magnitude, from 0.1% to 1.2% (Table 1).

The ability of representatives of these prokaryotic genera to transform BTs is currently unknown. However, some species of genus *Halothiobacillus* are able to degrade toxic azo dyes in contaminated river sediments (Ito et al., 2016), as well as to utilize thiocyanates, which are a part of waste from coke and precious metal plants (Sorokin et al., 2014). In addition, acidophilic iron-oxidizing *Halothiobacillus* bacteria are the dominant group in the microbial communities of acidic drainage waters of abandoned tin mines (Hallberg and Johnson, 2005). Acidophilic chemolithoautotrophic bacteria of the genus *Acidithiobacillus* are usually prevalent in mining process waters and in acid mine drainage waters (Chen et al., 2022a). *Acidithiobacillus ferrooxidans* and *A. thiooxidans* have been successfully used in the biomining of metals (tin, copper, nickel, zinc, uranium, etc.) and bioremediation of metal-contaminated environmental sites (Chen et al., 2022a; Willner et al., 2022; Zhang et al., 2018).

The observed increase in the abundance of bacteria belonging to the class Gammaproteobacteria in the process of TBT transformation may be associated with their high resistance to toxic effects of BTs. Many representatives of Gammaproteobacteria (*Aeromonas molluscorum*, *Klebsiella pneumonia*, *Moraxella osloensis*, *Pseudomonas* sp., *Shewanella putrefaciens*, and *Stenotrophomonas chelatiphaga*) are known as efficient degraders of butyltin compounds and are highly tolerant to toxicants (Cruz et al., 2014; Hassan, 2018; Khanolkar et al.,2015; Lee et al., 2012; Yáñez et al., 2015).

The increase in the relative abundance of representatives of the phylum Firmicutes by 5.3 and 7.4 times in sediment microcosms after 180 and 240 days during the process of BTs transformation was due to the predominance of bacteria represented by genera *Clostridium sensu stricto* and *Alicyclobacillus,* belonging to the families Clostridiaceae and Alicyclobacillaceae, class Bacilli (Table 1). Previously, resistance to organotin compounds was reported for *Staphylococcus xylosus*, *Kurthia zopfii*, *Listeria grayi*, *Bacillus cereus*, and *Bacillus thuringiensis*, belonging to the same class (Cruz et al., 2007).

Almost 1.5-fold increase in the abundance of representatives of the phylum Epsilonbacteraeota was due to the species belonging to the genera *Sulfurospirillum* and *Sulfurimonas* (families Sulfurospirillaceae and Thiovulaceae, order Campylobacterales); however, this increased was observed only for the period from day 0 to day 180. The share of genus *Denitrovibrio* (phylum Deferribacteres) significantly increased also by 180 days (Table 1). So far, the role of these bacteria in unknown in the biotransformation of BTs.

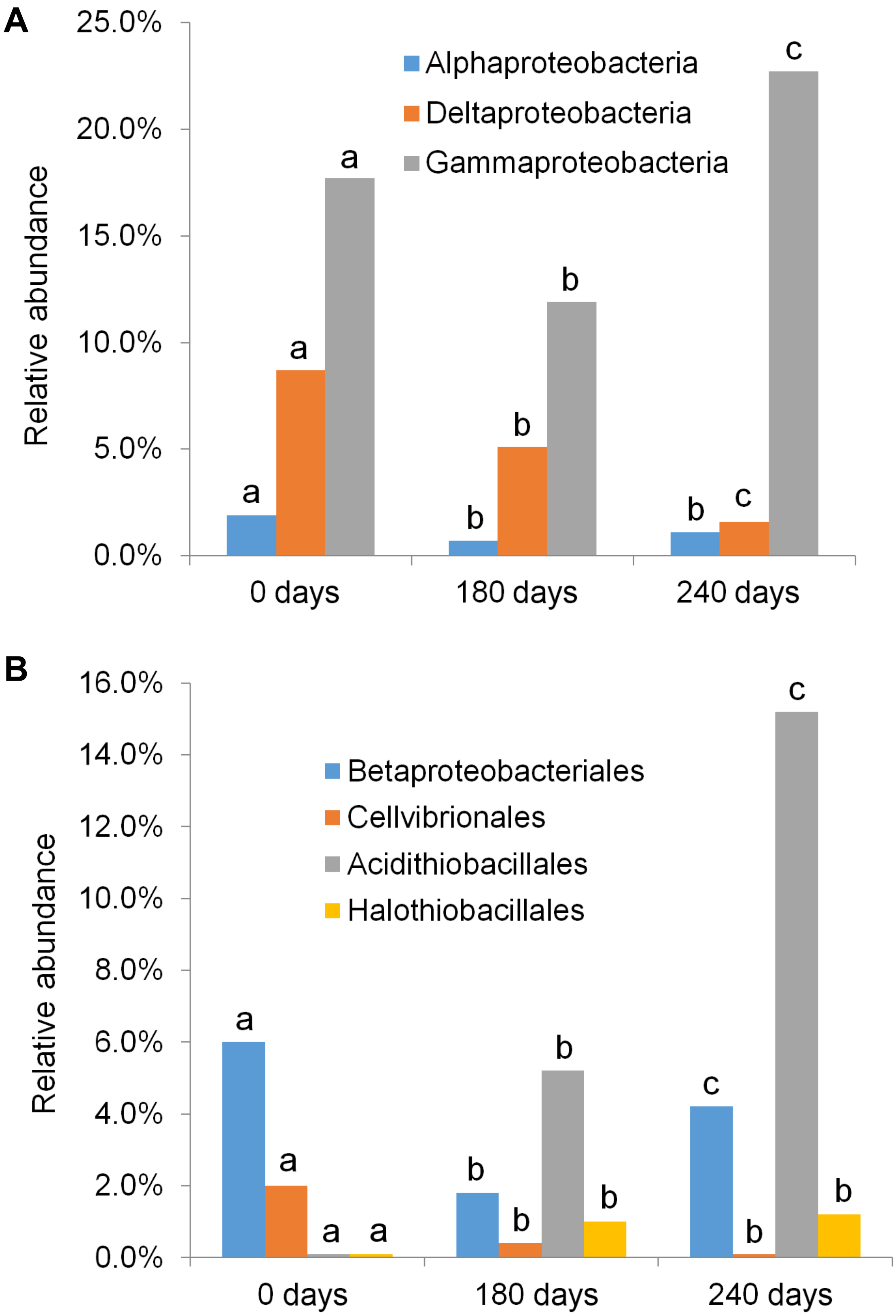

Fig. 3. Relative abundance (%) of 16S rRNA gene nucleotide sequences: **A** – taxonomic class level, dominant phylum Proteobacteria; **B** – taxonomic order level, class Gammaproteobacteria. Different letter indices within the same group indicate significant differences between microcosm samples (*p* < 0.05).

Table 1. Relative number of nucleotide sequences of the 16S rRNA gene of bacteria at the genus level in sediment microcosms. Different letter indices within the same genus indicate significant differences between microcosm samples ($p < 0.05$).

Genus	Relative abundance, %		
	Incubation period		
	0 days	180 days	240 days
Clostridium sensu stricto	0.1 ± 0.02^{a*}	0.6 ± 0.1^{b}	0.6 ± 0.2^{b}
Alicyclobacillus	0.1 ± 0.01^{a}	0.9 ± 0.2^{b}	2.4 ± 0.2^{c}
Sulfurospirillum	0.1 ± 0.03^{a}	1.0 ± 0.3^{b}	0.1 ± 0.05^{a}
Sulfurimonas	0.1 ± 0.02^{a}	0.7 ± 0.1^{b}	0.1 ± 0.02^{a}
Denitrovibrio	0.1 ± 0.02^{a}	1.3 ± 0.3^{b}	0.2 ± 0.1^{a}
Acidithiobacillus	0.1 ± 0.03^{a}	5.2 ± 0.4^{b}	15.2 ± 1.2^{c}
Halothiobacillus	0.1 ± 0.01^{a}	1.0 ± 0.2^{b}	1.2 ± 0.2^{b}
Methanolobus	0.1 ± 0.02^{a}	1.6 ± 0.3^{b}	0.1 ± 0.02^{a}
Geothrix	0.1 ± 0.04^{a}	0.7 ± 0.1^{b}	0.1 ± 0.01^{a}
Dyella	0.1 ± 0.03^{a}	0.6 ± 0.1^{b}	0.1 ± 0.03^{a}

Table 2. Indices of richness and diversity of sediment microcosms. Letter indices indicate significant differences between microcosm samples ($p < 0.05$).

	Index			
Incubation period	OTU	Shannon	Simpson	Chao1
0 days	737 ± 69^{a}	5.808 ± 0.121^{a}	0.994 ± 0.003^{a}	739.4 ± 63.1^{a}
180 days	554 ± 41^{b}	3.683 ± 0.105^{b}	0.833 ± 0.002^{b}	557.6 ± 40.3^{b}
240 days	355 ± 29^{c}	3.057 ± 0.087^{c}	0.848 ± 0.002^{c}	356.9 ± 28.7^{c}

It should be noted that the abundance of methanogenic archaea of the genus *Methanolobus* (phylum Euryarchaeota) increased greatly (from the initial 0.1% at day 0 to 1.6% at day 180) in the sediment microcosms during the process of TBT degrading, in addition to the observed changes in the taxonomic structure of the prokaryotic community (Table 1). Methanogenic archaea play an important role in wastewater treatment, often exhibiting resistance to a wide range of antibiotics with ability to degrade them (Chen et al., 2022b; Ng et al., 2015).

Microbial diversity in sediment microcosms

Diversity of bottom sediment microbiota is an important factor of their biological stability, as well as the intensity and direction of many biochemical processes. The diversity indices calculated in the present study evidenced that species diversity of prokaryotic sediment microbiomes significantly decreased during TBT transformation. After 240 days of sediment incubation, the number of detected taxa (OTU) and the estimated number of species (Chao1) decreased more than a half. According to Shannon index, the number of taxa decreased by 1.6 times after 180 days and by 1.3 times after 240 days of experiment. The Simpson index, allowing to assess the community diversity, also decreased significantly (Table 2). When assessing β-diversity on the basis of PCoA cluster analysis using the Bray–Curtis index, the taxonomic composition of sediment microbiomes has a rather low degree of similarity with control during TBT transformation after 180 and 240 days of incubation, when microbiomes formed non-overlapping clusters (Fig. 4). This indicate a decrease in species diversity in the bottom sediment microbiomes during transformation of butyltin compounds.

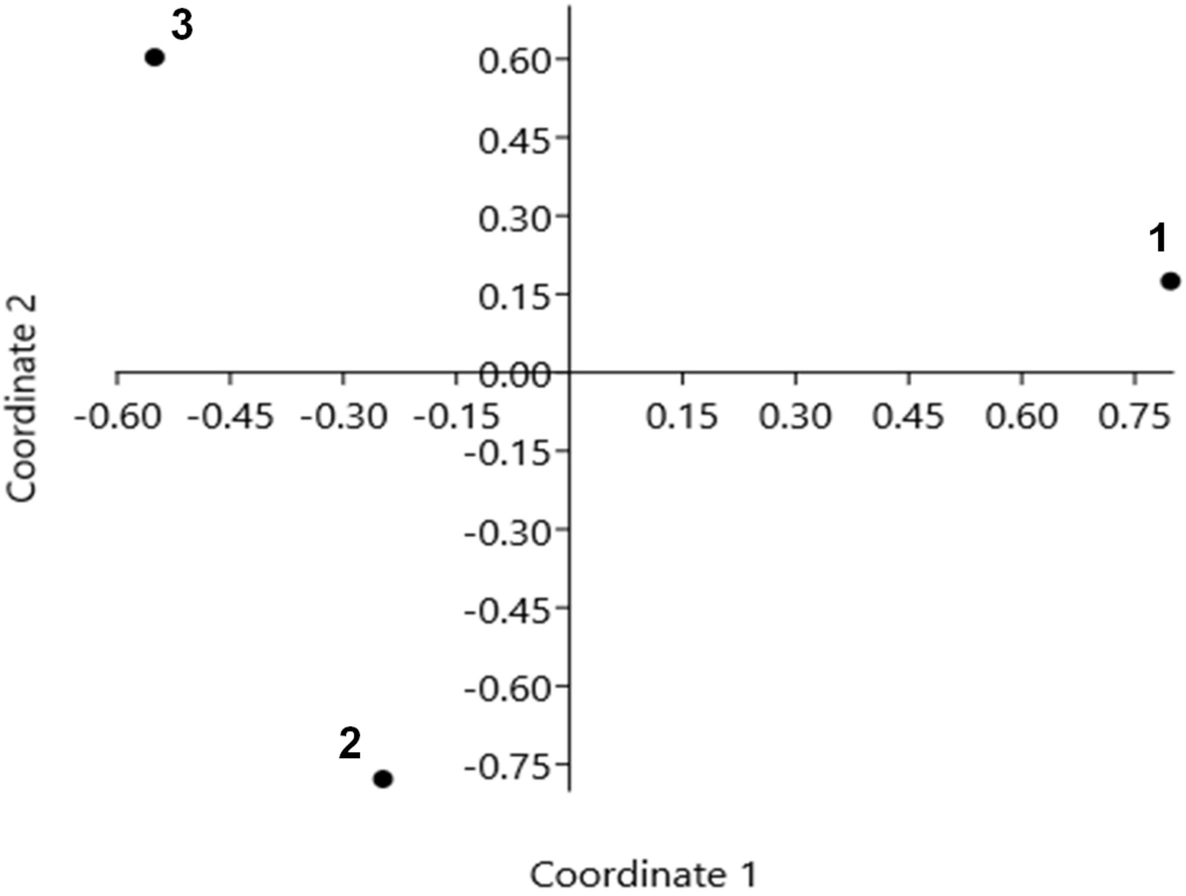

Fig. 4. Principal coordinates analysis (PCA) of sediment microcosms (based on the Bray-Curtis index). 1 – starting sediment at the day 0; 2 – sediment microcosm after 180 days of incubation; 3 – sediment microcosm after 240 days of incubation.

Conclusions

Under the controlled experimental conditions, the transformation of butyltin compounds was studied in bottom sediments of the Koporye Bay of the Gulf of Finland, as well as their effect on the taxonomic structure and species diversity of sediment microbiomes. In the samples heavily contaminated with dibutyltin and tributyltin, a loss of these toxicants is observed due to their biodegradation by autochthonous sediment microbiota. The increasing content of monobutyltin (MBT) in sediments indicates the process of biotransformation of tri- and dibutytin into a less toxic compound, MBT. As butyltin compounds biodegrade, significant changes in the taxonomic composition and species diversity of sediment microbiomes are detected. The relative abundance of bacteria of the dominant phyla *Proteobacteria*, *Bacteroidetes*, *Actinobacteria*, *Berrucomicrobia*, *Planctomycetes*, and *Chloroflexi* decreases, while the share of the minor phyla Firmicutes, Epsilonbacteraeota, and Deferribacteres increases. Bacteria of the genera *Acidithiobacillus* and *Halothiobacillus* predominate throughout the study period; these prokaryotes belong to the class Gammaproteobacteria, which representatives are known as active destructors of organotin compounds. In addition, the abundance of bacteria of the genera *Alicyclobacillus*, *Sulfurospirillum*, *Denitrovibrio* and methanogenic archaea of the genus *Methanolobus* increase also during the experiment. The dominance of these taxa may indicate their participation in the butyltin compounds' transformation. According to the number of detected OTUs, as well as Shannon, Simpson and Chao1 index values, the species diversity of microbiomes decreases during the biodegradation of butyltin compounds. The observed changes in the taxonomic structure of the bottom sediment microbiome may serve as a bioindicator of the ecological state of the marine environment. The results of this study may be used to identify bacteria with the ability to degrade toxic butyltin compounds in order to develop relevant methods for bioremediation of bottom sediments contaminated with organotin compounds.

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