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Influence of suspended matter on cadmium accumulation in sediment from Kaštela Bay, Adriatic Sea, Croatia

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Seasonal and spatial distributions of cadmium (Cd) and organic matter (OM) in sediment and suspended matter of the semi-enclosed Kaštela Bay (middle Adriatic Sea) were investigated at four stations during a one year period (April 2000 - April 2001). Samples of surface sediment and suspended matter were collected monthly. One sediment core was sampled at every station at the beginning of the sampling period. Cd concentrations in surface sediment and suspended matter were in the range of 0.082-0.904 mg kg^{-1} and 0.131-1.057 mg kg^{-1}, respectively. The distribution pattern of Cd between sediment and suspended matter clarified Cd transport throughout the bay. The suspended matter acts as a collector and carrier of Cd in the bay. The higher concentrations in suspended matter were associated with lower concentrations in the sediments, which indicated that sediment resuspension has an important role in the distribution of Cd between sediments and suspended matter. The spatial distribution of Cd in the sediment and suspended matter of Kaštela Bay was in accordance with the prevailing circulation in the bay and indicated a partially anthropogenic origin of Cd. The vertical distribution revealed a decrease of Cd concentration with sediment depth, which also indicated an anthropogenic origin. Seasonal differences in the accumulation of Cd were recorded with concentrations being higher during the autumn-winter season, in accordance with a higher degree of sediment resuspension during this period.

Key words: cadmium, suspended matter, sediment, Adriatic Sea

INTRODUCTION

In coastal areas, due to the settling of suspended matter of different origins, elevated amounts of trace metals are accumulated in sediment. In this way sediment becomes a repository compartment for continuing inputs, with the capability of releasing accumulated material to the overlaying water, even after anthropogenic inputs have been reduced or eliminated (BOGNER *et al.*, 2004; CAETANO *et al.*, 2002; PALANQUES *et al.*, 1995). Suspended matter which settled on sediment might be buried, then dissolved in sediment porewaters, or turned back to the water phase by the physical resuspension transport system. Resuspension occurs when shear stress (friction of the water against the bottom) is high enough to move the particles from the sediment. This leads to a change in ecological balance in the water column, especially at the sediment-water interface where water is stagnant and forms a diffusive water layer through which the diffusion process acts. Resuspended particles, modified or unmodified, can return to the sediment surface or can be transported along the sea floor by currents away from the point of pollution. The quantity of resuspended material depends of several factors such as erodibility and shear velocity (WAINRIGHT, 1990). Resuspension can be caused by natural events such as tidal currents, biological activity, storms, high bottom currents, or by anthropogenic perturbations such as trawling, dredging and marine traffic (ALLER & ALLER, 1998).

Trace metals distribution in the marine ecosystem includes partitioning between particulate and dissolved phases, as observed by different models of the exchange process. Dissolved and sediment-associated metals can be important sources of metal exposure for the biota. The relative importance of these sources strongly depends on the metal properties and on exposure conditions including the instability of the metals in the solid phase (FERRER *et al*., 2000).

During the twentieth century, Kaštela Bay became one of the most industrialized and contaminated areas in the eastern Adriatic, Croatia (KUŠPILIĆ *et al.,* 2009). The situation has improved from 1991 onwards due to the fact that most chemical industries were closed down during the war in Croatia (UJEVIĆ *et al*., 2000; BOGNER *et al.,* 1998). Transport of sediment and suspended particles in Kaštela Bay is controlled by the prevailing circulation in the bay which is mostly induced by wind (BEG PAKLAR *et al*., 2002). Possible sources of the suspended matter in the bay are river input (Jadro river), coastal erosion, marine organisms production and particles of anthropogenic origin. The aim of this work was to clarify the influence of suspended matter on the spatial and temporal distribution of Cd and organic matter content in the sediment of Kaštela Bay.

MATERIAL AND METHODS

Study area

Kaštela Bay is the largest bay in the central part of the Croatian coast (Fig. 1). It is located

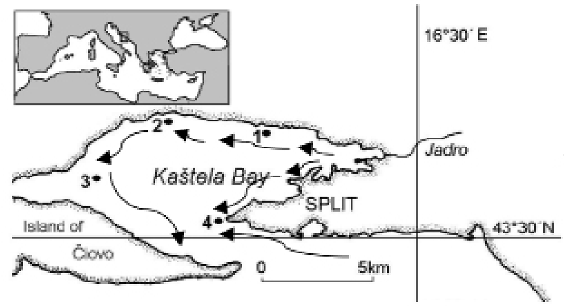

Fig. 1. Locations of experimental stations in Kaštela Bay (Adriatic Sea, Croatia)

in a highly urbanized (350.000 inhabitants) industrial and, agricultural region, characterized, by substantial discharges of domestic sewage, agricultural runoff and industrial wastes (from brewery, cement production, ports, recreational boating capacities, shipyard, iron-refining plant, etc.) which enter the bay without any treatment (UJEVIĆ *et al.,* 2000). An additional significant source of pollution is heavy traffic on the main road, which is located near the coast. Most of the industry is located in the eastern part of the bay, which is poorly aerated, particularly in the hot period of year, and very is thus susceptible to the impact of discharges of domestic and industrial waste waters and terrigeneous material brought by the River Jadro. Cd concentrations in marl deposits and carbonate rocks in the surroundings of the eastern Kaštela Bay (20 km from the bay) range from 0.380 to 1.088 mg kg^{-1} and from 0.654 to 1.346 mg kg^{-1}, respectively (BOGNER *et al.,* 1997), which indicates natural enrichment of Cd in the calcite lattice in the surrounding area.

Transport of sediment and suspended particles in the bay is controlled by the prevailing circulation which is mostly induced by local winds (BEG PAKLAR *et al.,* 2002). SE wind (scirocco) induces typical estuarine circulation with incoming flow in the surface layer and the outgoing flow in the deeper layers with turbulent vertical exchanges. NE wind (bora) is associated with an anticyclonic gyre in the eastern part of the basin and cyclonic circulation in the western part of basin, while water outflow in the surface layer is compensated for by an upwelling process. During calm weather the bay behaves as a typical dilution basin with an outgoing current in the surface layer and an incoming current in the deeper layers (ZORE-ARMANDA *et al.,* 1974). Vertical stratification is present from April to September, while in winter (from November to March) the water column is well mixed (ZORE-ARMANDA, 1980).

Sampling and analytical determination

In order to determine the spatial distribution of Cd in the bay 4 sampling sites were selected to represent a wide range of environmental conditions throughout the bay (Fig. 1). The water depth at all stations was 10 m, and the distance from the coast varied between 15 and 30 m. Sediment cores of different length (7-16 cm long) for investigation of vertical Cd distribution were sampled once, in March 2001, and sliced horizontally into 1 cm long sections. Surface sediments were sampled monthly, from March 2001 to March 2002, using a gravity corer and frozen until analysis. Samples of suspended matter were also collected monthly using sediment traps placed at three different depths at each station, except for S1 where five different depths were selected (Table 1, Fig. 2). Sediments traps were made from PVC tubes with a height: diameter ratio of 5:1; 50 cm height and 10 cm diameter (Fig. 2). Samples of collected sediment and suspended matter were lyophilized, homogenized and dry mass was determined.

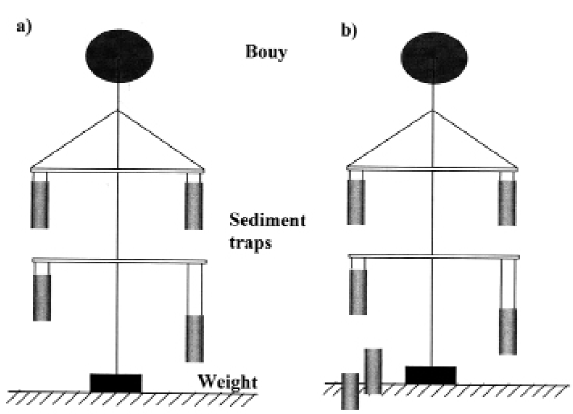

Fig. 2. Position of the sediment trap in the water column and sediment at (a) station 1 and (b) stations 2, 3 and 4 on the constructions which were built for the purpose of research

Organic matter (OM) content was determined by H_2O_2 treatment of sediment and suspended matter samples at 450°C for 6 hours. The loss of weight of the samples as the result of this treatment was assumed to be due to OM content. Granulometric composition of the sediment samples was determined by sieving (>63 μm) and hydrometric method (<63 μm). Sediment type was determined according to the Shepard classification (SHEPARD, 1954).

Samples of total sediment or suspended matter (~0.1 g) for determination of Cd were wet digested with a mixture of HNO_3 (65%, s.pure), HF (40%, p.a.) and $HClO_4$ (70%, s.pure) in a Microwave Digestion Unit. The digestion procedure included digestion of samples with acid in the microwave unit and subsequent evaporation to near-dryness on a hot plate in several steps. The detailed procedure was published earlier (UJEVIĆ *et al.*, 2000). The final evaporate was diluted to 25 mL with 0.1% HNO_3. An aliquot of the final solution was analyzed on the graphite furnace of Perkin-Elmer 1100 B spectrometer. The accuracy of the method was checked by simultaneous analyses of the certified reference materials SRM 1646, SDM/2TM, MESS-1 and BCSS-1. Results are shown in Table 2.

Table 1. Sediment trap depths, measured at the entrance of the trap, amount to accumulation depth of suspended matter collection in trap

Station 1	Station 2	Station 3	Station 4
3.8 m	4 m	4 m	5.2 m
5.8 m	8 m	8.5 m	6.5 m
6.8 m	9.2 m	10 m	8 m
8.2 m			
8.8 m			

RESULTS AND DISCUSSION

Granulometric composition of sediments and classification of sediment type according to the Shepard classification showed that fine-grained sediment at S1 is significantly different from coarse-grained sediments at stations 2, 3 and 4 (Table 3). Elevated concentrations of metals can usually be found in sediments with fine

Table 2. Quality assurance with certified reference sediment (mean value and standard deviation)

Cd (µg g^{-1})	SRM 1646	SDM/2TM	MESS-1	BCSS-1
n	20	18	18	16
certified	0.36 ± 0.07	0.113 ± 0.03	0.59 ± 0.10	0.25 ± 0.04
this study	0.41 ± 0.05	0.10 ± 0.08	0.62 ± 0.15	0.28 ± 0.06

grained particles, particularly in the silt and clay fractions due to the fact that adhesion processes of dissolved and colloidal metal species on fine grained sediment are more intensive and contribute to the increased trace metal accumulation (UJEVIĆ *et al.,* 2000). The highest OM content and Cd concentration were found in sediments at S1 (Fig. 3, 4), which is in good agreement with the granulometric composition of sediment in the investigated area (Table 3). S1 is located in the eastern part of the bay where a large amount of industrial and domestic waste waters containing various harmful organic and inorganic materials were discharged (UJEVIĆ *et al.,* 2000; BOGNER *et al.,* 1998). Increased concentrations of Cd in the fine-grained fractions of sediments in this area have been determined previously (UJEVIĆ *et al.,* 2000; BOGNER *et al.,* 1998).

The vertical distribution and concentrations of metals in sediment may be controlled by numerous factors such as granulometric and mineral composition of sediments, carrier substances, sediment surface area, OM content, bioturbation, etc. Bioturbation is the combined effect of all biological activities on particles and pore water dynamics at the sediment-water interface. The agents of bioturbation are mac-

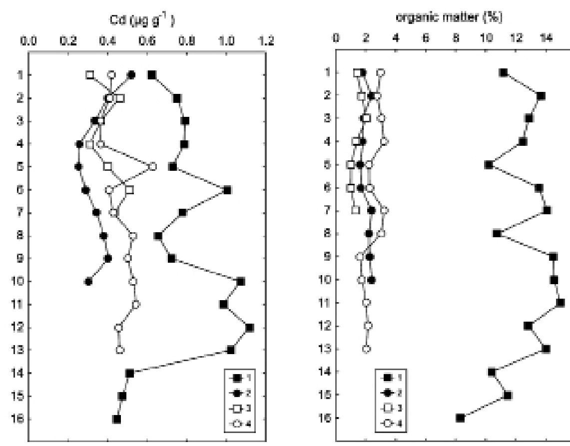

Fig. 3. Distributions of cadmium and organic matter content in sediment cores from Kaštela Bay stations

roinvertebrates such as oligohaetes, polychaetes, chironomides, crustaceans, mussels, molluscs and fish. The range of mechanisms of particle and water movement by macroorganisms is varied. In coastal sediments, deposit-feeding bivalves graze on organic detritus at the sediment surface, and worms inhabit tubes 30-40 cm below the sediment-water interface. Both species enhance particle transport through grazing and pore water transport through their respiration activities. (SANTSCHI *et al.,* 1990).

Table 3.Granulometric composition and sediment type (according to Shepard classification) of surface sediments at 4 stations in Kaštela Bay

Station	Gravel (%)	Sand (%)	Silt (%)	Clay (%)	Sediment type
1	1.8	26.8	64.6	6.8	sandy silt
2	7.3	85.2	3.8	3.7	sand
3	8.0	81.8	4.5	5.7	sand
4	16.6	75.9	3.3	4.2	sand

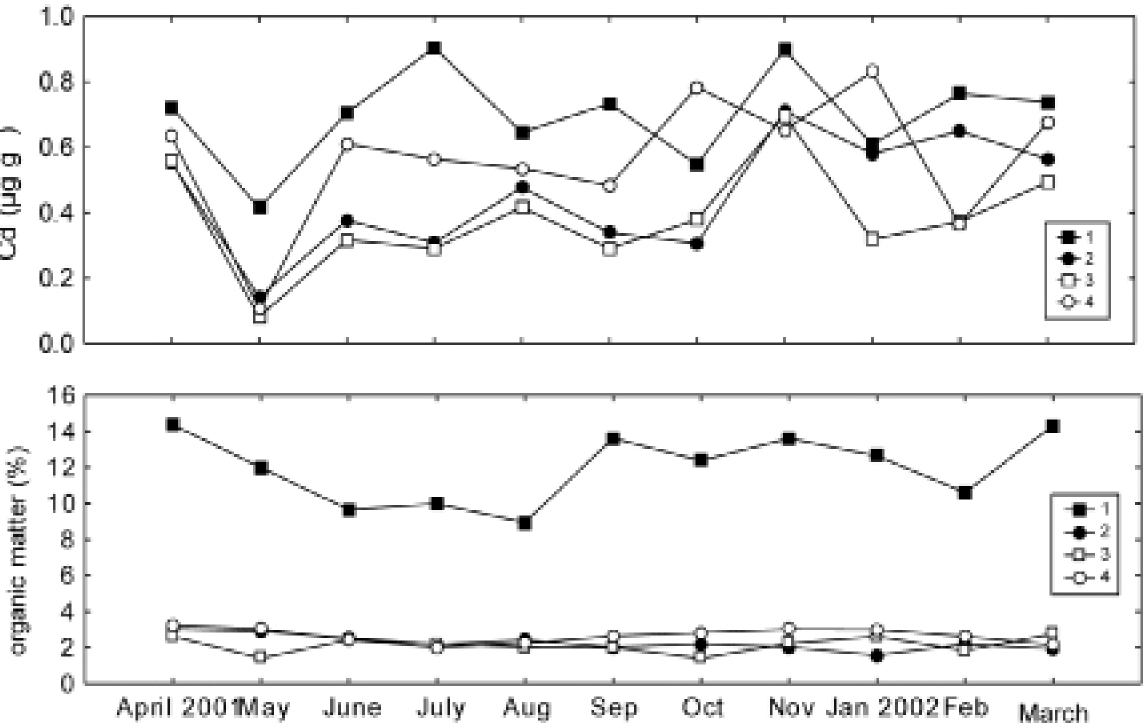

Fig. 4. Distributions of cadmium and organic matter content in surface sediment from 4 stations in Kaštela Bay

In Kaštela Bay sirocco wind induces outgoing flow in the deeper layers (BEG PAKLAR *et al.,* 2002) and can erode surface sediment and transport particles to the deeper part. The concentration of Cd in vertical profiles was significantly positively correlated with the concentration of OM only at S1 ($r=0.844$, $P<0.001$) (Fig. 3), which indicates that the interaction between OM and Cd is an important process for the removal and fixation of Cd in sediment at station S1. A strong correlation between OM content and trace metal concentration at a certain location, similar to one we have found for S1, usually indicates that the investigated area is polluted (UJEVIĆ *et al.,* 2000). A positive association between OM and Cd has often been related to the adsorption and complexation of Cd by organic matter.

A possible way to estimate anthropogenic input is to compare the concentrations of elements in upper layers of sediment with background concentrations of the same elements from deeper layers (KLJAKOVIĆ-GAŠPIĆ *et al.,* 2009). Results of the comparison of average concentrations in the mixed sediment layers (0-14 cm) with the last 3 cm of the cores showed that only at S1 Cd concentrations and OM content increase through upper layers of sediment (Fig. 3). The ratio between average Cd concentration for the upper 14 cm and the last 3 cm of the sediment core was higher than 1 (1.8) which confirms the anthropogenic influence in the eastern part of the bay. Data for stations 2, 3 and 4 did not defer between depths, probably because of the insufficient length of cores. Comparison of observed data with average values in marl (0.758 ± 0.357 mg kg^{-1}) and carbonate rocks (1.036 ± 0.352 mg kg^{-1}) from the surrounding area (BOGNER *et al.,* 1997) support our theory regarding a certain anthropogenic influence in the eastern part of the bay. Carbonate content in almost all investigated samples from the surrounding area and surface sediment from Kaštela Bay were up to 100% and 40-60%, respectively.

Concentrations of Cd and OM content in both sediment cores (Fig. 3) and surface sedia)

b)

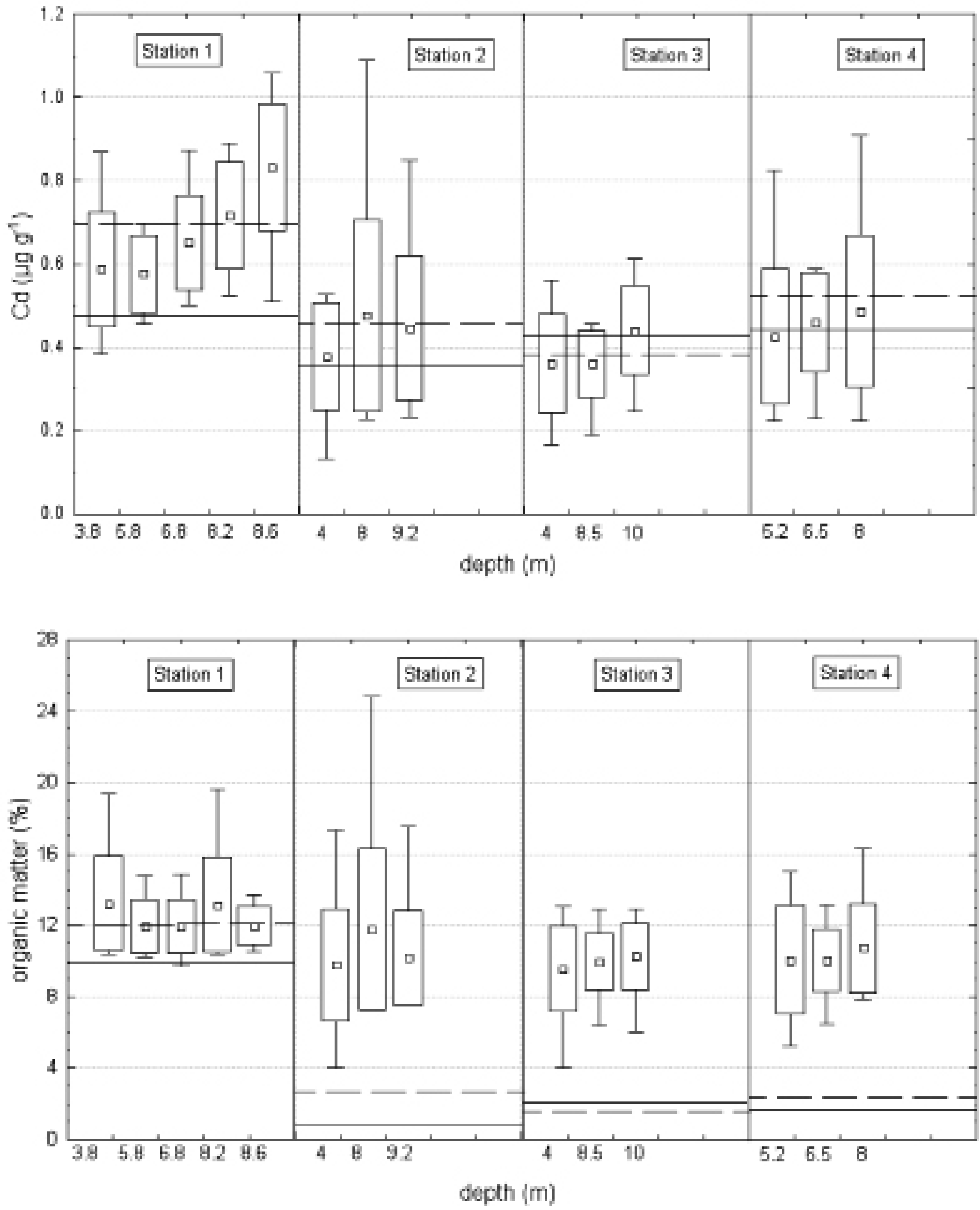

Fig. 5. Levels of (a) Cd and (b) organic matter in the suspended matter collected at defined depths of studied stations in the period from 20 March 2000 to 21March 2001. Squares (□) represent mean values, lower and upper box edges represent mean±1 SD, outlying bars are minimum and maximum values, solid horizontal line (__) represents mean Cd concentrations in the two deepest cm of the sediment cores, and dashed horizontal line (_ _ _) represents annual mean of Cd concentrations in surface sediments

ment (Fig. 4) were highest in the eastern and most polluted part of the bay (S1) and decreased with distance from it in either an anti-clockwise pattern or towards the exit of the bay (S1>S4>S3≈S2), depending on prevailing circulation. The spatial distribution indicates that the eastern part of Kaštela Bay, which is more shallow and contaminated, serves as a source of pollution for the remaining area.

Concentrations of Cd in the colder period of the year, from September to March (0.574±0.183µg g^{-1}), were higher in comparison to the late spring-summer period (0.468±0.217 µg g^{-1}) (Fig. 4). A seasonal change of OM content was recorded only at S1, where OM content increased from 11.0±2.2% in the warmer part of the year to 12.9±1.3% in the September-March period.

Research on factors which affect Cd accumulation in sediment needs to focus on a water column system, with clearly defined sources of metals and pathways of metal through the marine ecosystem. One of the most important factors influencing Cd migration in the marine water column is suspended matter.

Investigated stations are close to the coast (15–30 m), so we assumed that the lithogenic component should be dominant in suspended matter. Suspended material can be composed of particles of anthropogenic and/or natural origin, whether from the input of the local river, biological or atmospheric origin. According to BOGNER *et al.* (1997) Cd content in surrounding rocks indicate mostly their natural origin. Most particles may originate from resuspension of sediments, probably going through multiple deposition and resuspension cycles. Resuspension of sediments in coastal environments, especially during the winter has often been noted (NAMEROFF *et al.*, 2002; HEUSSNER *et al.*, 2006), and the same results were obtained in Kaštela Bay (Buljac, personal communication). Transport of resuspended fine grained particles from anoxic conditions to the oxic water column may lead to the formation of iron oxy-hydroxides which adsorb metals. As a result, metal concentrations in the water column decrease.

Figures 5a and 5b show Cd and OM distribution in the suspended matter collected at different depths. Cd concentrations in suspended matter (range: 0.131-1.057 mg kg^{-1}; mean: 0.504±0.191 mg kg^{-1}) were somewhat higher than in surface sediment (range: 0.082-1.116 mg kg^{-1}; average: 0.533±0.215 mg kg^{-1}) or the bottom 3 cm of sediment (Fig. 5a). Concentrations of Cd and OM in suspended matter were highest at S1 and decreased with distance from the eastern part of the bay (S1>S2, S4, S3).

Statistical analysis, the nonparametric Friedman ANOVA Chi Sqr. test, was conducted on Cd concentrations in the suspended matter of all stations. The differences between stations were statistically significant (χ^2=64.56, P< 0.001). Cd concentration at S1 also varied with the collection depth, but this was not the case for the OM. Cd concentrations increased from the surface (0.592 mg kg^{-1} at 3.8 m) to the deeper water layers (0.857 mg kg^{-1} at 8.6 m). Higher Cd concentrations in the suspended matter of deeper water layers indicated a significant input of Cd in the marine ecosystem (S1 is located at the most polluted part of the bay) and a more intensive resuspension process of fine grained sediment. Practically, particles of suspended matter can act as a collector for dissolved Cd from the water column.

OM content in collected suspended matter (2.5–14.7%) showed a uniform distribution with depth at all stations (Fig. 5b). OM content in suspended matter at S1 was similar to the OM content of sediment, while at remaining stations OM content in suspended matter was significantly higher than in the deepest layers of sediment or in surface sediment (Fig. 5b). Differences in OM content between S1 and other stations are a consequence of the considerably more intensive input of suspended particles either from natural or anthropogenic sources and more intensive precipitation in the eastern part of the bay where S1 is located.

CONCLUSIONS

The main source of Cd and organic matter in the sediment of Kaštela Bay is suspended matter from its own eastern part, which is the most contaminated part of the bay. Cd concentration decreased with distance from it in either an anti-clockwise pattern or towards the exit of the bay, depending on prevailing circulation. Cd concentrations and OM content are higher in suspended matter than in sediments. The spatial distribution of Cd and OM is affected by settling suspended matter as well as resuspension of sediments. Seasonal changes of Cd concentrations were recorded at all stations. Concentrations were higher during the winter season as the result of an increased resuspension of the sediment as well as increased inputs from the land and the atmosphere.

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Utjecaj suspendirane tvari na akumulaciju kadmija u sediment Kaštelanskog zaljeva, Jadransko more, Hrvatska

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SAŽETAK

U poluzatvorenom Kaštelanskom zaljevu istraživane su prostorna i sezonska raspodjela kadmija i ogranske tvari u sedimentu i suspendiranoj tvari tijekom jedne godine (travanj 2000. – travanj 2001.). Uzorci sedimenta i suspendirane tvari sakupljani su mjesečno na četiri postaje koje su 15-30 m udaljene od obale. Na početku istraživanja na svakoj je postaji uzorkovana po jedna sedimentna jezgra. Granulometrijski sastav sedimenta u istočnom onačišćenom području je pjeskoviti silt, dok je u ostalom dijelu zaljeva pjesak. Izmjereni maseni udjeli kadmija u sedimentu bili su od 0,082 do 0,904 mg kg^{-1}, a u suspendiranoj tvari od 0,131 do 1,057 mg kg^{-1}. Raspodjela kadmija između sedimenta i suspendirane tvari utječe na prijenos kadmija kroz zaljev. Suspendirana tvar je „sakupljač“ i „nositelj“ kadmija u zaljevu. Resuspenzija sedimenta je značajan proces koji doprinosi povećavanju masenih udjela kadmija u suspendiranoj tvari i smanjenju u sedimentu. Prostorna raspodjela kadmija određena je cirkulacijom u zaljevu i ukazuje na antropogeno podrijetlo kadmija. Smanjenje masenih udjela kadmija u dubljim slojevima sedimenta također upućuje na antropogeno podrijetlo kadmija u zaljevu. Povećanje masenih udjela kadmija zabilježeno je tijekom jesenske i zimske sezone kada je proces resuspenzije sedimenta značajniji.

Ključne riječi: kadmij, suspendirana tvar, sediment, Jadransko more