



The impact of highway pollution on the coastal sea

J. Faganeli*, B. Vrizer*, H. Leskovsek**, B. Cermelj*
& R. Planinc*

**Marine Biological Station, Fornace 41, 6330 Piran, Slovenia*

***Jozef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia*

Abstract

The impact of motorway pollution on the coastal sea in the Bay of Koper (Gulf of Trieste, northern Adriatic) was studied by the analysis of polycyclic aromatic hydrocarbons (PAH) and heavy metals (Pb, Zn, Ni, Mn, Cu, Cr, Cd) in marine sediments, and selected benthic organisms, namely, the bivalve *Mytilus galloprovincialis*, the gastropod *Patella coerulea* and seagrass *Cymodocea nodosa*. The impact of motorway pollution was evident in higher contents of PAH and heavy metals in bivalves and gastropods collected in the mediolitoral because the contaminants are prevalently bound to particles and accumulated by organisms. The accumulation of pollutants in the sediments of the Bay of Koper is less significant due to the sedimentological properties (mostly coarse sediment) of the nearshore zone of the Bay, and advective transport of particles. The sedimentological properties also govern the distribution and preservation of PAH in the study area since the degradation of PAH is slower in pelitic sediments at the Bay entrance and in the central part of the Gulf of Trieste. These are also the principal sites of pollutant accumulation. The described contamination of benthic organisms was, however, not reflected on the benthic community level which does not show any sign of degradation and any difference from the apparently uncontaminated northern shoreline of the Bay.

1 Introduction

Since most of the industrial and domestic emissions have now been reduced, traffic pollution represents the major source of pollution in urbanized areas¹. Besides air pollution which is the most recognized traffic-related problem, more recently vehicle emissions have been implicated in various health effects, noise, acid deposition, photochemical smog, global climatic changes, soiling of surfaces and implications for ecosystems in general. Only recently has motorway pollution been recognized as an important non-point source of pollution for aquatic environments due to pollutant input from the motorway surface and the modification of local hydrology imposed by the design and construction of the



motorway. Studies of the impact of motorway loading on aquatic systems are rather rare^{2,3,4}, but studies of these pollution effects on the coastal sea are, to our knowledge, at present nonexistent.

The aim of the present work was to study the long-term effect of highway pollution on coastal marine environment in the Bay of Koper (Gulf of Trieste, northern Adriatic) using chemical analyses of the traffic pollutants polycyclic aromatic hydrocarbons (PAH) and selected heavy metals (Pb, Zn, Cr, Cd, Ni, Mn, Cu) in surficial marine sediments and representative benthic organisms, namely a mussel, a gastropod and seagrass. The $\delta^{13}\text{C}$ values of sedimentary organic C were used to decipher the origin of organic matter in the studied sediments. In parallel, the impact of traffic pollutants on the littoral benthic community was studied using faunistic and floristic analyses. The results obtained were compared to those from an apparently uncontaminated nearshore area of the Bay.

2 Materials and methods

2.1 Study area

This study was conducted in the Bay of Koper (Gulf of Trieste, northern Adriatic) along the motorway between Koper and Izola (Figure 1). The 5 km long motorway was constructed in 1936 and has at present (1994) a traffic density of about 657000 vehicles annually of which about 10% have diesel engines. The estimated mean velocity of vehicles is about 75 km/h. The estimated annually average exhaust emission of pollutants from one vehicle is 0.8 g CO/km, 1.0 g hydrocarbons (HC)/km, 1.8 g NO_x/km, 0.22 g Pb/km, 1.5 g SO₂/km and 0.6 g of particles/km at a fuel consumption of 40 g/km⁵. The drainage system of the road consists of gulleys discharging the runoff water approximately every 50 m directly into the coastal sea. Location A was situated in a small relatively closed harbour.

2.2 Samples

Runoff water from a motorway gully and diluted runoff water with seawater were collected on June, 3 1993 in the morning during a storm event after a longer dry period using, polyethylene bottles.

Surficial sediment samples (2 cm top layer) and organisms were collected in March 1993 and March 1994 at the locations depicted in Figure 1. by SCUBA divers inserting plexiglass tubes (6 cm i.d.) into the sediment. For PAH and metal analyses the wet sediment samples were sieved so that up to 90% of the material passed through a 500 μm mesh sieve. For $\delta^{13}\text{C}$ analysis of sedimentary organic matter the sediment samples were freeze-dried, homogenized and sieved so that up to 90% of the material passed through a 100 μm sieve.

Animals and plants collected by hand were washed with distilled water, freeze-dried and ground to a fine powder.

Fauna was sampled in March 1994 at 5 locations along the motorway (locations C, G, B, A and F), and at 2 locations (J and L) along the northern shoreline of the Bay of Koper (Figure 1), apparently unpolluted by traffic.

2.3 Chemical analyses

Wet sediment and biological samples for PAH analysis were first spiked with internal standards and then extracted with CO₂ and 5% methanol with addition of Cu powder and wetsupport at a temperature of 100 °C for 1 hour using supercritical fluid extraction (Isco SFE System 2100). The extracted components were collected in dichloromethane solvent trap and determined by GC-MS (AutospecQ, VG Analytical) using a fused silica capillary column coated with SE-30 (Supelco). The quantification was based on calibration with internal standards SRM 1647c and SRM 1597 (NIST, USA; Supelco 4-8905, USA) corrected for extraction yields (30-60% for individual compounds).

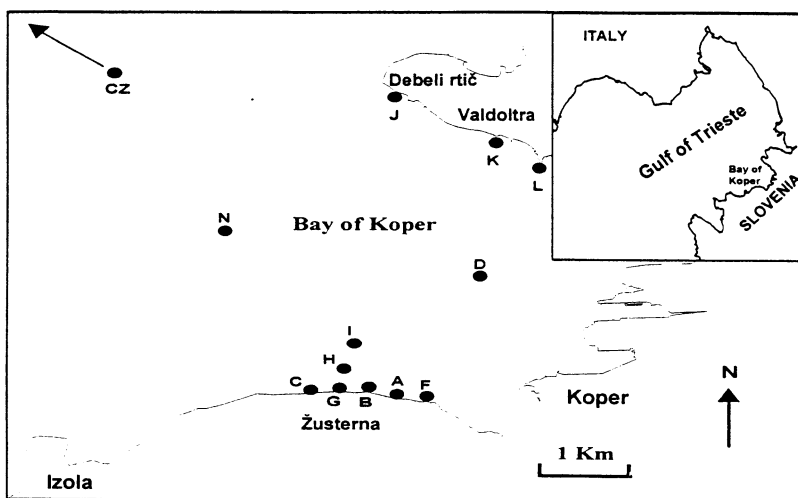


Figure 1: Sampling locations for analyses of sediments and organisms, and faunistic analyses in the Gulf of Trieste (northern Adriatic)

The PAH analyzed were fluorene (Fl), phenanthrene (Phe), anthracene (An), fluoranthene (Flu), pyrene (Py), chrysene (Chr), benzo(a)anthracene (B(a)An), benzo(a)pyrene (B(a)Py), acenaphthene (Ace), acenaphthalene (Aci). The analytical reproducibility for duplicate samples was 15%. The limit of detection for individual compounds was <0.1 ng/g.

Total hydrocarbons (HC) in the runoff water were extracted with hexane, analyzed fluorimetrically and the data reported as chrysene equivalents^{5, 19}.

The runoff water, wet sediment and dry biological samples for metal analyses were transferred to PTFE decomposition vessels and boiled with conc. HNO₃ at 120° C for 4 hours. Analysis of Pb, Ni, Cu, Cr, Cd were carried out by ETAAS, and Mn and Zn were analyzed by flame AAS using a Varian model SpectrAA 10 BQ atomic absorption spectrophotometer.

Statistical evaluation⁶ of the sediment PAH and metal data was performed using the Statgraphics (Stsc. Inc., USA) statistical programme.

Dry sediment samples for determination of the stable isotope composition of organic C were first acid treated with 3M HCl and then ignited to CO₂ in a recirculating stream of O₂. The stable isotopic composition of C was determined using a Europa 20-20 mass spectrometer and the results reported as deviations in ‰ from the ¹³C/¹²C ratio of the Chicago PDB standard ($\delta^{13}\text{C}$).

2.4 Biological analyses

Benthic fauna was sampled in the mediolittoral community which covers the limestone breakwater rocks between high and low tide. The faunal material was collected (scraped) from 50x50 cm squares, then taxonomically identified and counted. Only the most abundant species were estimated.

3 Results and discussion

3.1 Pollutant loading

The runoff of pollutants from the Koper-Izola motorway is mostly dependent on the seasonal variations of meteorological conditions. The runoff is especially intensive after spring and autumn rainfall after longer dry periods. The concentration of heavy metals in runoff water (Table 1) are similar to those reported by other researchers worldwide¹. The analysis of heavy metals and total HC in the diluted runoff water in seawater (Table 1) showed decreased concentrations of Pb, Zn, Mn, Cd, Cu, Ni and Cr relative to concentrations in the motorway runoff water. The greatest decrease observed for Pb is due to the prevalently bonding of Pb in insoluble particulate matter, while other metals are divided approximately equally between particulate and dissolved fractions, or are mostly present in the dissolved fraction¹. Also, the concentration of total HC were about 5-fold higher in the runoff than in runoff water diluted by seawater (Table 1).

Table 1: Heavy metals and total hydrocarbons (Tot. HC) in runoff water in gully (1) and diluted in seawater (2) during the storm event on June, 3 1993 in the morning ($\mu\text{g/l}$)

Sample	Pb	Zn	Cd	Cr	Cu	Mn	Ni	Tot. HC
(1)	1021	3413	3.3	42	70	318	18	115
(2)	15	485	0.3	26	12	8	7	20

A tentative budget for some traffic pollutants entering the coastal sea could be approximately constructed considering our emission data and the average pollutant loading from four USA highways² with traffic density comparable to our case. The use of US data is also possible because the analysis of traffic pollutants in the runoff water is similar to that obtained from US motorways. Considering these data and the mean precipitation rate of rainwater in the Koper area (approximately 1000 mm yr⁻¹) we can deduce that the mean annual loading amounts to 12 kg of Pb, 13 kg of Zn and 3500 kg of HC per km of motorway. Considering further the annual emission of Pb and that about 50% of particulate matter is composed of HC² we can deduce that less than 10% of the annually emitted Pb and particulate HC enter the coastal sea by the motorway runoff. This percentage is probably lower because of physical, chemical and biological processes (adsorption, sedimentation, uptake by plants) operative in the road gulleys².

3.2 PAH in sediments

In all recent sediment samples unsubstituted PAH with four and five condensed rings were present (Table 2) which are typical of a pyrogenic origin of PAH⁷ comprising the emissions (pollution) of motor vehicles. Pyrogenic PAH are prevalently introduced into the coastal marine environment by runoff as indicated by the difference in the PAH component ratio between recent marine sediments and a soil sample collected at location E in the vicinity of motorway (Table 2). The origin of PAH in this location is most probably atmospheric deposit. The pyrogenic origin of PAH in sediments is evident from the ratios of PAH components represented in the plot of the Phe/An and Py/B(a)Py ratios (Figure 2). Only in the samples collected in locations A, F and I could the PAH sources be partially attributed to oil (petrogenic) pollution.

Table 2: PAH concentrations in sediment samples (ng/g; dry weight)

Loca tion	Fl	Phe	An	Flu	Py	B(a)An	B(a)Py	Ace	Ac1	Total	$\delta^{13}\text{C}(\text{‰})$
F	221	10	275	183	184	230	151	10	259	1502	
A	600	500	300	600	1600	20	100			3720	-19.93
G	69	55	139	66	115	112	273	74	100	1005	-20.48
B	200	400	600	2300	1400	600	500			6000	-18.56
C	20	30	80	200	100	80	60			570	-17.93
H	218	176	230	178	56	10	158	10	408	1424	-18.09
I	284	397	33	35	201	35	10	10	281	1265	-21.47
J	10	70	79	10	75	288	421	10	299	1231	-22.91
K	206	235	232	280	291	10	106	10	160	1510	-21.98
L	10	296	167	157	86	10	104	10	305	1115	-19.86
D	30	50	60	80	50	10	80			360	-21.00
N	273	250	342	171	768	99	995	434	347	3679	-21.61
CZ	441	115	329	305	348	170	548	322	365	2943	-21.69
E	10	10	50	80	20	10	10			150	

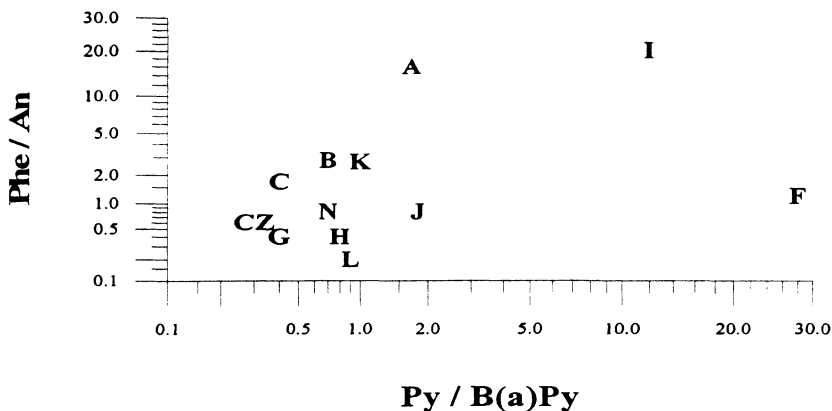


Figure 2: Relationship between Phe/An and By/B(a)Py ratios in sediment samples

The lowest PAH contents were detected in the reference locations I, J, K along the apparently unpolluted northern shore of the Bay of Koper (Figure 1). Elevated PAH contents along the Koper-Izola motorway were noticed only in locations A and B situated in a relatively closed small harbour, where the motorway runoff is directly discharged into the sea, and between two discharges from motorway gulleys, respectively. The highest PAH contents were found in locations N and CZ, situated at the Bay entrance and in the central part of the Gulf of Trieste, respectively (Table 2).

This areal distribution is further confirmed by cluster analysis of PAH showing similarities between all locations along the motorway and on the reference northern shore (locations L, K and J), except locations A and B situated in the small harbour and in the vicinity of the gully outlet, respectively. Similarities also appeared between locations N and CZ situated in the central part of the Gulf of Trieste. R-factor analysis of seven variables showed that the first three factors comprised 86.1% of the variance. The first factor (comprising 51.2% of the variance), related to Fl, Phe and Py, influences locations A and B, the second (comprising 24.3% of the variance) related to Flu, An and B(a)Py influences locations G and L, and the third factor (comprising 10.6% of the variance) related to B(a)An influences locations H, J and D. We concluded, therefore, that the influence of motorway discharges is evident at locations A and B situated in the small relatively closed harbour and in the vicinity of discharge, respectively, but other locations are less affected because the smaller particles with PAH are diluted, transported away, and mostly deposited in deeper parts of the Gulf.

PAH contents were higher in offshore (>10 m of depth), mostly pelitic sediments, compared to nearshore prevalently coarse sediment samples⁸. Similar areal distribution was observed for sedimentary organic C contents⁹. In nearshore sediments the sedimentary organic matter has an important imprint from benthic



macrophytes - algae and seagrasses¹⁰, as indicated by the rather high $\delta^{13}\text{C}$ values ranging between -18 and -20‰ (Table 2). In offshore sediments the active deposition of fine-grained sediment occurs⁸ (approx. 1.4 mm yr⁻¹), and the sedimentary organic matter, according to its $\delta^{13}\text{C}$ values ranging between -21 to -22‰ (Table 2), is of planktonic and microphytobenthic origin⁹. The described areal distribution of PAH contents is, therefore, most probably the result of transport processes of particles on which PAH are associated, and sedimentation and sorptive preservation on mineral surfaces in mostly pelitic sediments¹¹. The role of atmospheric deposition is less important, as indicated by the rather low PAH contents present in the soil sample in location E. Also, probably less important are other factors which could influence PAH degradation - chemical structure, bottom water oxygen concentration (anoxia), temperature and pH, and previous exposure to the microbial community¹², although periodic hypoxias and anoxias are known in late summer in the bottom water layer (below the pycnocline) in the central part of the Gulf of Trieste.

Our PAH results are higher than those reported for sediments of the central and northern part of the Gulf of Trieste¹³, probably because of different sampling techniques used (corer vs. grab), and higher than those found in the NW Adriatic^{13,14}. Both were described as the most polluted areas in the Adriatic Sea due to oil spills from tankers and the River Po outflow, respectively. Comparing our PAH results with others from polluted marine sediments in the Mediterranean^{15,16} and worldwide¹⁷ it emerges that ours are high and, therefore, the situation merits further investigation.

3.3 Heavy metals in sediments

The analysis of heavy metals in surficial nearshore sediments (Table 3) showed somewhat higher contents of Cr, Mn, Ni, Zn and Pb in comparison to those from the central part of the Gulf of Trieste and the "geochemical background" decoded at the depth of 1m in the nearby borehole MK-6 dated to 200 years BP⁹. Pb, Ni and Zn are important motorway pollutants originating from leaded fuel, lubricants and tyres, respectively². The rather small increase in concentrations is due to the rather low accumulation of heavy metals in coarse nearshore sediments⁹, influenced by tides, storms and sediment resuspension.

Cluster analysis showed that the locations at the Bay entrance and along the southern shore along the motorway are similar, and the locations situated along the northern shoreline and CZ located in the central part of the Gulf of Trieste show similarities. R-factor analysis of seven variables (Pb, Zn, Ni, Mn, Cu, Cr, Cd) showed that the first four factors comprised 94.9% of the variance. The first factor (comprising 41.9% of the variance), relating Ni, Mn and Cr, influences the locations G, I and D, the second factor (comprising 33.1% of the variance), relating Cu and Cd, influences locations L, K, J, the third factor (comprising 10.4% of the variance), relating Pb, influences location B, and the fourth factor (comprising 9.5% of the variance), relating Zn, influences locations A, B and D.

Locations A, B, G and I along the motorway were, therefore, affected by traffic runoff.

Table 3: Heavy metals in sediment samples compared with data from a horizon dated to 200 BP in the borehole MK-6⁹, and with the mean (\pm SD) concentration in surficial sediments of the Gulf of Trieste (TS)⁹ ($\mu\text{g/g}$; dry weight)

Metal	A	G	B	C	H	I	J,K,L	D	N	CZ	MK6	TS
Pb	57.4	44.3	262	28.8	69.3	53.6	122	19.5	75.9	58.9	9.1	18.9 \pm 5.9
Zn	194	102	162	124	86	94	109	139	113	114	91	102 \pm 3.
Ni	28.6	108	30.5	26.5	52	99.5	42.7	127	73.7	73.4	248	101 \pm 4
Mn	365	945	494	206	561	853	713	746	778	688	77	593 \pm 02
Cu	55.8	45.9	28.8	22	33	50.2	76.3	33.6	34.9	35.5	30	20.8 \pm 0.2
Cr	30.7	60	27.9	25.6	36.2	58.3	25.3	64.3	51.4	51.4	17.7	59.4 \pm 3.4
Cd	0.11	0.09	0.11	0.09	0.09	0.07	1.2	0.21	0.07	0.9	0.2	0.23 \pm 2

3.4 PAH and heavy metals in benthic organisms

In *Mytilus galloprovincialis* and *Patella coerulea* higher concentrations of An, B(a)An, Py and B(a)Py were found, i.e. the compounds with three- and four rings with lower molecular weight. The PAH contents in gastropods and bivalves were higher in samples collected along the motorway than in the reference area along the northern shoreline of the Bay (Table 4). This difference can be attributed to the impact of particulate pollutants originating from the motorway run-off and subsequently absorbed by both organisms. Bivalves are known to be useful organisms for monitoring HC pollution because they contain low or undetectable enzyme activity capable of metabolizing PAH¹⁸ and, hence, they bioaccumulate these compounds without alteration.

Table 4: PAH concentrations in *Mytilus galloprovincialis* in locations F and L and in *Patella coerulea* in locations G, F, L and J (ng/g; wet weight).

Mytilus galloprovincialis

PAH	Locations	
	F	L
Fl	95	126
Phe	178	333
An	253	153
Flu	229	10
Py	230	291
B(a)An	444	113
Chr	10	126
B(a)Py	149	115
Total	1588	1267

Patella coerulea

PAH	Locations				
	G	F	L	J	
Aci	54	14	10	10	
Fl	10	10	10	10	
Phe	31	10	11	10	
An	2062	788	138	153	
Flu	34	522	10	33	
Py	10	1905	108	29	
B(a)An	180	10	16	27	
B(a)Py	10	1926	39	10	
Total	2361	5125	301	242	

The analyses of heavy metals in bivalves and gastropods also showed higher concentrations of Pb, Ni, Cr and Cd in the samples collected along the motorway



(Tables 5 and 6) and can be attributed to the impact of motorway particulate pollutants. This is confirmed by the analysis of heavy metals in the seagrass *Cymodocea nodosa*, showing even higher concentrations in an apparently unpolluted location (Table 7). This plant does not accumulate particulate metals and is, therefore, not useful as an indicator organism for prevalently particulate pollutants in the marine environment.

Table 5: Heavy metal concentrations in *Patella coerulea* in locations G and J ($\mu\text{g/g}$; dry weight)

	Pb	Zn	Ni	Mn	Cd	Cr	Cu
G	8.51	69.49	5.25	7.66	8.51	6.08	14.78
J	4.47	56.30	3.50	8.72	4.34	1.68	11.88

Table 6: Heavy metal concentrations in *Mytilus galloprovincialis* ($\mu\text{g/g}$; dry weight; $\pm\%$ RSD, $n=10$) in locations F and L compared to mean concentrations from the southern part of the Gulf of Trieste (TS)¹⁸

	Pb	Zn	Ni	Mn	Cd	Cr	Cu
F	9.34 \pm 16	223 \pm 24	3.86 \pm 12	13.6 \pm 13	3.76 \pm 13	4.83 \pm 12	8.75 \pm 7
L	5.62 \pm 8	267 \pm 28	2.54 \pm 4	17 \pm 18	2.02 \pm 12	2.49 \pm 23	13.4 \pm 12
TS	6.04 \pm 41	177 \pm 35	2.96 \pm 2	16.6 \pm 23	3.1 \pm 27	1.31 \pm 19	9.95 \pm 25

Table 7: Heavy metal concentrations in *Cymodocea nodosa* ($\mu\text{g/g}$; dry weight) in locations G and J

	Pb	Zn	Ni	Mn	Cd	Cr	Cu
G	5.49	49.13	4.58	80.30	1.21	0.80	11.82
J	29.13	787.50	8.10	64.46	2.38	2.15	13.57

3.5 Effect of motorway discharges on mediolittoral fauna

The infralittoral macrobenthos (depth interval 0-10m) along the coastal motorway (Figure 1) consists of two distinct, partially mixed communities: a hard bottom phytal community with a variety of algae (mostly genus *Cystoseira*) and small areas of sandy bottom sea grass meadows with predominantly *Posidonia oceanica*, *Zostera marina* and *Cymodocea nodosa*. No destructive evidence of traffic impact was found in these well preserved highly diverse communities, hence, we focused our attention on the mediolittoral coastal fauna, between high and low tide, which was nearest to traffic pollution. In total 24 macrofaunal species were found there with small differences between sampling locations, varying from 9 to 13 species per location. The most abundant species were among Bivalvia (*Mytilus galloprovincialis*, *Mytilaster minimus*, *Crassostrea gigas*), Gastropoda (*Littorina neritoides*, *Patella coerulea*, *Monodonta articulata*), and Crustacea (*Chthamalus stellatus*).

On the basis of the presence of species, the Sorensen²⁰ similarity coefficient (QS) was determined for all possible combinations of sampling locations (Figure 3). Considering the 21 location pairs, the QS exceeded 50% at 19 locations, representing a high similarity level. There were some small quantitative differences between dominant species and between some locations, but without any evidence of traffic runoff impact. In our case the similarity analysis showed no taxonomic or structural differences between the motorway polluted and apparently unpolluted communities. This is in clear contrast to studies of freshwater benthos (especially along river flows), where a reduction of the number of species and total biomass of benthic invertebrates was documented due to motorway pollution^{21, 22}. However, motorway runoff impacts are often masked by rural, industrial and domestic outfall impacts. All this, taken with ecological peculiarities, make comparison between polluted and unpolluted areas difficult⁴.

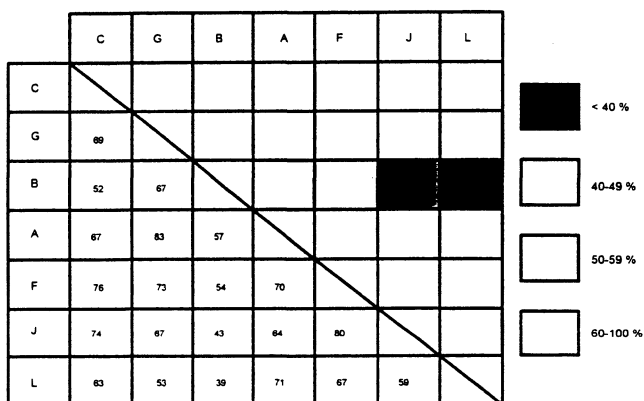


Figure 3: QS diagram of faunal affinity (% of species shared) among the mediolittoral assemblages at each location (C, G, B, A, F along the motorway, and J and L in an apparently unpolluted environment)

4 Conclusions

1. The Koper-Izola motorway, characterized by a rather high and constant traffic density mostly (>95%) composed of passenger cars, pollutes the environment with 52 tons of CO, 6.5 tons of HC, 12 tons of NO_x and 145 kg of Pb annually per kilometre. The annual loading of pollutants from each km of motorway averages 12 kg of Pb, 13 kg of Zn and 3500 kg of particulate HC. Considering the mean annual emission of Pb and particulate HC from vehicles, and the mean precipitation in the Koper area it emerges that <10% of emitted Pb and HC enter the coastal sea.

2. The analysis of PAH in nearshore surficial marine sediments in the Bay of Koper and offshore in the Gulf of Trieste revealed that PAH are mostly of



pyrogenic origin. The highest contents observed in the offshore locations demonstrate that PAH are introduced into the coastal sea mostly bound to finer particulate matter, transported away by tides and currents and sedimented in an offshore area composed of pelitic sediments. These sediment characteristics govern the distribution of sedimentary PAH through their persistence and by sorbing PAH on fine particles. The input of prevalently bonded PAH onto particulate matter is reflected in the higher PAH contents observed in the filter-feeder organism *Mytilus galloprovincialis* and *Patella coerulea* collected along the motorway in comparison to those collected in the apparently traffic unpolluted area along the northern shoreline of the Bay of Koper.

3. The analysis of the heavy metals Pb, Zn, Cr, Mn, Ni, Cd, Cu in surficial sediments along the Koper-Izola motorway showed only moderately increased concentrations of Pb, Cr, Mn, Ni and Zn in comparison to the surficial sediments in the southern part of the Gulf of Trieste, and to the "natural geochemical background" determined in a sediment horizon dated to 200 BP. This is probably due to the low accumulation of heavy metals, prevalently bonded onto particulate matter, in coarse nearshore sediments affected by tides, storms and resuspension. The significantly increased concentration of Pb, Ni, Cd and Cr observed in *Mytilus galloprovincialis* and *Patella coerulea* along the Koper-Izola motorway in comparison to an apparently unpolluted area along the northern shoreline in the Bay of Koper confirms the pollution role of particulate heavy metals originating from the motorway runoff.

4. A faunistic analysis, based on the abundance of species, and especially the calculated similarity index, showed that despite the described contamination of benthic organisms along the motorway this community biocenologically does not differ from that apparently uncontaminated by traffic pollutants along the northern shoreline of the Bay of Koper.

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