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RESEARCH ARTICLE

Distribution of tributyltin in surface sediments from transitional marine-lagoon system of the south-eastern Baltic Sea, Lithuania

Sergej Suzdalev · Saulius Gulbinskas · Nerijus Blažauskas

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Abstract The current research paper presents the results of contamination by tributyltin (TBT) compounds in Klaipėda Port, which is situated in a unique marine-lagoon water interaction zone. One hundred fifty-four surface sediment samples have been taken along the whole transition path from lagoon to the sea and analysed in order to quantify the contamination rate in specific environment of high anthropogenic pressure. The detected TBT concentrations ranged from 1 to 5,200 ng Sn g^{-1} of dry weight of sediment. The back-trace of horizontal distribution of TBT-contaminated sediments show obvious increase of tributyltin concentrations closer to port areas dealing with ship repair and places of dry-docking facilities. This is a clear indication that those activities are the main source of contamination in the study area. The estimated correlation of TBT concentration in sediments with total organic carbon and the amount of fine fraction (<0.063 mm) was significant for most of the stations. The TBT concentration in those sites varies from 1 to 100 ng Sn g^{-1}. This fact indicates that the most intensive accumulation of tributyltin is related to potential contamination source areas (ship repairing, dockyards) due to direct input of hazardous substances into the water.

Keywords Tributyltin compounds · Marine-lagoon environment · South-eastern Baltic · Contamination · Port

Introduction

Organotin compounds are described as the most toxic substances that have been deliberately released into the marine environment during the last 60 years (Hoch 2001). So far, attention has mainly been given to the pollution of water and sediments with tributyltin (TBT). Even very low concentration of this hazardous substance has high toxic impact to aquatic life (Chagot et al. 1990). TBT has been applied as an effective biocide additive since 1970s. TBT rich antifouling paint was widely used to coat hulls of pleasure boats, large ships and the docks (Fent 1996). Marine environment is being polluted by TBT through direct leaching from antifouling paints into the water. Pollutants are being adsorpt by suspended particulate matter and as a consequence of further sedimentation are fixed in the bottom sediments (Díez et al. 2006).

Due to the toxic, persistent, bioaccumulative and endocrine disruptive impact to the natural environment caused by TBT contamination, the topic is addressed in many international conventions, e.g. Convention on the Protection of the Marine Environment of the Baltic Sea Area (Helsinki Convention), Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) and the Convention on the Protection of the Black Sea Against Pollution (BSC). As a result, European Union has included TBT compounds into the priority list of compounds to be specifically monitored in water in order to control its presence and fate in the natural systems. Additionally, in 1999, the International Maritime Organization (IMO) has adopted the resolution banning the use of TBT in new antifouling systems starting from the 1 January 2003. The resolution also required to take environmentally sound actions to render harmless or remove the old TBT containing systems until 1 January 2008. Finally, in 2008, the use of organotin compounds in antifouling paints was completely banned by the International Convention on the Control of Harmful Antifouling Systems on Ships of the International Maritime Organization (IMO 2001). It was stated that ships cannot bear organotin compounds on their hulls or external parts or surfaces unless there is a coating that forms a barrier so that organotin compounds cannot leach out.

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S. Suzdalev (✉) · S. Gulbinskas · N. Blažauskas
Marine Science and Technology Centre, Klaipėda University, H. Manto str. 84, 92294 Klaipėda, Lithuania
e-mail: suzdalev@corpi.ku.lt

Despite the existing legislative restrictions, environmental pollution caused by highly toxic TBT compounds remains to be a severe problem in many countries (Garg et al. 2011; Kim et al. 2011). Recent studies have shown that most contaminated are semi-enclosed basins (of poor water exchange capabilities) where shipping or ship-related activities are intense or fish farming is being introduced (Batley 1996). Similarly, high TBT concentrations in bottom sediments were reported for ports (Bhosle et al. 2006; Cassi et al. 2008; Díez et al. 2006; Harino et al. 2007) and estuaries (Garg et al. 2010; Rodriguez et al. 2010) all over the world.

Shallow water zones of the south-eastern Baltic Sea including semi-closed bays (lagoons) are zones of high bio-productivity, recreation potential, or perspective for the mass development of tourism (Falandysz et al. 2002, 2006; Filipkowska et al. 2011; Radke et al. 2008). The Klaipėda Strait is a zone of natural geochemical barrier (Emelyanov 1998), has permanent water circulation (Stakėnienė et al. 2011) and an environment of intensive sedimentation (Trimonis et al. 2010). The anthropogenic pressure on the natural environment at the Klaipėda Strait is determined by the activities of the multipurpose, universal, deep-water port of Klaipėda. Port accommodates 15 big stevedoring companies, ship repair and shipbuilding yards, which provide cargo-handling and recreational services. The annual port cargo-handling capacity is up to 40 million tons in average. Therefore, the potential risk of TBT (as well as other harmful substances) pollution in the Klaipėda Strait is quite high.

The amount of scientific studies addressing the TBT contamination topic in the Baltic Sea sediments is quite limited (Biselli et al. 2000; Eklund et al. 2010). Even though there is some information on TBT values in Klaipėda Strait available (Dudutytė et al. 2007), extensive research on tributyltin distribution in this transitional marine-lagoon system (south-eastern part of the Baltic Sea) has never been carried out. TBT concentrations were measured only at few monitoring stations, thus cannot provide with a full picture of contamination extent in the area.

The present paper presents the results of quantitative analysis and distribution of TBT compounds in surface sediments from Klaipėda Port, situated in transitional marine-lagoon system.

Materials and methods

Study area

Klaipėda Strait is the only inlet connecting the freshwater Curonian Lagoon with the Baltic Sea. The area of the strait is 6.23 km^2. The strait is 12 km in length, while its width varies from 385 m in the north, at the entrance into the Baltic Sea to almost 1.5 km in the southern part, where the strait graduates into the Curonian Lagoon (Fig. 1).

Klaipėda Strait is often characterized as a very specific and dynamic area. Strait functions as natural geochemical barrier. The migration and transformations of the chemical elements are rather intensive. This results in the formation of sediments with anomalously high concentrations of chemical elements (Emelyanov 1998). Natural processes are often disturbed by morphology changes of the strait bottom due to the capital and maintenance dredging of the port area.

Two different sedimentation zones have been distinguished—natural and technogenic. The separation was based on the differences of water circulation and sedimentary conditions as well as the chemical composition of main lithological types of the sediments (Galkus and Jokšas 1997; Trimonis and Gulbinskas 2000). The zone of natural sedimentation extends along the western part of the strait and continues in the navigational channel of the port (central part of the strait). Water depth in this particular zone varies from 7.5 to 15 m, width ranges from 150 to 600 m. Various-grained sand with variable amount of silt are the typical sediments of this zone. Amount of the silt in the sand depends on the changes in hydrometeorological conditions and intensity of capital dredging works. More intensive accumulation of fine dispersed material is being observed in the area during the last years (Trimonis and Gulbinskas 2000). This is related to the intense deepening works being carried out in the port.

The remaining eastern part of the strait is the zone of technogenic sedimentation. It adjoins the quays and embankments of stevedoring, ship repairing companies and shipbuilding yards operating in the Klaipėda Port. The biggest (1.6-km long and 300–500-m wide) closed backwater area in Klaipėda Strait—Malku Bay—is also part of this particular zone of very slow water circulation. The bay embraces the mouth of small Smeltale River in the north-east and Wilhelm channel in the south. Moreover, southern part of the bay is occupied by floating docks, operating by a modern ship repair company. Another dockyard is located in the central part of the strait in the vicinity of established Yacht Port (Fig. 1). Accumulation of silty and silty/pelitic mud dominates in this zone. The lithologic composition of the sediments does not change considerably over the year.

Samples collection and analyses

The distribution of the bottom sediment samples (first 5 cm of the surface layer) collected in 2010–2012 cover the whole area of the strait. Position and the amount of the sampling stations were chosen in order to represent different hydrodynamic and morphological aspects of the Klaipėda Strait, to be able to compare the situation in natural environment against the zones impacted by different anthropogenic activities (Fig. 1). The sampling was carried out using the sediment grab of Van Veen

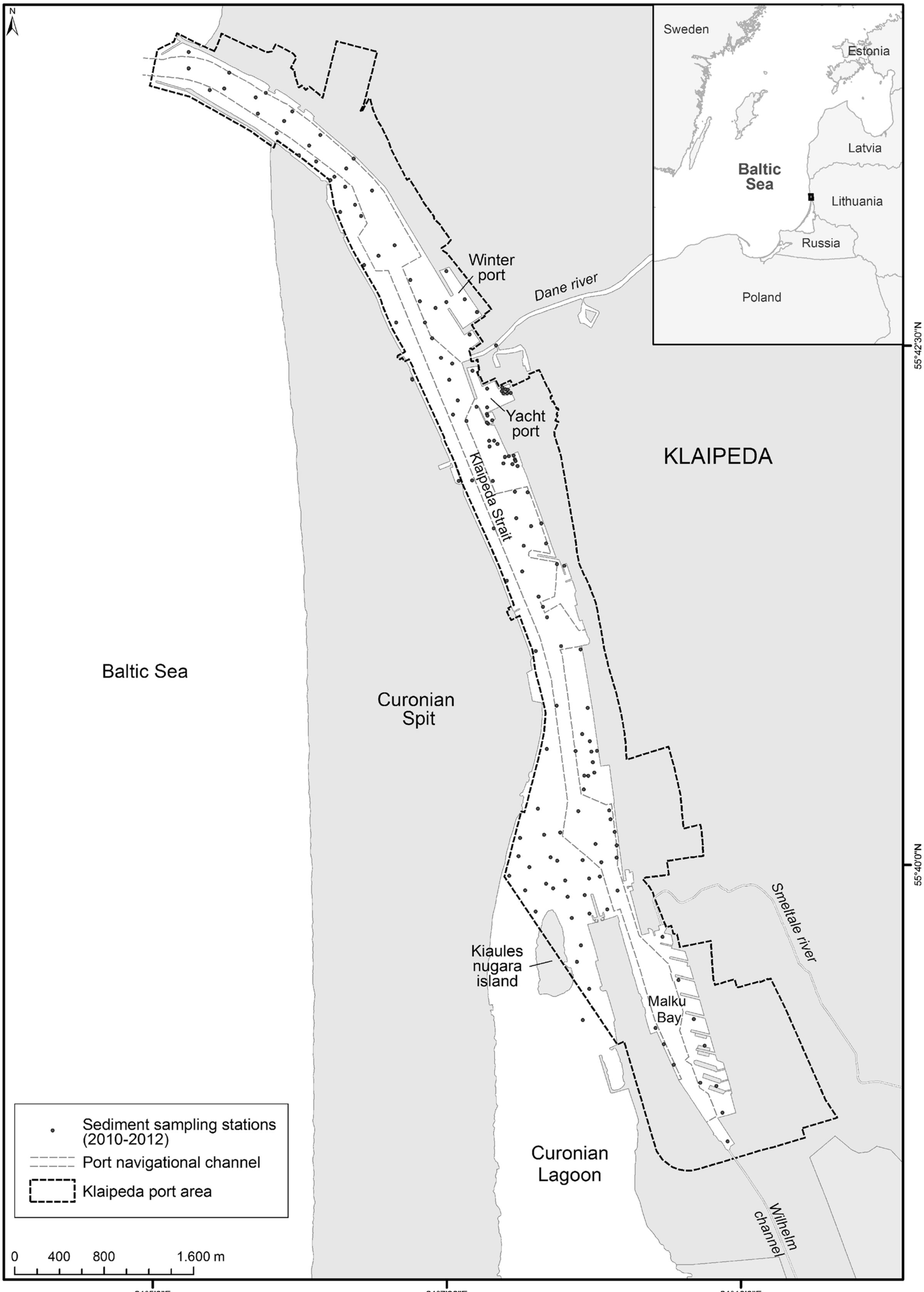

Fig. 1 Study area and sediment sampling stations

type. One hundred fifty-four samples have been collected, frozen and stored at−20 °C after the collection prior to being analysed for total TBT content and grain-size distribution. Sixty-nine samples were additionally analysed for the content of total organic carbon (TOC).

Laser diffraction method using laser particle analyser Analysette 22 Micro Tec Plus, Fritsch was used in order to perform grain-size analysis. The lithological classification of the sediments was done in accordance with the soil classification set in the Lithuanian normative document for dredging and dumping (LAND 46A-2002 2002). The following types of sediments have been distinguished: silty mud (<0.063 mm fractions >70 %), sandy mud (>0.063 mm fractions 30–50 %, <0.063 mm fractions 50–70 %), silty sand (<0.063 mm fractions 10–50 %) and sand (>0.063 mm fractions >50 %).

The content of TOC in sediments was determined by dry combustion with a nondispersive, infrared TOC analyser Shimadzu TOC-V CPH.

Gas chromatographic method was applied to analyse the content of TBT in the sediments. Analysis was done in accordance with the International Standard ISO 23161:2009 (soil quality—determination of selected organotin compounds—gas chromatographic method). The process of the analysis included the following: sample homogenization, TBT extraction using methanol, centrifuging of the extract and transfer to the separation funnel for derivatization. Derivatization was done in acetate buffer (pH 4.5) with sodium tetraethylborate used as derivatizing agent. Mass spectrometric detector (MSD) was used in order to determine the organotin cations. The accuracy of the analysis (detection limits) was 1 ng Sn g^{-1} d.w. Deuterated TBT-d_{27} was used as internal standard. Matrix spikes and blank samples were analysed in every sample patch in order to monitor the extraction efficiency and possible contaminations. BCR 646 certified reference material was used as quality control sample. TBT analyses were performed at the Ramboll Laboratory in Lahti, Finland (accreditation no. FINAS T039).

Statistical analysis

Spatial Analyst tool embedded in the ESRI ArcGIS software was used for linear interpolation of TBT concentrations in Klaipėda Strait. The relationships between the TBT content in the sediments and TOC content as well as amount of fine fractions (<0.063 mm) were evaluated after performed correlation analysis. Shapiro-Wilk test was applied in order to evaluate the possibility to use parametric testing. Results of the test showed that the conditions necessary for using the R-Pearson parametric linear correlation were not fulfilled (measured parameters cannot be adequately modelled by a normal distribution with 95 % confidence, $p<0.05$). Therefore, non-parametric R-Spearman correlation was applied. This method is less sensitive to extreme values (outliers) of the dataset. A correlation with $p<0.05$ was regarded as significant.

Results and discussion

Sediment characteristics

According to the earlier studies, the lithological composition of the bottom sediments in the Klaipėda Strait varied significantly—from boulders and pebbles to fine silty mud (Gulbinskas and Trimonis 1999). Recently, the distribution and composition of bottom sediments has changed significantly. The changes in the sedimentary pattern at the strait were due to intensive dredging (including capital dredging) of the port area (Trimonis and Gulbinskas 2000).

The main part of the bottom of eastern part of the strait including small bays and embayments is covered by silty mud. This fact points to the relative slowdown of hydrodynamic processes and established favourable conditions for the accumulation of fine sediments. Silty mud is also accumulated in small areas of the navigational channel dredged to the depth of 14.5 m. Sedimentation of silty sand is typical for the southern part of the strait and along the western shore and Klaipėda Port entrance channel in the north. Sandy mud has been identified in relatively small areas adjacent to zones of accumulation of fine silty sand and silty mud. Sand and coarser debris prevail in hydrodynamic transit zones, e.g. at the strait-lagoon junction in the south and at the entrance gates to the port in the north.

Most of the sediments contain high percentage of fines. Large percentage (ranging from 70 to 93 %) of fine clay and silt determines high sorption properties. Silty mud is mostly enriched with organic carbon. TOC values in those sediments range from 1.72 to 7.48 %. Maximum values of organic carbon are typical for muddy sediments of enclosed areas and dockyards, while sandy sediments of Klaipėda Strait showing the lowest values of TOC.

The positive correlation between fine fractions (<0.063 mm) and TOC ($\rho=0.87$, $p<0.05$) confirms the association of organic matter and the fine fraction of the sediments as reported elsewhere (Berg et al. 2001; Burton et al. 2004).

TBT concentrations

The variability of TBT concentration in different parts of the Klaipėda Strait area is high (mean value—55 ng Sn g^{-1} d.w., the coefficient of variation—350 %). TBT concentration in sediments is very much location-related and varies from 1 to 5,200 ng Sn g^{-1} d.w. Level of TBT contamination was classified according to the environmental quality levels proposed in the Lithuanian normative document for dredging and dumping (LAND 46A-2002 2002). TBT concentration less than 10 ng Sn g^{-1} d.w. is considered as "expected no-effect" (target level), values between 10 and 100 ng Sn g^{-1} points to slightly contaminated sediments, but still suitable for the sea dumping. A 100 ng Sn g^{-1} d.w. currently corresponds to maximum admissible concentration of TBT in dredged material. In this sense, all the values above 100 ng Sn g^{-1} are considered as extreme ones. We have also distinguished additional level of TBT contamination with concentrations above 1,000 ng Sn g^{-1} in order to highlight the most polluted areas of the strait (Fig. 2).

Performed frequency tabulation has shown that 78 % of the samples are low or slightly TBT-contaminated. TBT concentration in those samples is between 1 and 100 ng Sn g^{-1} d.w. (median value is only 17 ng Sn g^{-1} d.w.). TBT concentration is apparently lower in the western part of the strait (zone of natural sedimentation) and gradually increases towards the eastern bank. The TBT contamination in the semi-enclosed bays is relatively high. This is due to the activities of shipyards or ship repairing facilities located in those areas. The maximum TBT concentration (5,200 ng Sn g^{-1} d.w.) was recorded in sediments located in the central part of the strait—adjacent to the one of the oldest ship repair facilities (operating since 1857) and the largest shipyard in the port of Klaipėda (operating since 2003). Moreover, the site of the highest TBT concentration located close to the Dane River mouth. The drainage of Klaipėda City sewage water is carried out through this river. It has been known that municipal and industrial wastewater, sewage sludge and landfill leachates are regarded as additional tributyltin source (Barakat et al. 2001; El-Sayed et al. 1988; Fent and Hunn 1991).

Quite high TBT concentration (from 100 to 1,000 ng Sn g^{-1} d.w.) was detected also along the quays, operated by ship repairing facilities. Dockyard, located in the southern part of Malku Bay is also considerably contaminated (3,600 ng Sn g^{-1} d.w.). These results are in accordance with the results of the previous investigations (Dudutytė et al. 2007). TBT concentration in the Malku Bay by that time was also relatively high (1,920–2,400 ng Sn g^{-1} d.w.) if compared with central part of the strait (12.8–68.5 ng Sn g^{-1}) and at the entrance channel (35.8 ng Sn g^{-1}). This possibly indicates that primary source of TBT in this part of Klaipėda Strait is thought to be an antifouling agent, which historically leached from the ship hulls during ship repairing and dry-docking activities before the total ban came into force in 2008.

Relationship between TBT, grain-size and TOC

TBT levels in sediments usually show a steep concentration gradient from the pollution source such as marina or shipyard (Kim et al. 2011; Shim et al. 1999). However, due to intensive water exchange rate and variable sedimentation conditions of transitional waters TBT accumulation can be influenced by particle size (Langston and Pope 1995; Pempkowiak 1997; Stronkhorst et al. 2003) and organic carbon content in the sediments (Hoch et al. 2002; Radke et al. 2008). Therefore, an attempt was made to evaluate the impact of lithology and organic carbon on the variability in the TBT content. In this study, we investigated the relationship between the tributyltin concentration and the corresponding amount of fine fractions (<0.063 mm) as well as TOC content in the sediments.

Statistically significant positive relationship was obtained both between TBT concentration and fine particles (ρ=0.60, p<0.05) and between TBT and TOC (ρ=0.62, p<0.05) in sediments (Fig. 3).

Established correlation supports the findings of earlier studies (Berg et al. 2001; Burton et al. 2004; Fent 1996; Hoch and Schwesig 2004), suggesting that organotins are more likely to bind to fine particles associated with higher TOC content. Although statistical analysis shows the strong TBT-TOC-% of fine correlation, the graph (Fig. 3) clearly indicates that the relations are true for the low-contaminated samples (light grey dots on the graph) mainly. Samples that are highly TBT-contaminated (TBT>100 ng Sn g^{-1} d.w.) were collected near potential source of pollution (ship repairing, dockyards). TBT concentration there does not correlate much with the type of sediments and the amount of total organic matter. Statistically significant negative relationship between TBT and TOC (ρ=−0.72, p<0.05) and nonsignificant negative relationship between TBT and fines (ρ=−0.50, p>0.05) has been observed.

There is no doubt that TBT contamination is rather shipping and shipbuilding- and ship repairing-related. This has been proven by the number of investigations in shipyards and (or) dockyards worldwide (Berto et al. 2007; Bhosle et al. 2006; Ceulemans et al. 1998; Harino et al. 2007; Kim et al. 2011). Exceptionally high TBT value of 141,000 ng Sn g^{-1} d.w. was reported near a dock located in Antwerp Harbour, Belgium (Ceulemans et al. 1998). Extremely high TBT concentrations were recorded in front of a dry-dock and near surface runoff of the shipyard in Gohyeon Bay (36,292 ng Sn g^{-1} d.w.) in South Korea (Kim et al. 2011) and at one dockyard site in Venice Lagoon (39,300 ng Sn g^{-1}) in Italy (Berto et al. 2007). Little lower concentrations recorded around the shipyard located in Otsuchi Bay (14,000 ng Sn g^{-1} d.w.) in Japan (Harino et al. 2007) and in the vicinity of Cochin Shipyard located in Kochi Harbour (16,816 ng Sn g^{-1} d.w.),west coast of India (Bhosle et al. 2006).

The same is true for the southern Baltic Sea area, although published data are scarce and represent situation in Polish ports mostly. Studies carried out both before (Falandysz et al. 2006; Senthilkumar et al. 1999) and after (Filipkowska et al. 2011) the introduction of global prohibition to use harmful organotins in antifouling paints in 2008 showing relatively high contents of TBT compounds in sediments at shipyards and ports. For example, maximum concentration of TBT in the sediments of shipyard canals in the Gulf of Gdansk (detected in 1997/1998) reached 40,000 ng Sn g^{-1} d.w. (reported by Senthilkumar et al. 1999). Even higher TBT concentration (57,000 ng Sn g^{-1} d.w.) was recorded at the same shipyard canal in 2002 (reported by Falandysz et al. 2006). TBT concentration in the same ship repair yard measured right after the total ban came into force was much lower. Maximum

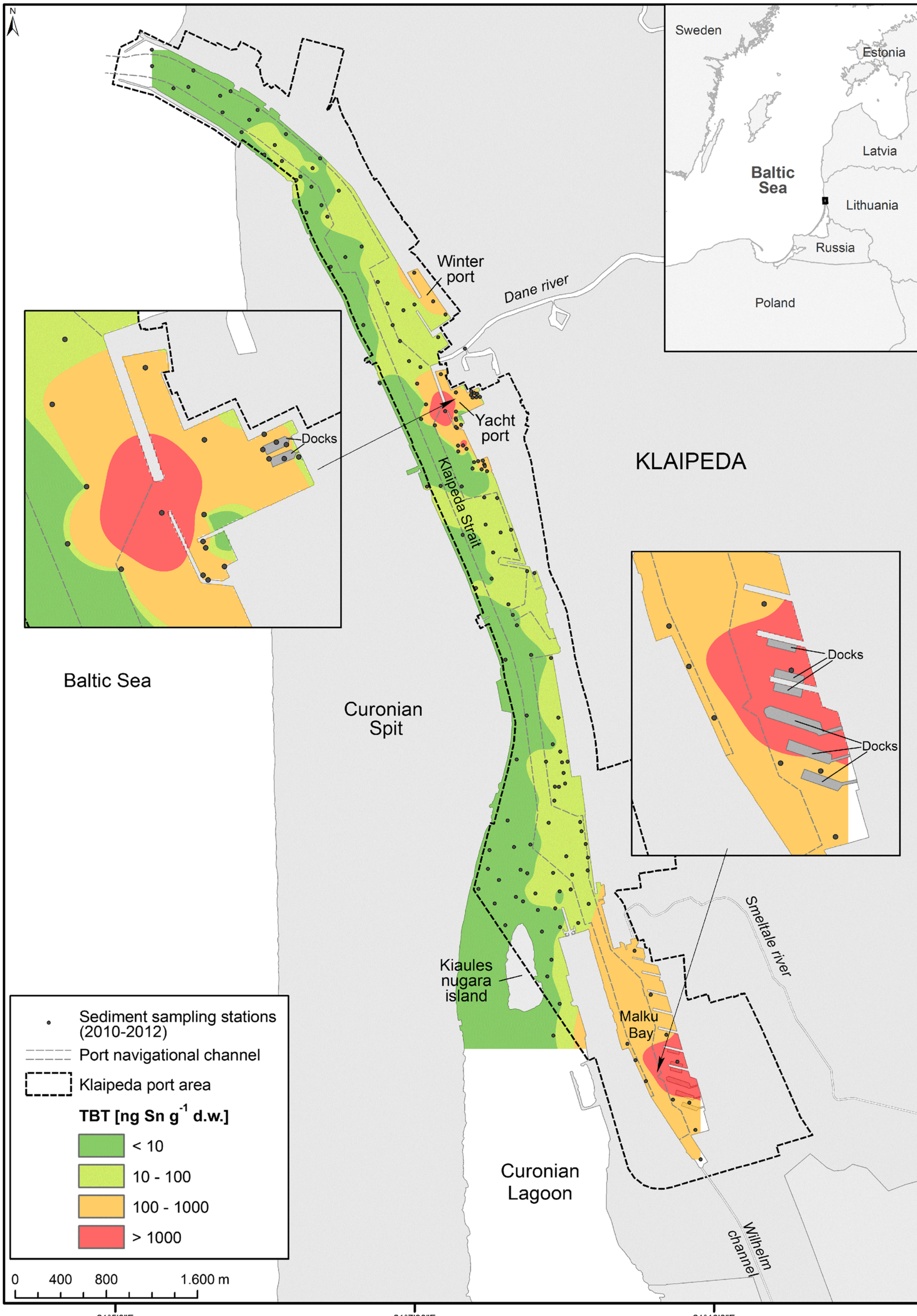

Fig. 2 Distribution of TBT-contaminated sediments in the Klaipėda Strait area

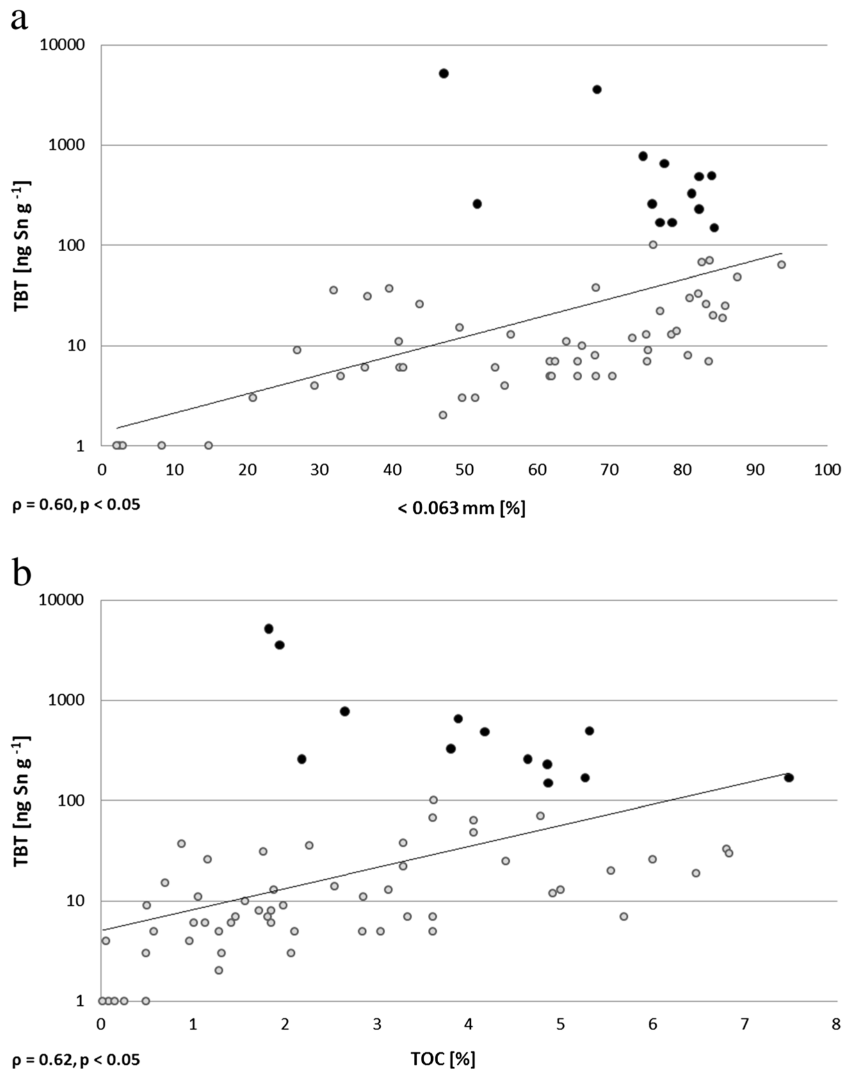

Fig. 3 Relationship between tributyltin concentrations (ng Sn g^{-1} d.w.) and fine particles, % (**a**) and TOC, % (**b**). *Solid black circles* group of stations considered as extreme values (TBT>100 ng Sn g^{-1} d.w.), *light grey circles* group of stations with values below 100 ng Sn g^{-1} d.w

TBT concentration reported by Filipkowska and others in 2011 reached 15,780 ng Sn g^{-1} d.w., and this was considerably lower than reported earlier (Falandysz et al. 2006; Senthilkumar et al. 1999), however still much higher compared to other places within the port of Gdansk.

Even though contamination in the Klaipėda Strait is relatively lower (reaching 5,200 ng Sn g^{-1} d.w.), such amounts of TBT compounds in bottom sediments are well ahead of the limits, established by Helsinki Commission (threshold equals to 2 ng Sn g^{-1} d.w.). Critical limit of allowed TBT concentration mentioned in the Lithuanian national regulations (LAND 46A-2002 2002) equals to 100 ng Sn g^{-1} d.w. The main threat of the TBT pollution is mainly related to the historic contamination but is still present in hot spots, associated with shipyards, ports and marinas.

TBT contamination in the Klaipėda Strait area is critical for dredging operations and further management of the dredged spoil. Appropriate adjustment of existing TBT limits needs to be done in order to ensure proper operation of Klaipėda port. Findings of the current study and results of other scientific studies focusing on TBT shall be used as background knowledge in order to set the reasonably sound limit values for the allowed TBT concentrations. Moreover, background concentrations in the region, effect of anthropogenic contamination as well as bioavailability of pollutant in dredged material need to be properly assessed.

Conclusions

Extensive research of tributyltin distribution in bottom sediments have been carried in Klaipėda Strait—transitional marine-lagoon system located in the south-eastern part of the Baltic Sea. The concentration of tributyltin compounds in the sediments varies and is depending on the location and sediment type. Exceptionally high TBT contamination is related to the ship repairing facilities and dockyards operating in the port. The areas of the highest TBT concentrations are contaminated via direct input of hazardous compounds into the strait area. However, lower contamination rates (less than 100 ng Sn g^{-1} d.w.) detected in the rest part of the strait may suggest that major sources of TBT pollution do not fully account for the general TBT presence in the system. Accumulation of TBT compounds in the study area is obviously governed by natural factors such as sedimentary conditions, water exchange rates, particle size and amount of organic carbon in the sediments.

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