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Contamination of Runoff Waters from Roads with High Traffic Intensity in the Urban Region of Gdańsk, Poland

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Abstract

Our paper presents the results of determination of road runoff water pollutants. Samples of runoff waters were collected at six sites with high traffic intensity located along a major transportation route from the city of Reda to the city of Gdańsk, and at one site with low traffic intensity located in Reda. Sampling was carried out over a period of two months in the fall of 2000. Additional samples were collected in March 2001. The analytes determined in the samples included organochlorine, organonitrogen and organophosphorus pesticides, selected volatile organohalogen compounds, petroleum hydrocarbons, as well as selected anions and cations. Total organic carbon (TOC) was also determined. In the petroleum hydrocarbons group, the highest concentrations were recorded for methyl tert-butyl ether (MTBE). Heptachlor epoxide and o, p'-DDE (DDT metabolite) were detected most often among the pesticides. Sulphate ions were found in all samples. Very high concentrations of sodium and chloride ions were measured in samples collected directly before the winter season. Overall, the results confirmed that road runoff waters are heavily polluted and their quality should be monitored.

Keywords: road runoff waters, toxic substances, urban area, environmental analysis, chromatographic analysis

Introduction

Vehicular traffic is considered a major source of air pollution in urban environments. A large number of pollutants are emitted to the urban atmosphere during combustion of gasoline, diesel oil and other fuels in internal combustion engines [1-9]. Tire wear also contributes to urban air pollution. Another important source of pollution is the combustion of fossil fuels to produce heat, particularly in small residential furnaces and central heating plants. Urban areas are often also industrial centres, with electricity production by power plants of medium and large sizes. Production of heat and electricity, as well as various industrial processes, contribute substantially to urban air contamination.

The most important pollutants in urban air include gases (CO, NO_x, SO_2), non-methane volatile organic pollutants (NMVOCs), as well as particulate matter and compounds sorbed on its surface, including major cations and anions, several trace elements and organic compounds. Some of the pollutants emitted to the atmosphere are transported with air masses out of the emission region. The remaining ones are deposited within the urban region through dry and/or wet deposition. Some pollutants undergo physical and chemical transformations, resulting in the formation of new pollutants, such as dioxins [10-19]. Precipitation is a very important deposition route from the air to other environmental compartments,

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particularly for water-soluble gases and chemical compounds sorbed on fine particles. The amount of air pollutants deposited through precipitation depends on the intensity of precipitation and its pH, as well as on aqueous solubility of the pollutants. While removed from the air by precipitation, air pollutants reach the terrestrial and aquatic ecosystems through runoff water. This water also carries the pollutants deposited on the roofs of buildings, roads, etc. Runoff water is an important transport medium for various air pollutants from the air to the surface- and ground water, and then indirectly to other environmental compartments [20-37].

Contamination of runoff water from roads with high traffic intensity in the urban area of Gdańsk, Sopot, Gdynia and Reda in Poland is discussed in this paper.

Material and Methods

Samples of runoff water were collected in the Gdańsk area, which is a large municipality in the eastern part of the Baltic coast of Poland. The population of the region is about 500,000 in the three cities of Gdańsk, Gdynia and Sopot. Major point sources of pollution in the region include harbours, shipyards, an oil refinery, power plants and a phosphate fertilizer plant. Traffic and combustion of coal and oil in small residential furnaces are the main diffuse sources of air pollution in the region. Road runoff samples were collected during the rainy season, from October 24 to December 18, 2000. Additional samples were collected following the winter season, on March 9, 2001. The samples were collected at the following seven sites, six of which (nos. 2-7) were located along the major route connecting the city of Reda and the city of Gdańsk (see Fig. 1): **1.** Reda, M. Buczka street (Traffic intensity app. 100 cars per hour); **2.** Reda, Gdańska Street (near a railway station); **3.** Gdynia, Morska Street; **4.** Sopot, Aleja Niepodległości Street; **5.** Gdańsk-Oliwa, Aleja Grunwaldzka Street; **6.** Gdańsk-Wrzeszcz, Aleja Grunwaldzka Street and **7.** Gdańsk, Podwale Grodzkie Street; (sites **2** – **7:** traffic intensity approximately 1,000 to 3,000 cars per hour).

Road runoff was collected during or immediately after (15 min maximum) a rain event. The samplers were plastic containers, from which samples were dispensed to amber glass bottles prior to their transportation to the laboratory. Samples were stored at low temperature (4-7°C) in a dark room, with no preservatives added. In general, all analyses were performed within 24 hours of sample collection, with those involving volatile compounds within 2 hours.

Due to the high contents of solid particles (sand, dust, etc.) in the road runoff samples, inclusion of a filtration stage was necessary in some analyses. Analytical procedures used in the study (including the parameters of chromatographic determination of pesticides, volatile organohalogen compounds, petroleum hydrocarbons and the cations and anions, as well as the detection limits of the different methods) were described previously [38].

Total organic carbon was determined in the samples by converting all organic compounds into CO_2 (sample mineralization) and determining its amount coulometrically. The determinations were performed using the CM 5300 Furnace Apparatus Version 1.0 (UIC INC. COULOMETRICS).

Results and Discussion

Analysis of the samples yielded a large body of data on the occurrence and levels of a wide spectrum of analytes in road runoff. Table 1 presents the concentration ranges and the frequency of occurrence of the analytes from the different groups in the samples. Following is a brief discussion of the results.

Even though pollutant concentrations often vary by several orders of magnitude during a runoff event, a single index known as event mean concentration (EMC) can be used to characterize runoff constituents. EMC represents a flow average concentration defined as the total pollutant load divided by total runoff volume [39-41]. EMCs for three events are presented in Table 2.

Pesticides

Heptachlor epoxide (metabolite of heptachlor; formed in plants, soil and homeothermic organisms), and o, p'-DDE (DDT metabolite) were the most often detected pesticides in the runoff water samples. Fig. 2A, presenting the sums of the concentrations of organochlorine pesticides in the samples, illustrates the

Fig. 1. Locations of the sampling sites.

Table 1. Maximum and mean analyte concentrations in road runoff water samples collected in the study.

Analyte	Concentration		Units	Total number of samples (number of samples in which analyte was detected)
	Maximum	Mean		
Volatile organohalogen compounds				
$CHCl_3$	0.53	0.29	µg/dm^3	53 (53)
$CHBrCl_2$ +C_2HCl_3	0.08	0.02		53 (45)
$CHBr_2Cl$	0.01	0.01		53 (20)
CCl_4	0.01	0.01		53 (24)
Petroleum hydrocarbons				
MTBE	269	116	µg/dm^3	24 (11)
Hexane	9.66	4.41		24 (17)
Benzene	12.7	3.97		24 (15)
Heptane	9.71	2.86		24 (17)
Toluene	38.4	9.72		24 (22)
Isooctane	3.56	3.56		24 (1)
Octane	3.71	2.96		24 (12)
m-xylene	5.08	2.59		24 (11)
Decane	6.09	3.25		24 (10)
Anions				
F^-	7.92 (0.63)	3.15 (0.17)	mg/dm^3 (meq/dm^3)	59 (11)
Cl^-	4770 (136)	306 (8.73)		59 (59)
NO_2^-	3.48 (0.08)	3.48 (0.08)		59 (1)
NO_3^-	13.8 (0.22)	2.79 (0.04)		59 (29)
PO_4^{3-}	371 (11.7)	218 (6.88)		59 (2)
SO_4^{2-}	163 (3.4)	25.9 (0.54)		59 (59)
Cations				
NH_4^+	14.8 (0.82)	3.50 (0.19)	mg/dm^3 (meq/dm^3)	59 (17)
Na^+	1328 (57.8)	107 (4.67)		59 (59)
K^+	21.9 (0.56)	7.82 (0.20)		59 (59)
Mg^{2+}	6.87 (0.57)	2.22 (0.19)		59 (59)
Ca^{2+}	70.0 (3.5)	24.3 (1.22)		59 (59)
Organonitrogen and organophosphorus pesticides				
Simazine	0.48	0.27	µg/dm^3	54 (9)
Atrazine	4.10	0.71		54 (17)
Propazine	12.1	3.25		54 (40)
t-Butylazine	12.4	2.38		54 (14)
Bromophos	2.86	0.27		54 (34)
Malathion	0.46	0.11		54 (10)
Chlorfenvinphos	0.13	0.06		54 (16)
Fenitrothion	0.13	0.08		54 (3)

Continued Table 1

Organochlorine pesticides				
α-HCH	5.83	3.36	µg/dm^3	54 (3)
γ-HCH	61.4	31.3		54 (7)
Aldrine	21.1	2.68		54 (14)
Heptachlor epoxide	57.1	8.75		54 (24)
o,p'-DDE	127	18.1		54 (17)
p,p'-DDD	8.38	3.09		54 (11)
o,p'-DDD	1308	212		54 (10)
p,p'-DDE	213	29.4		54 (9)
o,p'-DDT	2.41	1.38		54 (3)
p,p'-DDT	2.88	1.66		54 (6)
Methoxychlor	5.70	2.63		54 (13)
TOC				
	2.03	113	mg C/dm^3	49 (49)

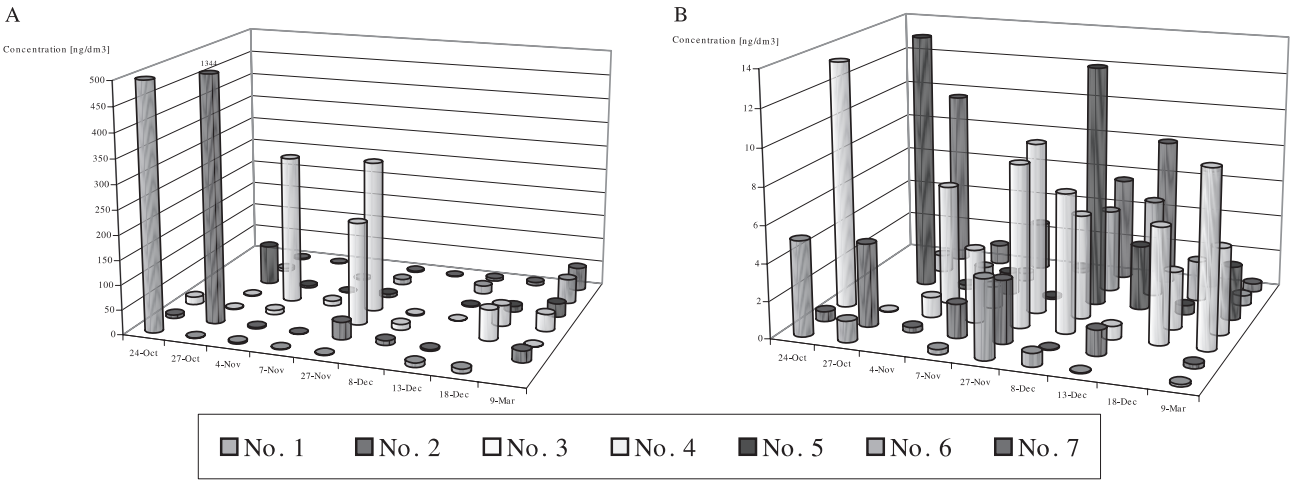

Fig. 2. Calculated sums of pesticide concentrations in road runoff samples collected at the seven sites: A – organochlorine pesticides, B – organonitrogen and organophosphorus pesticides.

Table 2. Range of event mean concentration (EMC) for three events (24, 27 october and 4 november 2000).

EMC	
Anions mg/dm^3	
Cl^-	10.5 – 23.5
SO_4^{2-}	61.4 – 88.3
Cations mg/dm^3	
Na^+	12.8 – 17.1
K^+	8.5 – 11.1
Mg^{2+}	1.56 - 1.59
Ca^{2+}	23.8 - 26.5

total load of the samples with pesticides from this group. The highest value for this sum (1.3 µg/dm^3) was recorded for the sample collected on October 27, 2000, at site no. 2. Fig. 2B presents the sums of the concentrations of organonitrogen and organophosphorus pesticides in the samples of road runoff waters collected at the different sites. Propazine and bromophos, used as selective weed killers, were detected the most often among the pesticides from these two groups. The highest value for the sum of the concentrations of organonitrogen and organophosphorus pesticides (14 ng/dm^3) was recorded on the same day (October 27, 2000) at site no. 5, located in Gdańsk-Oliwa. The concentration of organochlorine pesticides in this sample was also relatively high at 81 ng/l. Elevated pesticide concentrations in runoff waters were also observed for samples

collected on October 24, 2000. This may have been related to the fact that those were the first precipitation events following a prolonged dry spell. Both sites (nos. 2 and 5) are characterized by very heavy traffic volume. However, the pesticide contamination of road runoff was quite clearly related to agricultural uses rather than to traffic.

Volatile Organohalogen Compounds

Volatile organohalogen compounds (VOX) were found in all samples examined. In this group, the most often detected analytes included chloroform ($CHCl_3$; used among others as a solvent, degreasing agent, insecticide carrier, PVC modifier and heat transfer agent), dichlorobromomethane ($CHBrCl_2$) and trichloroethene (C_2HCl_3). The highest chloroform concentration (0.53 µg/dm^3) was recorded for the sample collected on December 8, 2000, at site no. 2 (city of Reda, railway station).

Fig. 3A presents the calculated sums of the concentrations of the individual VOX in the samples collected. Each series was characterized by two maxima, recorded on November 4, 2000 (all sites except no. 5) and December 8, 2000 (all sites except no. 6). The first maximum was observed at site no. 5 on November 7, while the second maximum at site no. 6 occurred on December 13. The latter was the only exception to the general trend of decreasing VOX concentrations in road runoff water samples collected after December 8, 2000. The VOX presence in runoff waters can be explained most likely by their widespread use as solvents and degreasing agents in various branches of industry in the area (shipyards, ports, chemical industry, etc.) and in small car repair shops and dry cleaners.

Petroleum Hydrocarbons

In this group, the most frequently detected analytes included methyl tert-butyl ether (MTBE, detected in 11 of 24 samples examined), benzene and toluene. Their presence in road runoff waters can be explained by emissions from vehicles through spills, evaporation from fuel tanks and emissions with exhaust gases following incomplete combustion of the fuel. Other sources of petroleum hydrocarbons in the environment include evaporation and accidental spills during crude oil drilling and processing and during handling of the fuels. Figure 3B illustrates the sums of the concentrations of petroleum hydrocarbons in road runoff water samples. The highest values of this parameter were recorded on December 13, 2000 (sites no. 1 and 5) and on December 18, 2000 (sites no. 1, 3 and 7).

Cations and Anions

Na^+, K^+, Mg^{2+} and Ca^{2+} were detected in all samples, with the highest concentrations recorded for Na^+ and Ca^{2+}. Among the anions, the highest concentrations were found for Cl^- and $SO_{4^{2-}}$. The presence of sulphate ions in all samples was most likely related to the beginning of the heating season, as traffic emissions contribute relatively little to overall SO_2 emissions. The power plants in the area, supplying heat for central heating, are coal fired (coal may contain as much as 5% ww. of sulphur). In addition, many apartments are also heated with coal furnaces.

Nitrite ions were detected only in a single sample collected on October 24 at site no. 3, while phosphate ions were detected in two samples collected on December 18 (sites no. 2 and 6). Fluoride ions were detected most frequently at sites no. 7 and 6. On December 18, they were

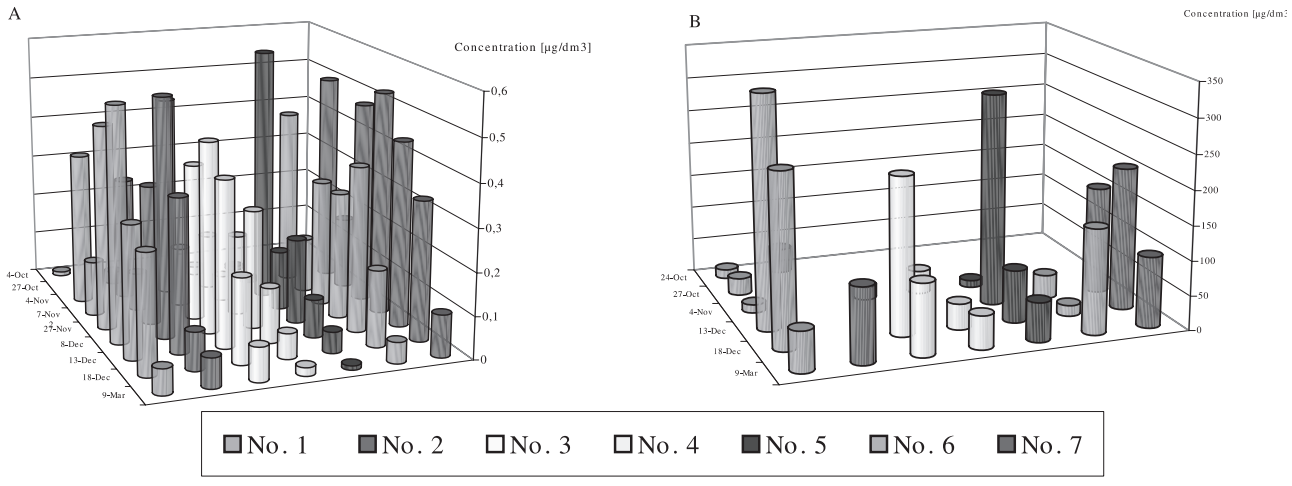

Fig. 3. Calculated sums of volatile hydrocarbon concentrations in road runoff samples collected at the seven sites: A – organohalogen compounds, B – petroleum hydrocarbons.

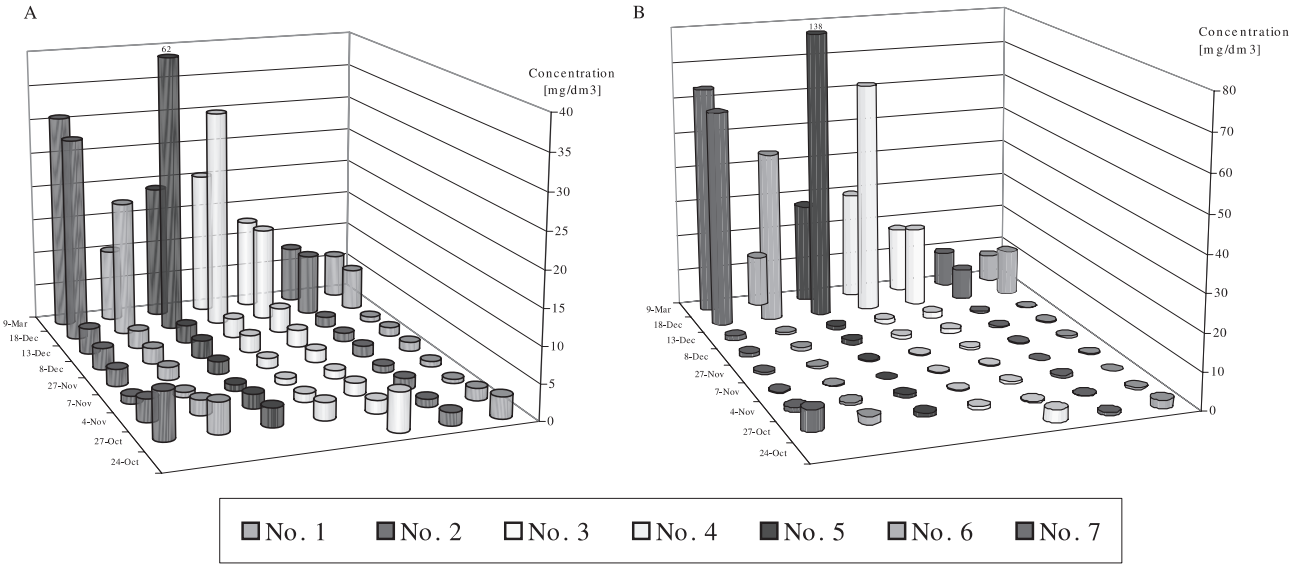

Fig. 4. Calculated sums of the concentrations of (A) cations and (B) anions in road runoff samples collected at the seven sites.

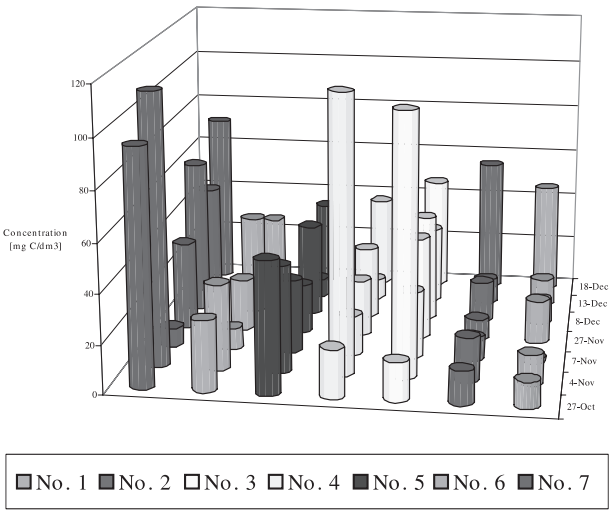

Fig. 5. TOC levels in road runoff samples collected at the seven sites

detected in samples collected at all seven sites. The presence of fluorides in runoff waters may be related to atmospheric migration of pollutants originating from the phosphogypsum stack near the phosphate fertilizer plant. Figs. 4A and 4B present the calculated sums of the concentrations of the cations and anions determined in the study. December 18, when the highest values were recorded for these parameters, was the last rainy day before snowfall. It was also the coldest of all the sampling days. Very high concentrations of chloride, sodium and magnesium ions were found in samples collected on this day. In the case of the chloride ions, the extremely high concentrations (4,770 mg/dm^3 at site no. 5) can only be explained by the use of road salt to prevent icing in the area.

Total Organic Carbon

The highest TOC values were observed on November 4, 2000 for samples collected at sites nos. 3 (city of Gdynia), 4 (city of Sopot) and 7 (city of Gdańsk). Fig. 5 illustrates the TOC levels in the samples collected in the study.

Temperature Dependencies

Figs. 6 and 7 present examples of the dependencies of hydrocarbon and TOC concentrations in road runoff waters on ambient temperature. In general, hydrocarbon concentrations decreased when air temperature increased. In the case of volatile organohalogen compounds (Fig. 6A), the results were scattered and the overall quality of the correlation was poor. The trend was clearer for petroleum hydrocarbons (Fig. 6B). The occurrence of the trend for volatile organohalogen compounds and petroleum hydrocarbons was not surprising taking into account that analytes from both groups are volatile, therefore tend to partition into the gas phase at higher temperatures. A similar trend was observed for TOC (Figs. 7A and B), which seems to indicate that the majority of the organic compounds present in the road runoff samples in this study were volatile.

Conclusions

The study was carried out over a period of 2 months (from October 24 to December 18, 2000), with an additional set of samples collected on March 9, 2001. Overall, 59 samples of road runoff were collected along the major highway connecting the cities of Gdańsk and Reda. The samples were analyzed for the presence of analytes from 5 different groups. A total of 403 analyses were per-

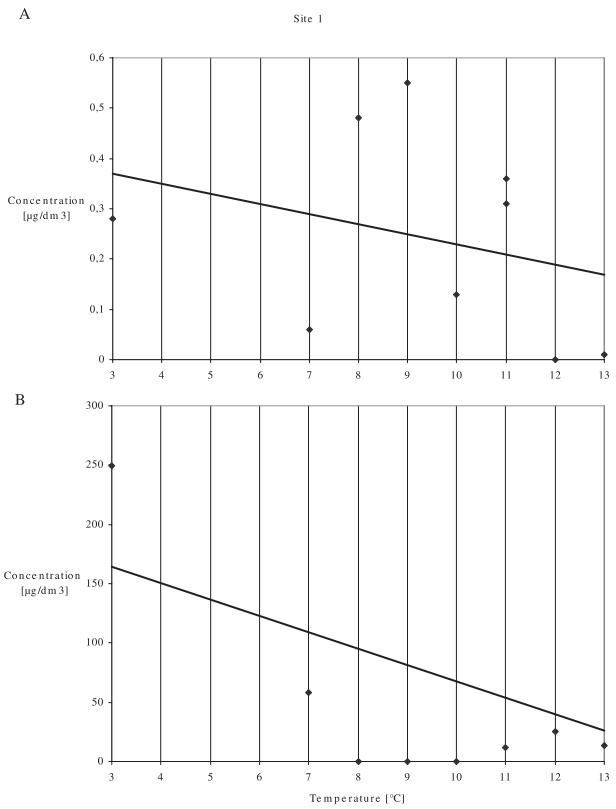

Fig. 6. Relationship between the levels of volatile hydrocarbons and ambient temperature for road runoff samples collected at site no. 1 (city of Reda): A – organohalogen compounds, B – petroleum hydrocarbons.

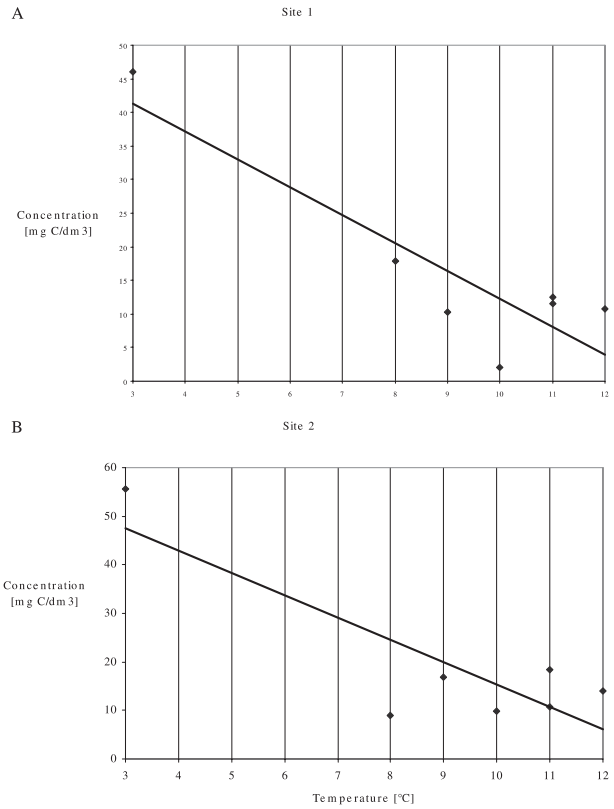

Fig. 7. Relationship between TOC levels and ambient temperature for road runoff samples collected at (A) site no. 1 and (B) site no. 2.

formed. A comparison of the results with literature data indicates that both the types of the pollutants and their levels were comparable to those found in road runoff waters in other geographical locations. For example, petroleum hydrocarbon levels in runoff waters ranged from 160 to 340 µg/dm^3 in France [42] and from 300 to 500 µg/dm^3 in USA [43], compared to 10 – 335 µg/dm^3 in this study. The levels of sulphate ions varied from 2.29 to 163 mg/dm^3 in this study, compared to 60.4-270.9 mg/dm^3 in the UK [23], 4.9-520 mg/dm^3 in Norway [25] and 5-650 mg/dm^3 in the USA [20]. Sulphate levels determined in a similar study carried out between October 1999 and March 2000 in Gdańsk ranged from 1.81 to 201 mg/dm^3 [38]. Finally, the TOC levels found in this study (2.03 – 113 mg C/dm^3) were similar to the levels found in Norway (8-320 mg C/dm^3 [25] and 3.5-48 mg C/dm^3 [44] and in the USA (4.5-141 mg C/dm^3 [20]).

The analytes determined in the study can originate from many different sources, including road deposition (e. g. from tire wear, break wear, fluid leaks), atmospheric deposition (transport of particulate matter and gaseous pollutants), road wear (cracks and potholes in asphalt and concrete pavements), road maintenance practices (use of de-icers or pesticides) and accidental spills [20]. Overall, road runoff carries with it a substantial load of toxic substances and can be potentially dangerous to the environment. Consequently, it is our opinion that the quality of road runoff should be monitored, especially in large urban agglomerations with heavy traffic.

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