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POLYAROMATIC HYDROCARBONS POLLUTION ASSESSMENT OF THE SLOVENIAN SEA

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ABSTRACT

The paper assesses pollution of the Slovenian sea in terms of polyaromatic hydrocarbons content in sediment samples and mussels. For this purpose, fluorescence spectroscopy was applied as a screening method. Elevated concentrations of polyaromatic hydrocarbons were obtained in municipal harbours of the three Slovenian coastal cities, as well as in marinas along the coast. The Bay of Koper is more polluted in comparison to the Bay of Piran, which seems to be the less polluted area of Slovenian coastal waters. The Bay of Koper is affected by various port activities and maritime traffic. In the central part of the Gulf of Trieste, a certain influence of the city of Trieste with its port and maritime traffic was observed. In general, the Slovenian sea could be considered as moderately polluted.

Key words: polyaromatic hydrocarbons, pollution, sediments, mussels, fluorescence spectroscopy, Slovenian sea

VALUTAZIONE DI INQUINAMENTO IN MARE SLOVENO CON L'AUSILIO DI IDROCARBURI POLIAROMATICI

SINTESI

Nell'articolo viene presentata la valutazione dell'inquinamento del mare sloveno con l'ausilio delle concentrazioni di idrocarburi poliaromatici in campioni di sedimento e bivalvi. A tale scopo è stata adoperata la spettroscopia di fluorescenza. Concentrazioni elevate di idrocarburi poliaromatici sono state registrate nei mandracchi dei tre comuni costieri come pure nelle marine costiere. La baia di Capodistria è risultata più inquinata di quella di Pirano, che sembra essere l'area meno inquinata del mare sloveno. La baia di Capodistria viene condizionata dal porto e dal traffico navale. La parte centrale del Golfo di Trieste risente dell'influsso della città di Trieste, del rispettivo porto e del traffico navale. In conclusione, il mare sloveno risulta essere moderatamente inquinato da idrocarburi poliaromatici.

Parole chiave: idrocarburi poliaromatici, inquinamento, sedimenti, bivalvi, spettroscopia di fluorescenza, mare sloveno

INTRODUCTION

Hydrocarbons of natural and anthropogenic origin are widely distributed in different environmental compartments, i.e. soils, sediments, natural waters and biota. The awareness of the various sources of introduction into the natural environment, dispersal of these compounds, transport mechanisms and the concentrations in different parts of the environment are very important to adequately assess the state of the environment. The most important sources of pollution by hydrocarbons are oil seepage, oil spillage, combustion of various fossil fuels, traffic, domestic and industrial waste waters and sewage, as well as urban runoff (IMO, 1993). Because of their low solubility in water and rather low vapour pressures, the adsorption of hydrocarbons onto solid particles is a very important process in the natural environment (Means *et al.*, 1980). Hydrocarbons are relatively stable in the natural environment and they accumulate in sediments and biological communities (Guzella & de Paolis, 1994; Quintero & Diaz, 1994). For this reason, sediment and biota samples are usually used for the analyses since the concentration in seawater is usually very low due to the fast dilution. The most used marine organisms for this purpose are mussels. They are widespread in coastal waters, and owing to their filtrating ability they accumulate contaminants in their tissue.

The study of the impact and fate of hydrocarbons is therefore of great importance, especially in marine coastal waters since these areas are biologically active and receive considerable pollutant inputs from land-based sources via coastal and river runoff.

The aim of the present work was to investigate the state of marine environment in the Slovenian sea in terms of pollution with polyaromatic hydrocarbons. For this purpose, sediment samples and mussel tissue (*Mytilus galloprovincialis*) collected in 2005 and 2006 were analyzed. Sediment samples were not separated according to the grain size. It is well known that the adsorption of different compounds onto sediment particles is dependant on the grain size since smaller particles have bigger specific area for the adsorption. The aim of this work was merely to compare different coastal areas and to assess the pollution with polyaromatic hydrocarbons and not a detailed study of polyaromatic hydrocarbons distribution. The content of polyaromatic hydrocarbons was determined using UV fluorescence spectroscopy. This technique was established for the determination of total hydrocarbons, although the major part of fluorescence arises from polyaromatic hydrocarbons (Mason, 1987). This method is quite fast and very sensitive. Its weakness could be that it can only provide information concerning the classes of compounds (Mason, 1987) whereas its application for the determination of the concentrations of individual compounds is rather limited (Ehrhardt & Knap, 1989). On the other hand, the

fluorescence may also be due to degradation products generated from oil in the natural environment (Ehrhardt & Knap, 1989). These compounds are important constituents of the unresolved complex mixture (UCM), determined usually as a background signal in GC chromatograms. The UCM was considered a better measure of the level of petroleum hydrocarbons than the resolved components (Farrington *et al.*, 1982). In addition, the fluorescence spectroscopy is less susceptible to the influence from biogenic compounds, since biological tissue does not usually contain significant amounts of fluorescing species (Mason, 1987). This method is thus suitable as a screening method for the assessment of the state of pollution in a certain area (Ehrhardt & Knap, 1989).

Fluorescence spectroscopy was used for determination of petroleum hydrocarbons in sea water (El Samra *et al.*, 1986; Dujmov & Sucevic, 1989; Ehrhardt & Knap, 1989; Ehrhardt & Petrick, 1989; Ferrer *et al.*, 1998), marine sediments (Badawy *et al.*, 1993; Ehrhardt & Burns, 1993; Al Lihaibi & Al Omran, 1996; Peterson *et al.*, 2001; Gonzales-Macias *et al.*, 2007) and biota (Mason, 1987; Badawy *et al.*, 1993; Ehrhardt & Burns, 1993; Watson *et al.*, 2004; Rodriguez-Sanmartin *et al.*, 2005). Chrysene and crude oil of different origin were most frequently used as standards (Dujmov & Sucevic, 1989; Ehrhardt & Petrick, 1989; Badawy *et al.*, 1993; Rodriguez-Sanmartin *et al.*, 2005), although mixtures of different PAHs were also examined (Mason, 1987; Owen *et al.*, 1995; Peterson *et al.*, 2002). Lately, some new techniques of this method, e.g. synchronous fluorescence spectroscopy, have also enabled a better determination of some individual fluorescing compounds (Disanayake & Galloway, 2004; Hua *et al.*, 2007).

MATERIAL AND METHODS

Study area

The investigated area to the southeast of the Gulf of Trieste is part of the northern Adriatic (Fig. 1). Slovenian coastal area is one of the fast developing regions of Slovenia (Turk & Potočnik, 2001). The number of inhabitants has increased in the last decades and has now reached 80,000 (in the summer period this number may increase five fold). The most important industries in the Slovenian coastal region are metal manufacturing, organic chemicals production and food industry. The agriculture is mostly oriented towards the production of wine, fruit and olive growing, and vegetable cultivation. The Port of Koper has become one of the most important ports in the northern Adriatic and is still increasing its activities. It handles about 10 million tons of cargo per year, among them over 1.5 million tons of oil and oil products and more than 100,000 tons of chemicals. In the studied area, the nautical tourism has also developed with three marinas, in Portorož, Izola and Koper.

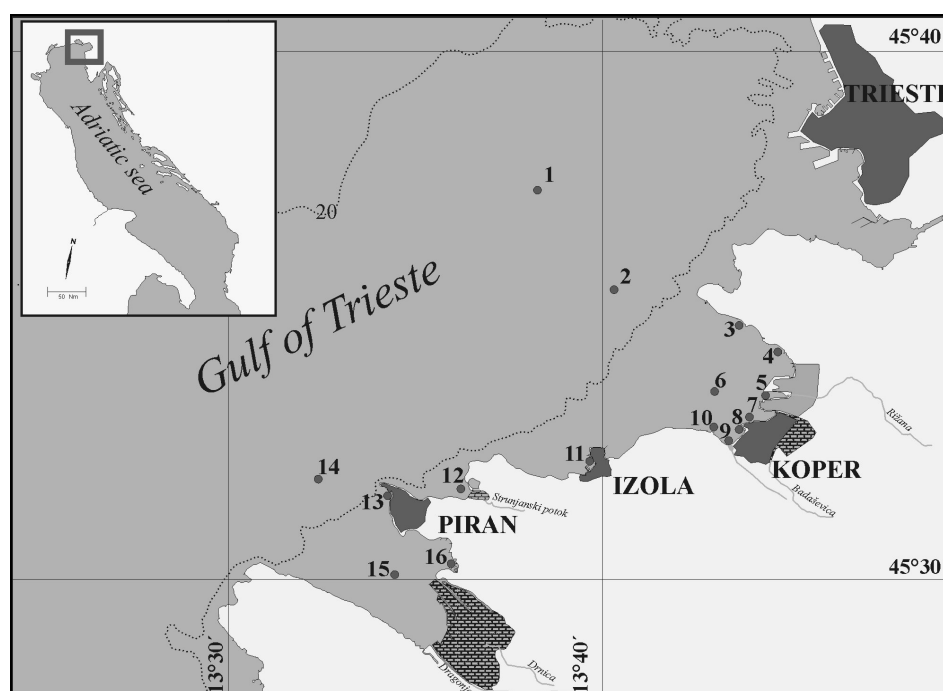


Fig. 1: Sampling sites within the investigated area.
Sl. 1: Vzorčevalna mesta na preiskovanem območju.

One of the severest problems concerning marine environment in this region is sewage discharge. Only two central sewage treatment plants are active in the cities of Koper and Piran, and even there only primary treatment is operating. The sewage from the community of Izola is discharged almost without any treatment. There are also some minor outlets discharging sewage from some smaller towns on the coast. Apart from the above mentioned point sources of pollution, some diffuse sources should also be considered. These are road runoff from the coastal highway, especially significant during the high tourist season, runoff from agricultural areas, river discharges and atmospheric deposition. This area is also affected by pollution sources from other parts of the Gulf of Trieste, sewage discharges from the city of Trieste and intense maritime traffic to the ports of Trieste and Monfalcone (over 30 million tons of oil and oil products per year). A very important source of pollution is the river Isonzo, collecting industrial and municipal wastewaters and discharging them in the northern part of the Gulf of Trieste.

The distribution of sampling sites in Slovenian coastal waters is presented in figure 1. A very short description of sampling sites is given in Table 1. It has to be mentioned that even though the mussels are very widespread in our sea it was not possible to sample them at all sampling sites.

Experimental

"For residue analyses quality" hexane, methanol and methylene chloride (Fluka) were used for the extraction of hydrocarbons. All other chemicals were of analytical grade, purchased from Merck (Germany). Na_2SO_4 was precleaned by Soxhlet extraction with methanol and hexane and additionally precombusted at 400°C for 6 hours. SiO_2 and Al_2O_3 were precleaned by Soxhlet extraction with methanol and hexane for 8 hours and then dried at 105°C. Before use both oxides were activated at 200°C for 4 hours and then partially deactivated by adding 5% of precleaned water. Cu was activated with HCl and rinsed several times with distilled water, acetone and hexane. It was kept in hexane before used.

Sediment samples were taken by gravity corer. The top 2 cm layer was used for further analyses. Samples were frozen and eventually freeze-dried. Dry sediment was homogenized. About 10 g of sediment was transferred in a Soxhlet apparatus and extracted for 8 hours with 200 ml of hexane/methylene chloride (1:1). The extracts were dried with Na_2SO_4 , concentrated on a rotary evaporator and additionally in a nitrogen stream. Sulphur was removed overnight by the addition of activated Cu. After additional concentration of extracts under nitrogen stream, the partitioning in aliphatic and aromatic fraction was performed on $\text{SiO}_2/\text{Al}_2\text{O}_3$. Aromatic fraction was concentrated and used for the analyses.

Tab. 1: Short description of the sampling sites.**Tab. 1: Kratek opis vzorčevalnih mest.**

Sampling site	Description	Sample
1	Central part of the Gulf of Trieste	Sediment
2	Outer margin of the Bay of Koper	Sediment
3	Northern part of the Bay of Koper	Mussels
4	Small harbour	Mussels
5	Port of Koper	Sediment
6	Central part of the Bay of Koper	Sediment
7	Marina of Koper	Mussels
8	Municipal harbour of Koper	Sediment, mussels
9	Mouth of the river Badaševica	Sediment, mussels
10	Southern part of the Bay of Koper	Mussels
11	Municipal harbour of Izola	Sediment, mussels
12	Strunjan protected area, cultivation of mussels	Sediment, mussels
13	Municipal harbour of Piran	Sediment
14	Outer margin of the Bay of Piran	Sediment
15	Central part of the Bay of Piran	Sediment
16	Marina of Portorož	Sediment

Mussels were sampled manually using a home-made scraper. Each sample was composed of 15 mussels and analyses were performed in 5 parallels. Samples of the whole mussel tissues were homogenized and then freeze-dried. Dry samples were extracted in a Soxhlet apparatus with 200 ml of methanol for 6 hours. Lipids were saponified by the addition of KOH solution and additionally extracted for 2 hours. Hydrocarbons were extracted with hexane, extracts dried with Na₂SO₄ and concentrated on a rotary evaporator. After additional concentration under nitrogen stream, the hydrocarbons were partitioned in two fractions as already described in the case of sediment samples. Extractable organic matter (EOM) was determined from the mass difference after the evaporation of a certain volume of hexane extracts.

Concentrations of polyaromatic hydrocarbons were determined on a Perkin Elmer LS 30 spectrofluorometer. The excitation and emission wavelengths were 310 nm and 360 nm, respectively. The calculations were made on a chrysene standard basis.

RESULTS AND DISCUSSION

The concentrations of polyaromatic hydrocarbons in marine sediment samples are presented in Table 2. They ranged from 4.2 µg/g to 227.0 µg/g. The highest concentrations were obtained in the municipal harbours of three coastal cities, Koper, Izola and Piran. The water exchange in these areas is very limited and the accumulation of different pollutants is quite important. Higher content of polyaromatic hydrocarbons is an indication of pollution from smaller boats due to exhaust emissions, leaking from engines and reservoirs as well as accidental spillage during the maintenance activities. The harbours

are also affected by the runoff from the city streets. The atmospheric deposition seems also not to be negligible, especially in the Izola harbour due to important emissions in the past from the chimney stack of the fish industry. Concentrations in these three harbours are even significantly higher than in the Port of Koper and Marina of Portorož, which are usually thought to be the most polluted areas in the Slovenian sea. Comparison of the polyaromatic hydrocarbons content in the Bay of Koper and Bay of Piran reveals that the latter is less polluted. This area, extended to the area off Piran (sampling site 14), emerges as a rather unpolluted part of the Slovenian sea.

Tab. 2: Concentrations of polyaromatic hydrocarbons in sediment samples (in µg/g dry sediment, chrysene equivalents).**Tab. 2: Koncentracije poliaromatskih ogljikovodikov v vzorcih sedimenta (v µg/g suhega sedimenta, v ekvivalentih krizena).**

Sampling site	Concentration (µg/g)
1	17.2
2	8.6
5	18.0
6	14.0
8	49.6
9	9.4
11	68.6
12	17.2
13	227.0
14	4.2
15	6.2
16	13.5

The Bay of Koper is affected by pollution from the Port of Koper. A decreasing gradient is observed from the port to the outer margin of the bay (sites 5, 6 and 2). Concentrations in the central part of the Gulf of Trieste do not follow this gradient, since they are significantly higher. This is most probably due to the influence of the city of Trieste (including sewage discharge) and intensive maritime traffic to the Port of Trieste. The rather high concentrations of polyaromatic hydrocarbons at sampling site 12 (Strunjan Bay) were surprisingly higher than expected. This area is a protected area and it was usually used as a reference site. The results of this study revealed comparable polyaromatic hydrocarbons content to those in the Port of Koper and central part of the Bay of Koper. In the case of the Port of Koper, however, the dredging activities can affect the content of different pollutants in marine sediments and direct comparison could sometimes be difficult. One of the possible sources of pollution with hydrocarbons at sampling site 12 could be a small stream, which is bringing runoff waters from the crossroad on the main coastal road to Croatia.

The second part of the work comprised the analyses of polyaromatic hydrocarbons in mussel samples collected along the Slovenian coast. Mussels are usually used for this purpose, since they accumulate pollutants in their tissue. General characteristics of the mussel samples are presented in Table 3. Some of these characteristics are usually used as "normalization factors" for better comparison of results, since the accumulation of pollutants is also time dependant. The content of pollutants in different environmental samples is significantly dependant on the conditions in the environment. This is the reason why the extractable organic matter was used to normalize the results (Gonzales-Macias *et al.*, 2007).

The highest concentration of polyaromatic hydrocarbons in mussel tissue was obtained at sampling site 11 in the Bay of Izola. This is in accordance with high polyaromatic hydrocarbons content obtained in sediments at the same sampling site. Much lower, but still elevated concentrations were obtained in the marina and municipal harbour of Koper. Concentrations at other

sampling sites in the Bay of Koper were rather low, except those at site 4, situated in a small harbour at Sv. Katarina, an area with important sea-grass meadow of *Cymodocea nodosa*. This area is close to the Port of Koper and as such affected by port activities and maritime traffic. Elevated concentrations of polyaromatic hydrocarbons in sediments at sampling site 12 are not reflected in a higher amount in mussel sample. This area is used for mussel cultivation and the time of accumulation of pollutants is most probably shorter. This could also be an important observation, taking into account human health.

As previously mentioned, the different time of accumulation of pollutants, e.g. different mussel age, can affect the interpretation of the results and may lead to incorrect conclusions. For this reason the concentrations of polyaromatic hydrocarbons in mussel samples were expressed on the amount of extractable organic matter (Fig. 2). The distribution pattern remained very similar, while the difference in concentrations between sampling sites 4, 7 and 8 became less significant (Tab. 4).

There are not many results in the literature about the hydrocarbons pollution of sediments or biota assessment using fluorescence spectroscopy. Some authors (Gonzales-Macias *et al.*, 2007) established the background concentration of total aromatic hydrocarbons in sediments of the Mexican Salina Cruz Bay at 5 µg/g. The highest concentrations in this area exceeded 2000 µg/g. In the Arabian Gulf, the concentrations up to 90 µg/g were related to moderate chronic pollution, while the content in heavily polluted areas reached 1448 µg/g (Massoud *et al.*, 1996). Concentrations in surface sediments of the Black Sea were in the range from 2 µg/g to 310 µg/g and could be comparable to those encountered in the Mediterranean (Readman *et al.*, 2002). Some fairly old results for the Mediterranean (Mille *et al.*, 1982; Barnier *et al.*, 1986) confirm this established range of concentrations in sediment samples. Concentrations in sediments from the open Adriatic Sea were below 1 µg/g (Dujmov & Sučević, 1989). Some authors (Bajt, 2000; Notar & Leskovšek, 2000; Notar *et al.*, 2001) presented some results of the analyses of polyaromatic hydrocar-

Tab. 3: Characteristics of mussel samples.

Tab. 3: Značilnosti vzorcev školjk.

Sampling site	Length (cm)	STD	Width (cm)	STD	Weight, wet (g)	STD	EOM (mg/g)
3	5.7	0.1	2.7	0.1	4.1	0.3	143
4	5.2	0.2	2.9	0.2	3.8	0.3	150
7	5.0	0.2	2.6	0.2	2.4	0.3	236
8	5.9	0.1	2.7	0.1	1.8	0.2	180
9	5.4	0.1	2.9	0.1	4.2	0.6	194
10	4.6	0.2	2.4	0.2	2.9	0.6	148
11	4.5	0.1	2.3	0.1	1.8	0.2	125
12	6.9	0.1	3.4	0.1	4.8	0.4	207

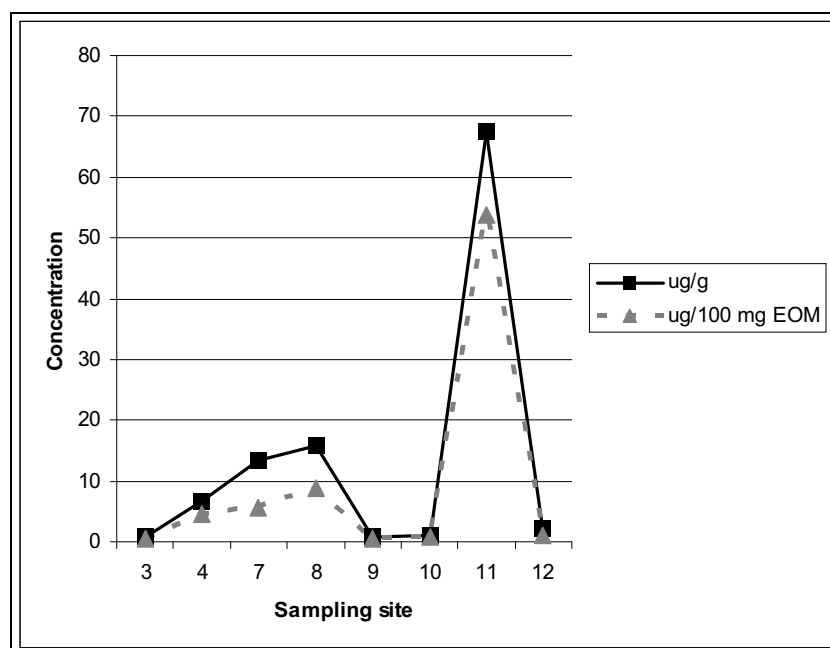


Fig. 2: Concentrations of poliaromatic hydrocarbons in mussel tissue normalized on EOM.
Sl. 2: Koncentracije poliaromatskih ogljikovodikov v tkivu školjk, normalizirane na EOS.

bons (PAH) in sediment samples and mussel samples from the Gulf of Trieste. These analyses were performed by gas chromatography. Unfortunately, the direct comparison of these results is not possible. The content obtained by fluorescence spectroscopy is usually higher, considering that it also includes degradation products and other fluorescing poliaromatic hydrocarbons.

Tab. 4: Content of poliaromatic hydrocarbons in mussel samples (in $\mu\text{g/g}$ dry weight, chrysene equivalents).
Tab. 4: Vsebnost poliaromatskih ogljikovodikov v vzorcih školjk (v $\mu\text{g/g}$ suhe teže, v ekvivalentih krizena).

Sampling site	Concentration ($\mu\text{g/g}$)	STD
3	0.8	0.1
4	6.7	1.6
7	13.4	3.1
8	15.9	0.8
9	0.9	0.1
10	1.0	0.1
11	67.3	7.1
12	2.2	0.3

Fluorescence spectroscopy was used for the analyses of biota samples as well. Concentrations below $1 \mu\text{g/g}$ (wet weight) of hydrocarbons in farmed mussels *Mytilus galloprovincialis* from Galicia (Spain) were detected (Rodriguez-Sanmartin *et al.*, 2005).

Comparison of the results obtained in the present study with the above presented results from the literature

leads to the conclusion that Slovenian sea is, in general, only moderately polluted with poliaromatic hydrocarbons. There are some areas (harbours, marinas) with elevated content of poliaromatic hydrocarbons due to the limited exchange of seawater and consequently higher accumulation.

CONCLUSIONS

The fluorescence spectroscopy was used for the assessment of the state of Slovenian sea in terms of the pollution with poliaromatic hydrocarbons. The results of the analyses of sediment and mussel samples showed elevated concentrations of poliaromatic hydrocarbons in municipal harbours of the three Slovenian coastal cities, as well as in marinas along the coast. The Bay of Koper is more polluted in comparison to the Bay of Piran, which could be considered a less polluted area of the Slovenian coastal waters. The Bay of Koper is affected by port activities and maritime traffic. In the central part of the Gulf of Trieste, a certain influence of the city of Trieste with its port and maritime traffic was observed. In general, Slovenian coastal sea may be considered moderately polluted with poliaromatic hydrocarbons.

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OCENA ONESNAŽENJA SLOVENSKEGA MORJA S POLICIKLIČNIMI AROMATSKIMI OGLJIKOVODIKI

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POVZETEK

V članku je predstavljena ocena onesnaženosti slovenskega morja z vidika vsebnosti poliaromatskih ogljikovodikov v vzorcih sedimenta in školjk. V ta namen je bila uporabljena fluorescenčna spektroskopija. Povišane koncentracije poliaromatskih ogljikovodikov smo zasledili v komunalnih mandračih treh slovenskih obalnih mest in tudi v marinah vzdolž obale. Koprski zaliv je bolj onesnažen v primerjavi s Piranskim zalivom, ki je videti tudi kot najmanj onesnaženo območje slovenskega obalnega morja. Koprski zaliv je pod vplivom pristanišča in pomorskega prometa. V osrednjem delu Tržaškega zaliva je opazen tudi vpliv mesta Trst s pristaniščem in pomorskim prometom. V glavnem lahko zaključimo, da je slovensko morje le zmerno onesnaženo s poliaromatskimi ogljikovodiki.

Ključne besede: poliaromatski ogljikovodiki, onesnaženje, sedimenti, školjke, fluorescenčna spektroskopija, slovensko morje

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