

**Total petroleum hydrocarbons in surface sediments of the Lithuanian coastal area  
of the Baltic Sea*****Sergej Suzdalev, Saulius Gulbinskas***

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**Abstract** Operation of large oil import/export terminals and intensive shipping activities together with input of hazardous substances from terrestrial runoff and constantly developing cities makes the Lithuanian part of the Baltic Sea especially sensitive to contamination with oil products. The paper presents an overview of total petroleum hydrocarbons (TPH) distribution in surface sediments at the Lithuanian near shore and within the Klaipėda State Seaport area – transitional marine-lagoon system. The study is based on the results of examination of surface sediment samples carried out in 2010–2012. The variations of TPH content in bottom sediments are explained by differences in grain size and the genesis of the investigated sediments as well as the degree of organic material decomposition. Extreme values obtained in the Klaipėda Strait area indicate presence of additional TPH contamination sources possibly of anthropogenic origin.

**Keywords** • petroleum hydrocarbons • bottom sediments • lithology • organic matter • contamination

✉ *Sergej Suzdalev (sergej.suzdalev@jmtc.ku.lt), Saulius Gulbinskas, Klaipėda University Marine Science and Technology Centre, H. Manto 84, 92294 Klaipėda, Lithuania*

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**INTRODUCTION**

Petroleum hydrocarbons are ubiquitous pollutants in marine sediment because of industrial discharges, accidental spills, shipping activities, atmospheric fallouts, and marine oil and gas explorations (Ye *et al.* 2007; de Mora *et al.* 2010). Once released into the environment, all of these compounds are subjected to continuous and variable changes due to bacterial degradation, photo-oxidation and evaporation.

The Baltic Sea is one of the largest bodies of brackish water in the world. Its environment is contaminated from numerous point sources of oil pollution. In particular, growing maritime traffic, including oil transportation and handling in several ports have increased the risk of oil spillages and further risk for marine environment (Pikkarainen, Lemponen 2005). According to the estimations of Helsinki Commission (HELCOM) 20,000 to 70,000 t of oil enter the Baltic Sea annually and 10 % of the total amount comes from illegal discharges from flushing of machinery systems or cargo tanks of vessels (HELCOM 2003).

Presence and distribution of total petroleum hydrocarbons (TPH) in the Baltic Sea has been extensively reviewed in several earlier studies (Andrulewicz 1992; Andrulewicz, Rohde 1987; Dahlmann 1990; Granby 1987; Jörgensen *et al.* 1985; Melvasalo *et al.* 1981; Rudling 1976). Quite limited analytical data were available for the south–eastern part of the Baltic Sea. Results on presence of oil products in bottom sediments of Lithuanian waters are also quite patchy. There are just a few investigations available (Pustelnikovas 1994, 1998; Galkus 2004; Jokšas *et al.* 1998, 2005; Stakėnienė 1996), however a general assessment of pollution in the bottom sediments of the Lithuanian coastal area was still missing.

The fragile marine environment here is constantly threaten by the two large oil import/export enterprises (Klaipėdos Nafta, Būtingė Oil Terminal), intensive shipping activity, wastewater discharges from Klaipėda and Palanga cities and constant input of contaminants with waters of Nemunas River. Regular observations of TPH in water and bottom sediments of Lithuanian coastal zone, open sea and transitional waters are carried out in the frame of Lithuanian national monitoring programme.

In this study an overview of TPH distribution in surface sediments is given. Results are based on the investigations carried out in 2010–2012. The comparative analysis of TPH distribution in different sedimentary environments that are impacted from various pollution sources provides general picture of TPH contamination pattern in the Lithuanian coastal zone.

## MATERIAL AND METHODS

### Study area

Three different study sites have been selected in order to reflect differences of TPH contamination depending on the potential source of pollution. Those are: (1) marine area in the vicinity of operating Būtingė Oil Terminal; (2) deep-water dumping site used for dredged spoil utilization and (3) Klaipėda Strait as heavily modified transitional (marine and lagoon) water body accommodating the main port of Lithuania (Fig. 1).

### Būtingė Oil Terminal area

Būtingė Oil Terminal is located in the northern part of the Lithuanian coast close to the Latvian State border. It is the only buoy type oil harbor in the Baltic Sea. The single point mooring buoy is situated 7 km offshore (at the depth of ~20 m) from the coast and is connected to the onshore terminal via submerged 9.8 km long pipeline. The terminal can export up to 14 million tons of crude oil each year. Although the risk of pollution during the oil transfer is reduced to a minimum, accidental oil spill may occur during the disconnection operation of the oil carrier from the buoy during emergency situations. Several oil spills have been registered in the terminal since 1999 (reported in response plan of Būtingė Oil Terminal). The big accidents occurred in 2005, releasing 59 t of crude oil, in 1999 – 3.4 t, 2001 – 48.2 and 3 t and in 2008 – 6.5 t.

### Deep-water dumping site

The deep-water dumping site is located almost 20 km south-west from the Klaipėda Port gate at the water depth of approximately 50 m. The dumping site is in operation since 1987 and has been used for dumping of glacial and recently deposited sediments, dredged in the Klaipėda Port. By dredging and dumping the port sediments offshore, pollutants (variety of chemical substances including TPH) that have been accumulated in the fine-grained and organic-rich matter of the port are being discharged into marine environment. Therefore, sediments dumped in the deep-water dumping site may act as a potential secondary pollution source.

### Klaipėda Strait

Klaipėda Strait is a transitional marine-lagoon system with permanent water circulation (Stakėnienė *et al.* 2011) and intensive sedimentation (Trimonis *et al.* 2010). It also acts as a natural geochemical barrier zone (Emelyanov 1998). The strait accommodates the Klaipėda multipurpose port. This is the area of intensive transfer and settling of sedimentary matter provided by the Nemunas River into the Curonian Lagoon. Sediments of the strait are rich in technogenic products being handled by the port and city enterprises and therefore are the most anthropogenic loaded sea bottom sediments in Lithuania (Stakėnienė 1996). Natural processes are often influenced by the changing bottom morphology due to the capital and maintenance dredging of the port area. Risk of TPH contamination is also posed by intensive transportation of oil products to and from the port area (21 % of total cargo handled in the Klaipėda Port in 2012) with tankers of up to 100,000 t carrying capacity.

### Sample collection

Sampling of the surface sediments was carried out during the period of 2010–2012. Samples from the bottom of the sea were collected during the cruises of R/V *Darius* while sampling in the strait was done on motorboat *Emma*. Bottom sediments of the uppermost layer (0–5 cm) were collected using a Van Veen grab sampler.

Eight monitoring stations were sampled in the marine area of the Būtingė Oil Terminal (Fig. 1-1) in June 2012. Sampling stations were selected to represent the influence of variable environmental conditions and impact rate from different possible pollution sources. Station B-4 is located close to the mooring buoy, stations B-5, B-6 and B-3 are within the buoy impact zone. Station B-2 is situated close to the shore and monitors possible impact of the urban sewage discharges on the marine environment. Possible inflow of contaminants from the Šventoji River is monitored at B-1 station while B-7 is situated in the anchorage area of oil tankers. Station B-8 is chosen in order to obtain the reference value of the TPH contamination in the surrounding environment. Investigation of TPH concentrations in the sediments at the deep-water dumping site and its impact zone (Fig. 1-2) was based on the sampling at 12 monitoring stations (ED 1–12) completed in June 2012.

Sampling at Klaipėda Strait area was completed in frame of the international project SMOCS ([www.smocs.eu](http://www.smocs.eu)) during the period from April 2010 to November 2012. Altogether 152 samples have been collected in order to map TPH contamination in whole area of the strait including natural environment and sites of very intensive port activities in the open waters as well as in semi-enclosed bays.

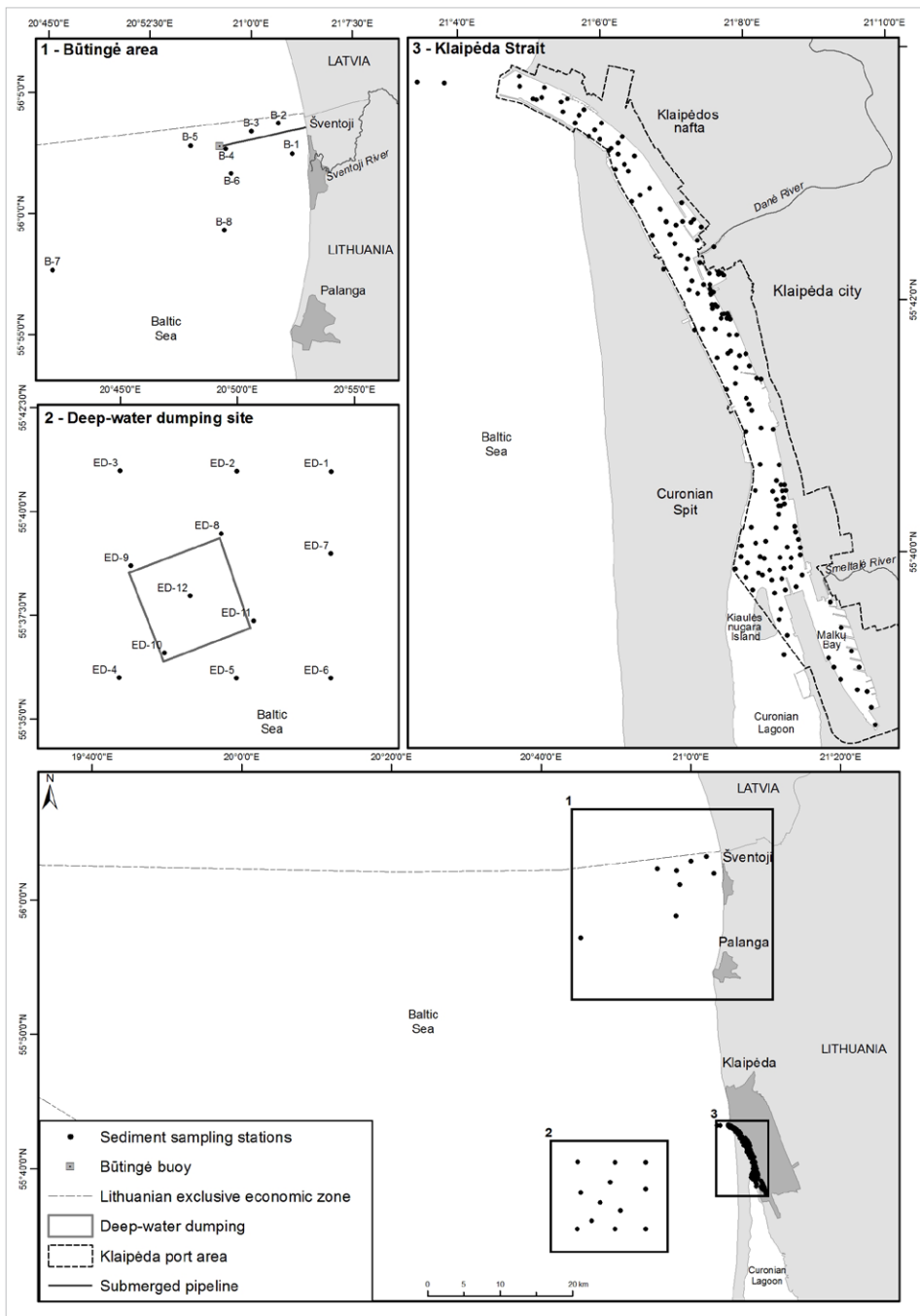


Fig. 1 Study area and sediment sampling stations. Compiled by S. Suzdalev, 2014.

## Analyses

All collected samples have been analysed for grain size distribution and TPH content. Sediments from Būtingė area and offshore dumping site were also analysed for the content of organic matter, expressed by the loss of ignition (LOI). It is characterised by the weight loss of dried sediment sample heated at 550°C and expressed in dry weight percentage. Most dominant lithological types of sediments from Klaipėda Strait were analysed for the content of total organic carbon (TOC).

The grain size composition of the sediment samples was analysed by laser diffraction method using laser particle analyser *Analysette 22 Micro Tec Plus*,

*Fritsch*. Based on the results of grain size analysis sediments were classified into six fractions in accordance with Lithuanian rules for dredging and dumping (LAND 46A-2002): coarse and fine grained sand, 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 analysis of TPH was performed in two laboratories. Samples from Būtingė and deep-water dumping area were analysed at the accredited laboratory of the Department of Environmental Protection, Maritime Institute in Gdansk. Samples from the Klaipėda Strait area were analysed at the accredited laboratory Ramboll Finland Oy, Ramboll Analytics. Content of mineral oil (C10–C40) was determined by gas chromatography with a flame ionization detector (FID). The extraction was done using penthane.

Prior to determination of TPH content, samples were homogenized and extracted with a mixture of acetone and hexane. Following procedures included removal on acetone by deionized water and drying of hexane fraction containing mineral with sodium sulfate. Polar compounds were removed with florisil and the concentration of TPH was determined by gas chromatography (GC-FID) according to the International standard method ISSO 16703:2004.

Diesel, lubricating oil and n-alkyl homologues were used as standards. The method detection limit for mineral oil is < 10 mg/kg dry weight.

## Statistical analyses

Spatial Analyst tool embedded in the ESRI ArcGIS software was used for linear interpolation of TPH concentrations in Klaipėda Strait. The relationships between the total petroleum hydrocarbons (TPH) in the sediments and organic carbon content (TOC) 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.

Statistical analysis was not applied for samples taken from Būtingė area and deep-water dumping site due to the small number of samples.

## RESULTS

### Būtingė Oil Terminal area

Silty sand was identified in the stations B1, B-2 and B-8. The amount of fine material ( $<0.063$  mm) in those stations varies from 10 to 30 %. The amount of sand fractions is bigger in the stations closer to the mooring buoy. In the stations B-4 and B-6 fine-grained ( $0.25-0.1$  mm  $> 50$  %) and medium-grained ( $0.5-0.25$  mm  $> 50$  %) sand is prevailing. At the B-3 and B-5 sand is mixed with variable amount of pebble and gravel. Grain size analysis of the sample taken at the station B-7 indicates that sea bottom in the anchorage area for oil tankers is covered with fine-grained sandy sediments.

The amount of organic matter in analysed sediments is relatively low ranging between 0.35 and 1.5 %. The highest content of organic matter (18 %, expressed as LOI) was identified in the B-2 station (Fig. 2) located close to the Latvian State border. Maximum concentration of TPH (5.44 mg/kg) was also recorded in the sample taken at the same station (B-2) close to Palanga City sewage discharger. Such increase in TPH concentration presumably is caused by the discharge of petroleum hydrocarbons rich wastewaters into the marine environment rather than terminal operations itself. The sediments taken at this station also contains higher amounts of organic matter (Fig. 2).

The TPH values identified in the vicinity of Būtingė Oil Terminal mooring buoy (0.34 mg/kg in station B-3 and 0.44 mg/kg in B-4) as well as near the mouth of Šventoji River (0.52 mg/kg in B-1) are much lower. Results of sediment analysis in remaining (B-7 and B-8) stations were below the detection limits (0.01 mg/kg).

### Deep-water dumping site

Analyses of grain size distribution of the sediments sampled in the dumping site revealed that accumulation of fine-grained silty sand is prevailing in the area. The deep-water dumping site was used for the disposal of mixed (including hard glacial till) sediments dredged from the port area in the Klaipėda Strait. Nevertheless, remains (washed out till) of glacial sediments were found in the central part (ED-12 station) of the dumping area only (Fig. 1-2).

TPH concentrations measured in the sediments around the dumping site were rather low if compared to the values obtained for the Būtingė area. The highest values (0.51 and 0.44 mg/kg) were detected in the central and western part (station ED-12 and ED-11 respectively) of the dumping site and this seems to be associated with higher amount of organic matter in the sediments (Fig. 3). Concentrations reaching 0.40 mg/kg) were also observed in the sediments sampled outside and north of the dumping area (ED-1). Concentrations of TPH measured in the rest of the samples did not exceed the detection limits (being less than 0.01 mg/kg).

### Klaipėda Strait

The capital dredging has changed the sedimentation conditions, distribution and composition of bottom sediments in the Klaipėda Strait significantly (Trimonis, Gulbinskas 2000). Results of the study show, that currently silty mud occupies almost all eastern part of the strait including small bays of low hydrodynamic intensity and favourable conditions for the accumulation of fines. Silty mud is also present locally in the navigational channel at the depths of 14.5 m. Sandy mud is deposited in relatively small areas of the strait, bordering accumulation zones of fine silty sand and silty mud. Silty sand is typical for

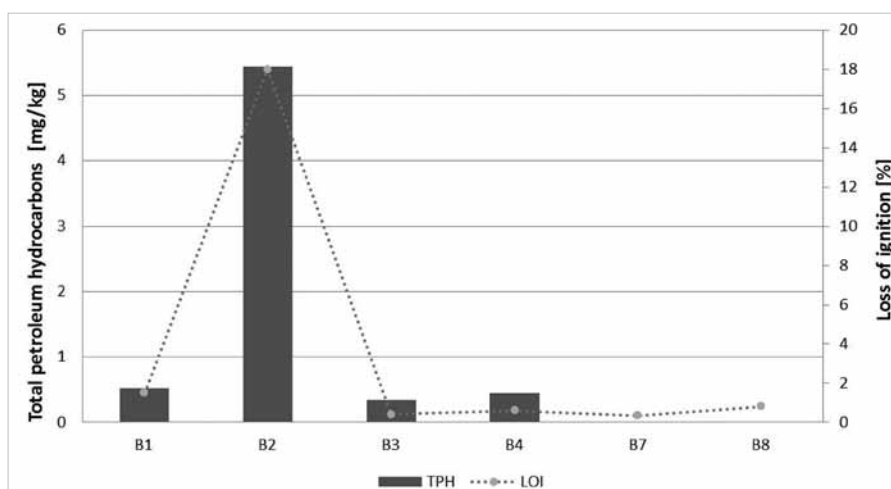
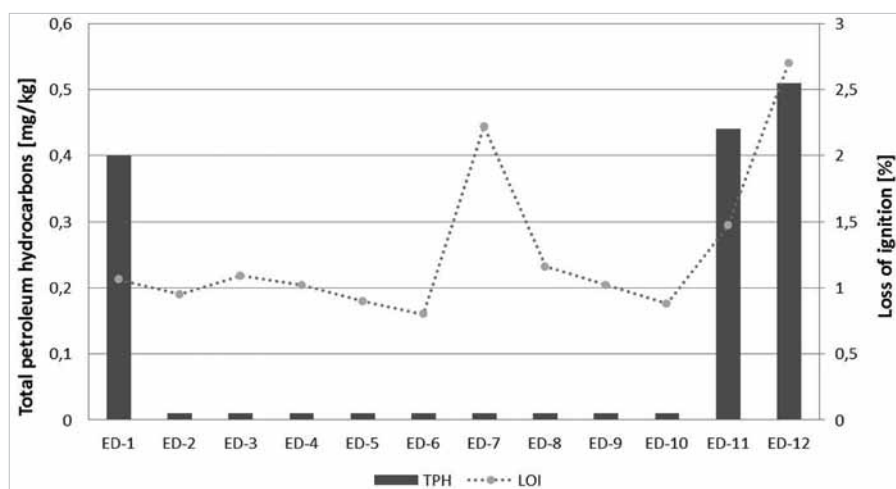


Fig. 2 TPH concentrations (mg/kg) and organic matter (LOI, %) in the sediments at Būtingė area. Compiled by S. Suzdalev and S. Gulbinskas, 2014.



**Fig. 3** TPH concentrations (mg/kg) and organic matter (LOI, %) at dumping area. Compiled by S. Suzdalev and S. Gulbinskas, 2014.

the southern part, where the strait transits into the Curonian Lagoon; along the western shore and at the northern entrance channel of the Klaipėda Port. Sand and coarser debris prevail in the sediment transit zones, e.g. the strait-lagoon junction in the south and at the entrance to the port in the north.

Most of the sediments contain high percentage (ranging from 70 to 93 %) of fines (< 0.063 mm). This determines high sorption properties of the sediments. The TOC content is highest in the silty mud (ranging from 1.72 to 7.48 %). Maximum values of TOC are typical for muddy sediments of enclosed areas and dockyards, while sandy sediments of Klaipėda Strait contain the lowest amount of organic carbon. The positive correlation ( $\rho = 0.87$ ,  $p < 0.05$ ) between fines and TOC in the sediments suggests a clear association of organic matter content and amount of fine particles in the sediments. Horizontal distribution of TPH in the bottom sediments shows an apparent gradient increase of concentrations towards the eastern bank of the strait and semi-enclosed bays (Fig. 4).

The highest quantities of TPH were identified in the semi-enclosed bays, where the water exchange is limited and silty mud is accumulating intensively. The maximum TPH concentration (1600 mg/kg) was identified in the Yacht Port close to the dockyard, which can be regarded as potential source of anthropogenic hydrocarbons. Similarly high values (500–1500 mg/kg) are common for sediments in Winter Port, along the quays operated by ship repairing companies and in the northern part of Malkū Bay at the mouth of small Smeltalė River. Least contaminated are the central and southern parts of the strait and northern entrance to the Baltic Sea. The increase of TPH concentration at the entrance of the channel can be a result of a geochemical/sedimentary barrier due to mixing of salty marine and fresh lagoon waters.

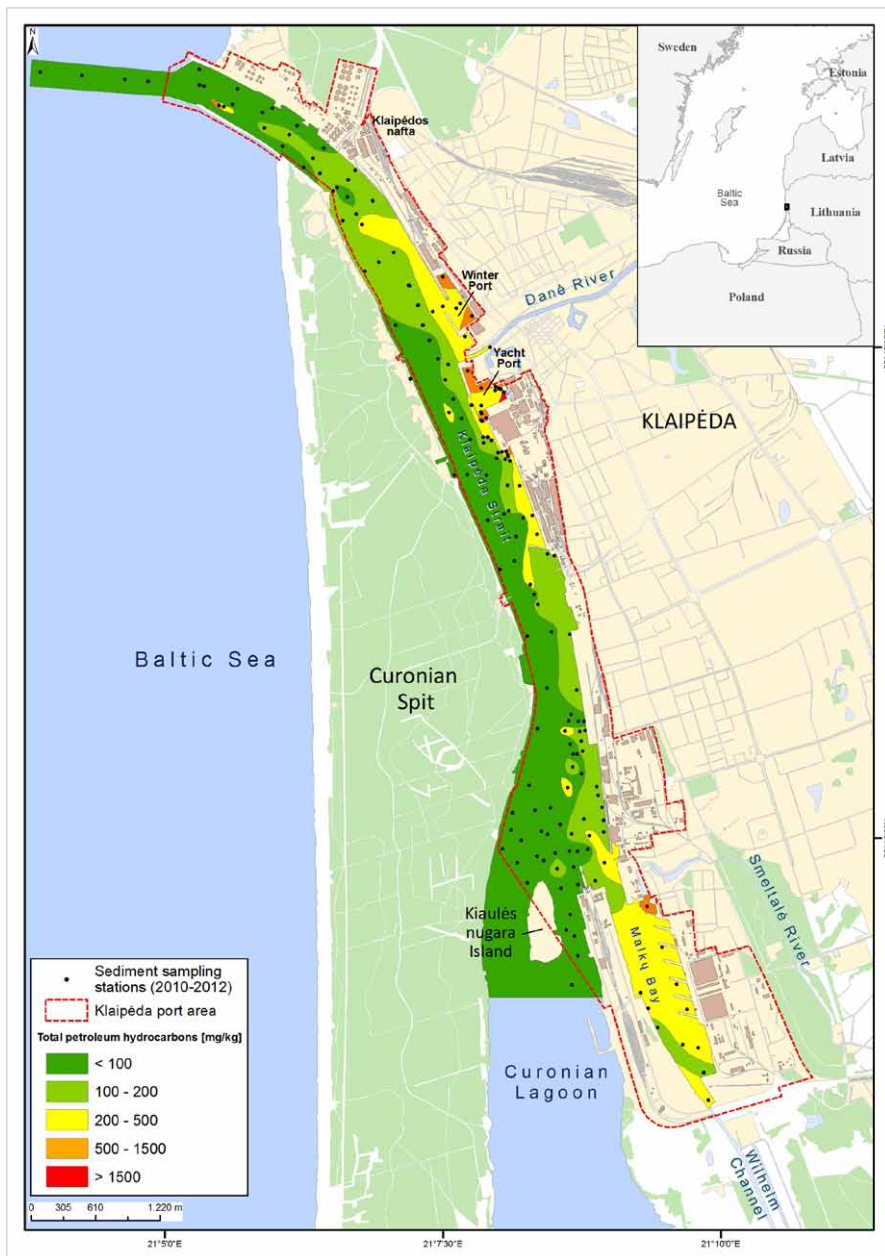
TPH accumulation can be much influenced by the amount of fine terrigenous (silt and clay) as well as biogenic material in the sediments (Stakėnienė 1996; Jokšas *et al.* 2005). Current study evaluates the impact of lithological composition and content of organic carbon on the variability in the hydrocarbons concentration in the sediments. The relationship between the TPH concentration, amount of fine fractions (< 0.063 mm) and TOC content in sediments was analysed (Fig. 5). Statistically significant, moderately strong positive relationship was established both for TPH concentrations and amount of fine particles ( $\rho = 0.69$ ,  $p < 0.05$ ) and for TPH and TOC ( $\rho = 0.73$ ,  $p < 0.05$ ) in the sediments. The findings point to the conclusion, that in natural conditions TPH are more likely to bind to fine particles associated with higher TOC content.

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## DISCUSSION

Oil pollution caused by large accidents or long-term small-scale spills and leakage is recognised as one of the greatest hazards for the marine environment. As reported earlier the amount of oil hydrocarbons in surface sediments of the open Baltic Sea area varies from 1 to 326 mg/kg dry weight (Nemirovskaya 2004). Obviously, such huge variations of TPH concentration in the Baltic Sea sediments cannot be explained by lithological composition of bottom sediments and amount of organic matter only. Results from this study show that TPH concentrations in the sediments can be much higher in the areas close to the direct pollution sources – in the ports and near the river mouths.

Variations of TPH concentrations in bottom sediments and water of the Lithuanian coastal area (open sea, near shore zone up to 20 m depth, offshore dumping area, Curonian Lagoon and Klaipėda Strait) are often used as indicator for the assessment of anthropogenic pressure related to oil transportation, shipping, illegal discharges of oil in the open sea, dredging of port areas and further disposal of sediments at the offshore dumping sites. Results showed that distribution of petroleum hydrocarbons in the Baltic Sea bottom sediments correlates well with distribution of fine-grained sediments in general. The concentration of TPH increases with increasing amount of organic matter in bottom sediments. Concentrations of petroleum hydrocarbons in coarse-grained bottom sediments are low (1–2 mg/kg).



**Fig. 4** Distribution of TPH (mg/kg) in the surface layer (0–5 cm) of Klaipėda Strait. Compiled by S. Suzdalev, 2014.

Relatively high TPH concentrations (reaching 56.5 mg/kg) in marine bottom sediments near Klaipėda were reported (Jokšas *et al.* 2003). Similarly high values of TPH (59 mg/kg) were identified in the local depressions near the Klaipėda Port gate during the complex research of pollutants carried in 2006 (Garnaga *et al.* 2008).

Certain increase of TPH concentrations was also recorded at Būtingė marine area. For instance in 2006 the concentration of TPH in bottom sediments close to the Būtingė buoy reached 25 mg/kg (Jančiauskienė, Jokūbauskaitė 2003). The pollution is possibly related to the big accident occurred in 2005, when 59 t of crude oil were released to the marine environment. Oil products, if accumulated, can remain in the bottom sediments for relatively long period of time

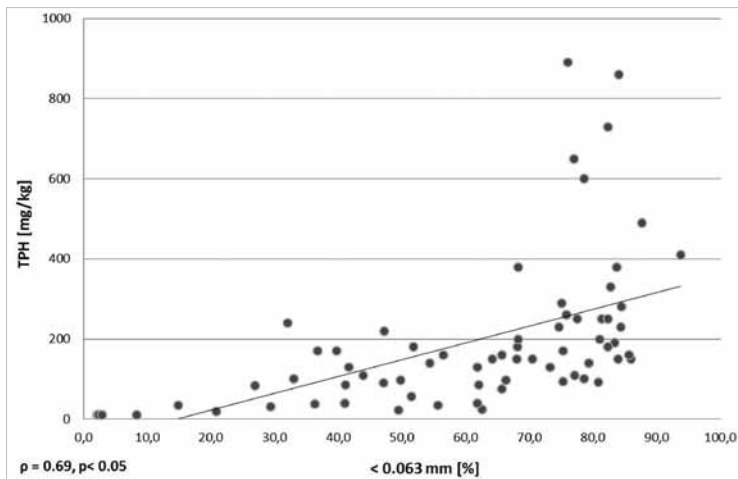
(Nemirovskaya, Pustelnikov 1984). Usually, sediments from Būtingė area are little or not contaminated with TPH. Therefore, it might be assumed that operation of the oil terminal does not affect the marine environment considerably. On the other hand, accumulation of hydrocarbons is influenced by sedimentation conditions in this region. There are no areas of permanent accumulation of fine sediments within the terminal's impact zone. Therefore, higher concentrations of hydrocarbons can be observed right near the places of direct pollution perhaps caused by permanent pollution.

Long-term investigations carried out in the Baltic Sea have revealed that concentration of petroleum hydrocarbons in the bottom sediments at the dumping area is higher than observed in the open sea (Jokšas *et al.* 2005). Average concentrations of TPH in deep-water dumping site measured for the period from 1996 to 2006 varied between 2 and 18 mg/kg dry weight (Garnaga *et al.* 2008). Maximum values were usually associated with disposed loamy sediments dumped from the Klaipėda Port area. According to the results of current study, maximum concentration of TPH was identified in mixed sediments of clayey and sandy loam (0.51 mg/kg). Such amount is considerably lower than reported earlier (Garnaga *et al.* 2008) and does not prove the serious impact

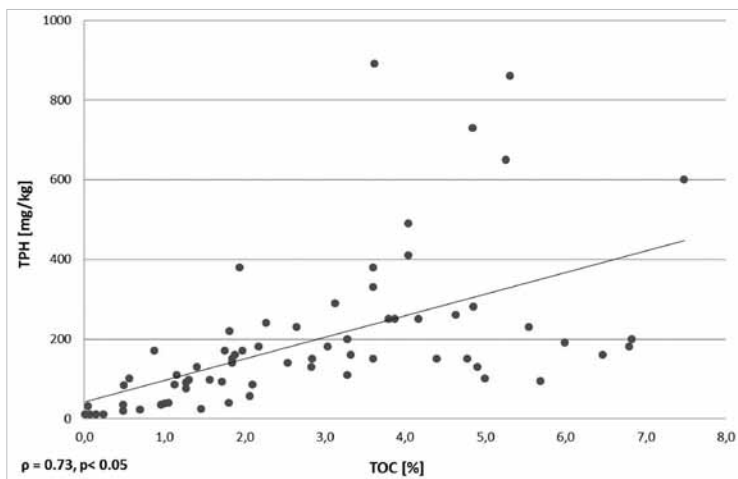
of dumping activities on the marine environment.

Current study reveals that Klaipėda Strait is heavily TPH contaminated. Sediments of this particular transitional environment accumulate petroleum hydrocarbons from different sources – atmospheric discharges, terrestrial runoff, technogenic loads of the Klaipėda Port and city, accidents and the shipping (Jokšas *et al.* 2005).

According to the data of 1994–1997, the concentration of TPH in the bottom sediments of Klaipėda Port varied from 0.26 in sandy to 2029 mg/kg in silty clayey sediments (Stakėnienė 1999). Similar regularity of TPH accumulation was identified during the current study. Comparison of average TPH concentrations in different lithological types of Klaipėda Strait measured in different periods is presented below (Fig. 6).

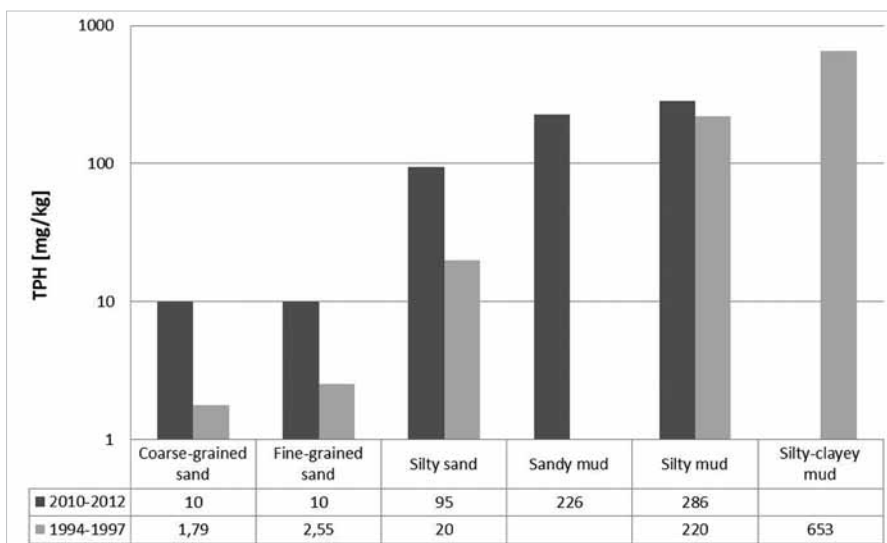


a



b

**Fig. 5** Relationship between TPH concentrations (mg/kg), amount of fines, % (a) and TOC, % (b). Compiled by S. Suzdalev, 2014.



**Fig. 6** Average concentrations of TPH in sediments at Klaipėda Strait in 1994–1997 and in 2010–2012. Compiled by S. Suzdalev and S. Gulbinskas, 2014.

The TPH contamination of particular types of sediment has increased by five times and more since 1997. Results clearly demonstrate that oil related activities have been intensifying during last years. The amount of TPH has a general tendency to be higher in fine-grained sediments with high content of TOC. However, this rule is not valid in the areas close to the direct influx of TPH from the pollution source. Ship repairing facilities are among the most dangerous sources of potential contamination within the port area, while other part of pollutants reaches Klaipėda Strait from Danė and Smeltalė rivers.

## CONCLUSIONS

Concentrations of TPH in surface sediments at the Būtingė Oil Terminal marine area range between 0.01 and 5.44 mg/kg. At the deep-water dumping site the amount of petroleum hydrocarbons was from 0.01 to 0.51 mg/kg. The variations of TPH concentrations are mostly associated with quantity of biogenic matter in the sediments.

Much higher values of TPH were identified in the area of Klaipėda Strait. Sediments of transitional (between lagoon and the sea) water system trap petroleum hydrocarbons originating from different sources. Maximum concentrations (reaching 1500–1600 mg/kg) of TPH were identified in silty muds of semi-enclosed bays and jetties. Amount of TPH content in the natural environment correlates well with the amount of fine sediments and organic material. Whereas, in the port areas the TPH contamination pattern is significantly related to the existing pollution sources and economic activities of port related industry.

## Acknowledgements

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