

Influence of Mobile Sources on Pollution of Runoff Waters from Roads with High Traffic Intensity

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Abstract

This paper presents the results of monitoring and evaluating the degree of environmental pollution in Gdańsk, Poland, based on the analysis of rainwater and runoff waters from roads with high traffic intensity. Rainwater and road runoff were collected at two sites located in Gdańsk. The concentrations of the following analytes were determined: petroleum hydrocarbons (PH), polycyclic aromatic hydrocarbons (PAH), heavy metals, anions, cations and pH. Road runoff from the two sites was slightly basic with average pH values ranging from 7.04 to 7.45. Heavy metals concentrations in runoff water samples were higher than in rainwater samples. Higher concentrations of heavy metals were observed in precipitation samples and runoff samples during the day, which is connected with traffic intensity. The concentration of ions, PH and PAH in runoff waters was higher than in precipitation waters and increased during the day, together with increasing traffic intensity. Overall, the results confirmed that road runoff waters are heavily polluted and their quality should be monitored.

Keywords: rainwater, road runoff water, urban area, environmental analysis

Introduction

Urban areas are characterized by high population densities and economic development. The resulting pollutant emissions place increasing pressure on the air quality of these areas. Whereas in the past the major reasons for poor air quality were industrial activity and domestic heating, nowadays, as a result of the rapid increase in mobility, the major current urban air pollutants come from road traffic [1, 2]. The influence of transport on the environment is a very complex and multi-aspect phenomenon. Road transport interferes with the environment, influencing all of its elements: the atmosphere, soil, water, the shaping of the land, the geological structure,

plant life and the animal world. Today, occupation of space by the linear and point infrastructure of transport is, besides air pollution from exhaust fumes, one of the most important problems connected with transport [3]. Sustainable transport has become a key global transport objective. One of the major ways of improving sustainability is to reduce car ownership and use and encourage a modal shift towards public transport [4]. To design an air pollution abatement strategy it is first necessary to identify pollution sources and to quantify their emissions [2]. Emissions from road vehicles in Europe are expected to fall markedly by the year 2010 in response to planned and probable control measures across the European Union (EU) [5].

Year by year the interference of transport in the natural environment along a wide front, together with an increasing number of cars and transport devices, are increasing the lev-

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Fig. 1. Location of the sampling sites; (1) – Armii Krajowej Street, (2) – Słowackiego Street

el of threat to the environment along transport routes. One of the important ways through which pollutants connected with these activities enter the environment are road runoff.

Several investigations have been carried out to determine the concentrations of common heavy metals (e.g. Cd, Pb, Zn) in road runoff during short periods [6]. Apart from runoff, heavy metals can be transported from the road through splash and particulate transport [7]. Rainfall runoff from urban pavements often contains significant quantities of dissolved metal elements (DME), particulate-bound metal elements (PME), and suspended, colloidal and volatile fractions of particulates [8]. These diffuse constituents result from traffic activities, roadway maintenance operations and atmospheric deposition. Tire wear is a source of zinc and cadmium. Brake wear is a source of copper, lead, chromium and manganese. Engine wear and fluid leakage is a source of aluminium, copper, nickel and chromium. Vehicular component wear and detachment is a source of iron, aluminium, chromium and zinc [9, 10].

Automobile exhaust gases contain many non-polar polycyclic aromatic hydrocarbons (PAHs) such as benzo[a]pyrene and benzo[e]pyrene, and also oxygenated polycyclic aromatic hydrocarbons (oxy-PAHs) such as anthraquinone and xanthone. They are generally formed from oil and coal upon incomplete combustion [11, 12]. However, methyl-t-butyl ether (MTBE) and ethanol (used as fuel) are the most frequently used oxygenate additives

in gasoline to enhance fuel octane and to reduce carbon monoxide and ozone. Although exhaust gas pollutants from traffic are selectively reduced by catalytic converters, they may contribute to MTBE concentrations in ambient air [13], as MTBE itself can be present in the exhaust gas when MTBE-containing gasoline is used [14].

The study presented in this paper is an evaluation of the contamination of roads with high traffic intensity in big urban agglomerations by selected organic and inorganic compounds, quite often cancerogenic and mutagenic [15, 16]. The research results presented in this paper are a continuation of previous research data that were published previously [10, 17-19].

In accordance of pollutants presented above in rain and road runoff water, the following groups of compounds

Table 1. Limit of detection, expanded uncertainty and intermediate precision of analytical methods.

Parameter	Anions	Cations	PH	PAH
Limit of detection	0.01 mg/l	0.01 mg/l	0.03 µg/l	0.005 µg/l
Expanded uncertainty	1	1	2	2
Intermediate precision	2	2	4	5

Table 2. Concentration range and mean concentration of analytes found in samples of rainwater and road runoff.

Analytes	Armii Krajowej Str.				Słowackiego Str.			
	Concentration		Number of samples	Frequency of occurrence (%)	Concentration		Number of samples	Frequency of occurrence (%)
	Range	Mean			Range	Mean		
Rainwater								
pH	6.40-7.09	6.62	13	100	6.04-6.90	6.67	8	100
Cl ⁻ [mg/l]	0.79-26.1	5.53	13	100	1.69-17.5	4.91	8	100
NO ₃ ⁻ [mg/l]	0.44-3.82	2.03	13	100	0.43-3.47	1.66	8	100
SO ₄ ²⁻ [mg/l]	0.45-10.6	4.94	13	100	0.99-14.8	4.84	8	100
NH ₄ ⁺ [mg/l]	0.043-0.15	0.096	13	100	0.013-0.063	0.038	8	100
Pb [µg/l]	0.40-53.1	8.10	13	100	0.30-69.3	15.2	8	100
Cd [µg/l]	0.10-5.80	0.62	13	77	0.10-5.41	1.90	8	87
Zn [µg/l]	112-856	312	13	100	93-1080	267	8	100
Σ PAH [µg/l]	-	0.051	1	100	-	0.055	1	100
Σ PH [µg/l]	18.7-24.5	20.6	4	100	10.2-16.32	13.9	4	100
Road run-off								
pH	6.80-7.35	7.08	13	100	6.78-7.45	7.10	8	100
Cl ⁻ [mg/l]	32.7-15564	3541	13	100	21.8-13576	1953	8	100
NO ₃ ⁻ [mg/l]	0.89-5.90	2.70	13	62	0.60-8.10	3.05	8	62
SO ₄ ²⁻ [mg/l]	11.9-139	53.8	13	100	3.53-133	63.2	8	100
NH ₄ ⁺ [mg/l]	0.16-0.81	0.36	13	100	0.091-0.36	0.19	8	100
Pb [µg/l]	1.0-880	258	13	100	8.1-1600	380	8	100
Cd [µg/l]	0.20-79	19	13	85	0.20-80	14.2	8	87
Zn [µg/l]	73-864	355	13	100	107-1090	314	8	100
Σ PAH [µg/l]	-	0.092	1	100	-	0.068	1	100
Σ PH [µg/l]	17.2-379	108	4	100	7.67-21.2	16.1	4	100

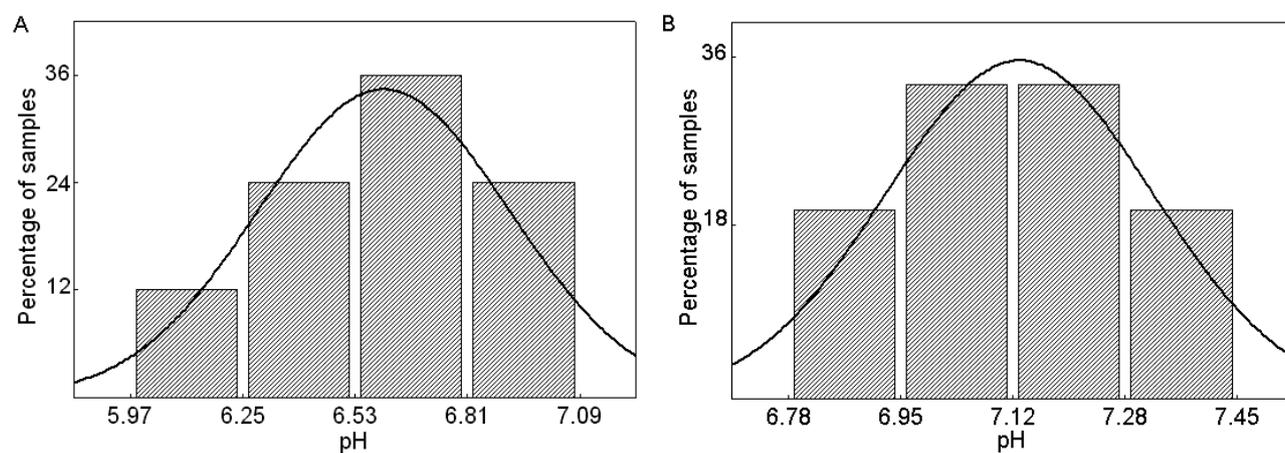


Fig. 2. Percentage distribution of pH for 21 rainwater samples (A) and 21 road runoff samples (B).

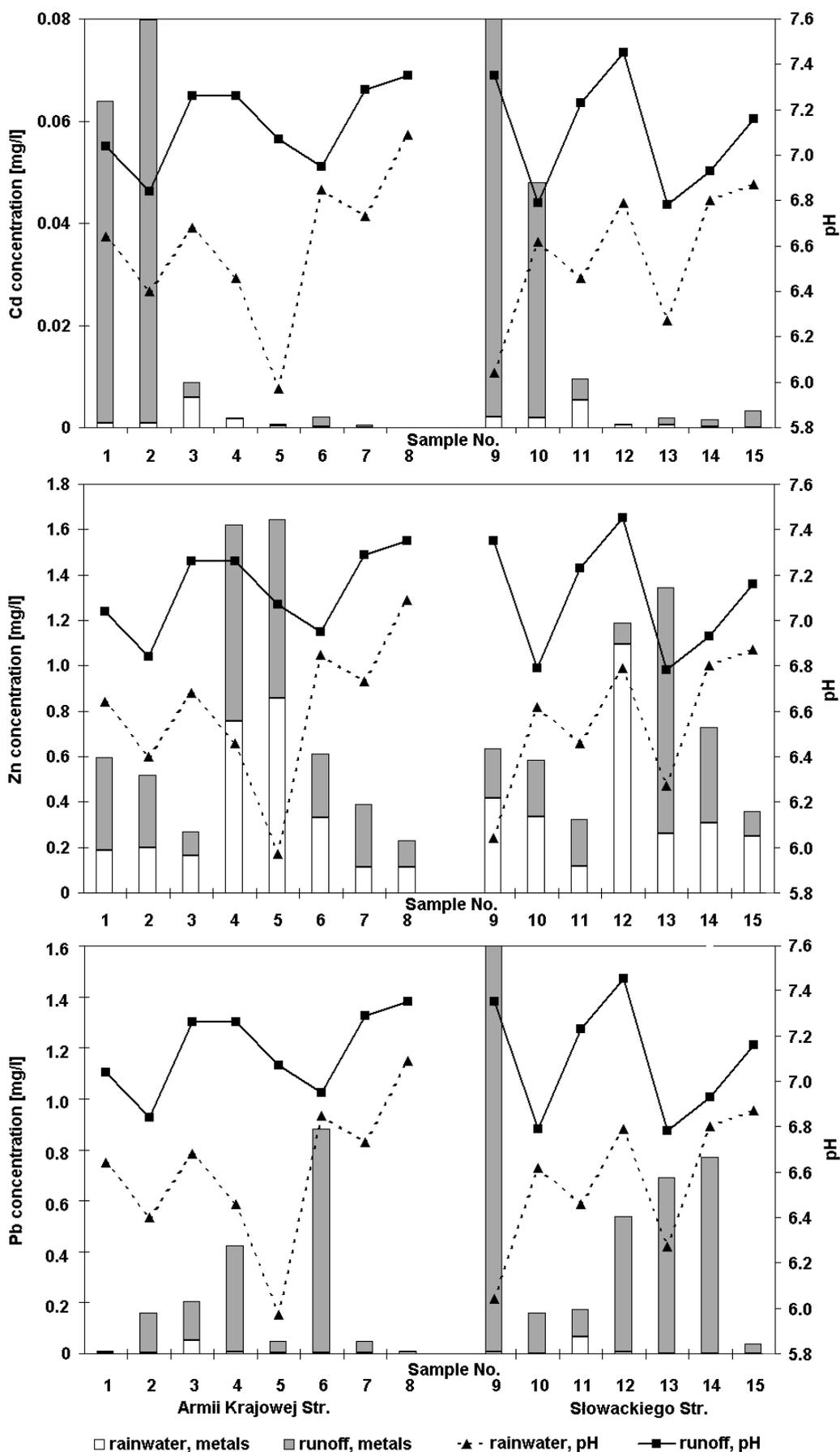


Fig. 3. Average concentration of heavy metals and pH in rainwater and runoff water samples: Cd (A), Zn (B), Pb (C).

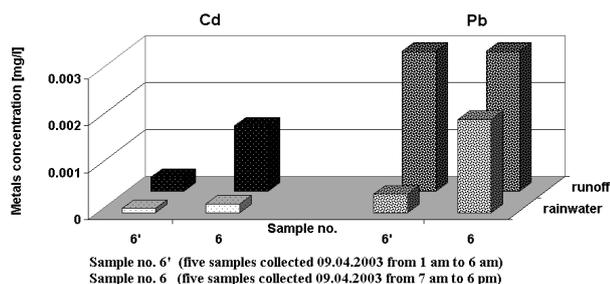


Fig. 4. Average concentration of heavy metals in rainwater and run-off water samples during day and night.

were identified: PAHs, PH (petroleum hydrocarbons), heavy metals, anions and cations. Due attention was paid to the influence of the sampling site on the amount and diversity of pollutants present in the analyzed forms of atmospheric precipitation (rain and road runoff).

Methods

Sampling

Samples of rain and runoff waters from major highways were collected from January 2003 to May 2003 at two sites (day and night) in the city of Gdańsk: Site 1: Armii Krajowej Street (day: 3000 vehicles/hour; night: 1700 vehicles/hour) and Site 2: Słowackiego Street (day:

2100 vehicles/hour; night: 1000 vehicles/hour). Fig. 1 illustrates the locations of the sites. Samples were collected using a sampler based on the design described by Chiwa et al. [20], Bannerman et al. [21], Waschbusch et al. [22]. Samples were collected during or immediately after a precipitation event (8 events; 21 of rain and 21 of runoff water samples; rainfall above 2 l/m²), stored at 4°C without any chemical preservatives and analyzed within 24 hours.

Analytical Methods

Table 1 lists limit of detection, expanded uncertainty and intermediate precision of analytical methods. The samples were analyzed for PAH and PH by means of gas chromatography with mass spectrometry (GC-MS) and flame ionization detection (GC-FID), respectively [19, 23]. Selected anions and cations were determined using ion-suppressed chromatography and quantified against synthetic rain standards (RAIN-97, CRM 409) [24, 25]. Heavy metals (dissolvent fraction) were determined by Atomic Absorption Spectroscopy [26]. pH was measured with a CX-315 pH-meter [19].

Results

The conducted research has allowed us to gather a large amount of data regarding the content of selected ana-

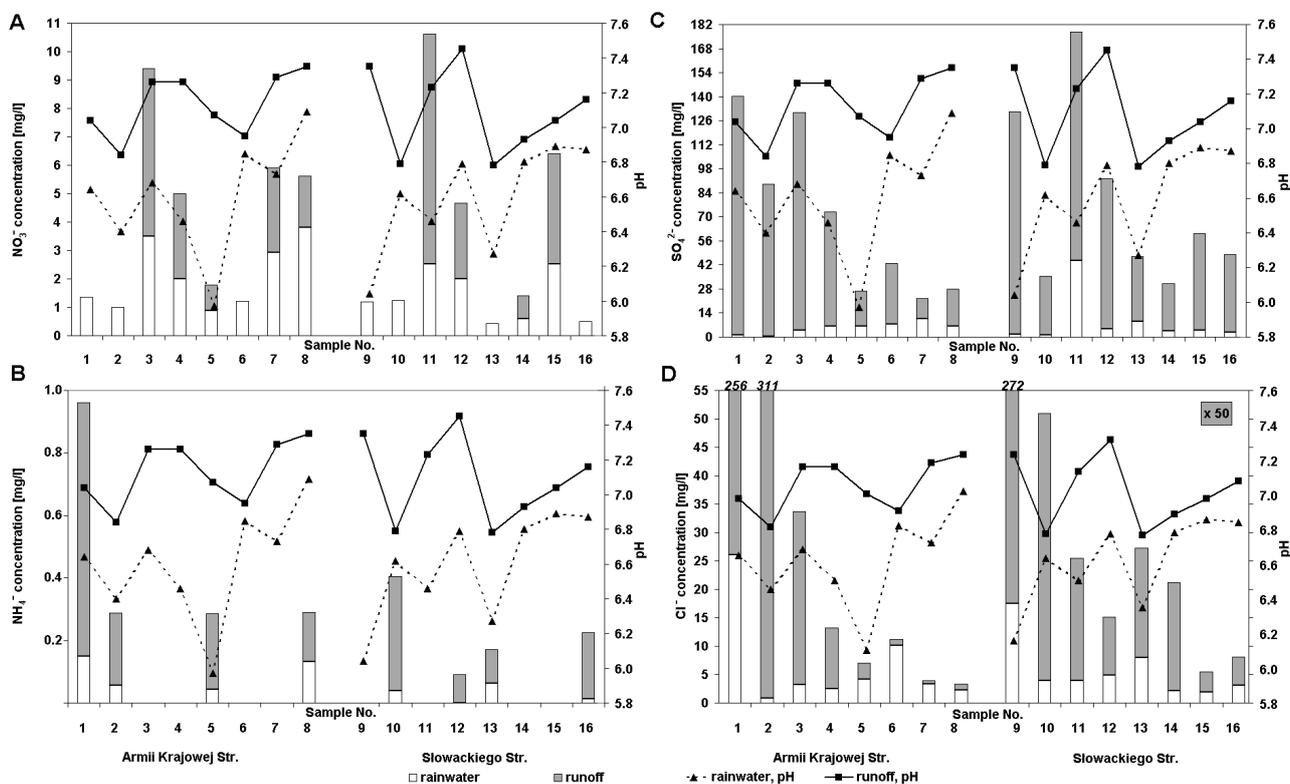


Fig. 5. Average concentration of ions and pH in rainwater and runoff water samples: A-NO₃⁻, B-NH₄⁺, C-SO₄²⁻, D-Cl⁻ (concentration of chloride ions in runoff waters multiplied by 50).

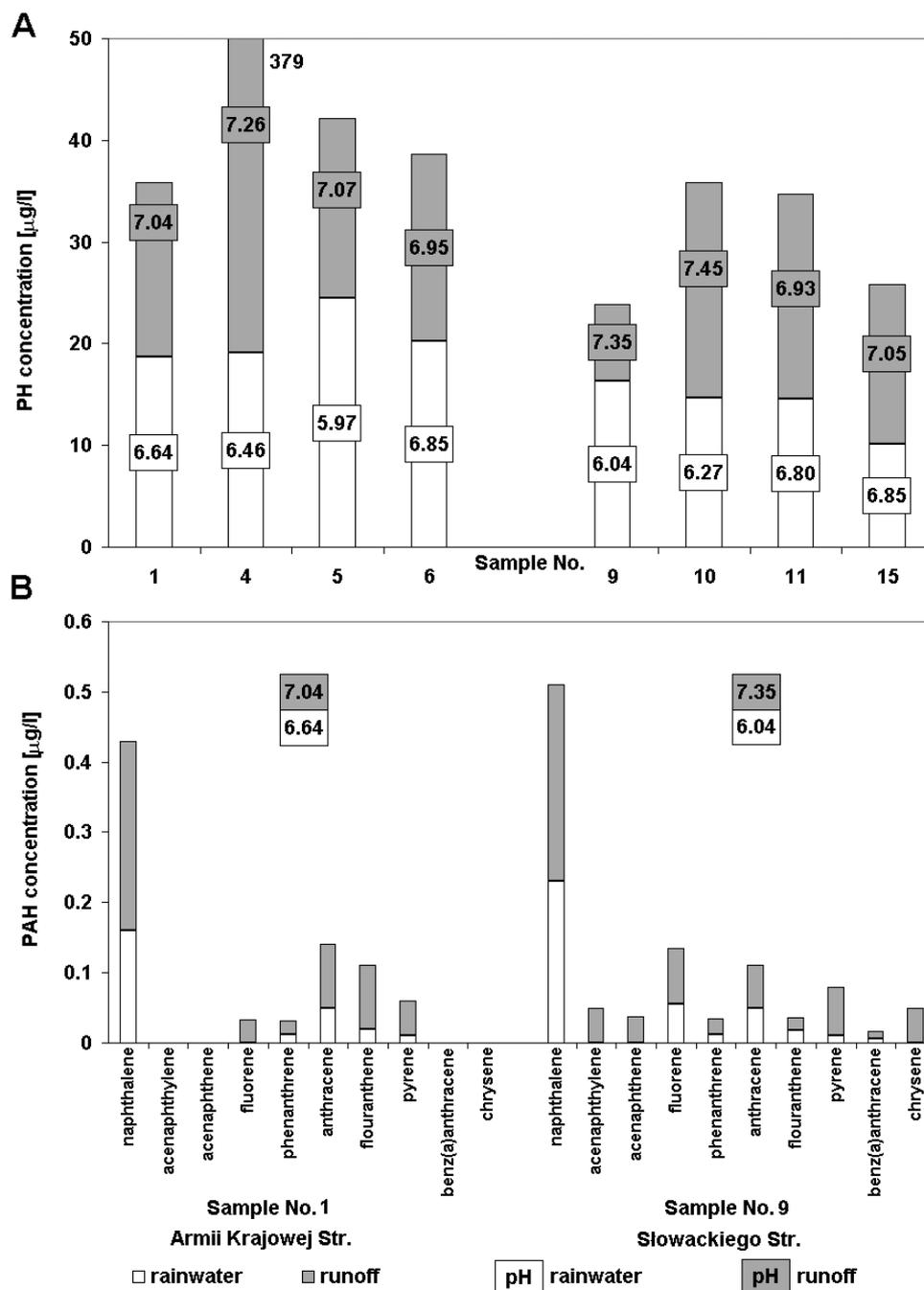


Fig. 6. Average concentration of PH (A) and PAH (B) in rainwater and runoff water samples.

lytes from the organic and inorganic compound groups in rain and runoff water samples from roads with high traffic intensity in Gdańsk. Table 2 lists the ranges of concentrations determined (the final result of determination was calculated as a mean value of three concurrent measurements) and mean concentrations of PH (benzene, ethylbenzene, hexane, heptane, toluene, MTBE, xylene, nonane, butylbenzene, isopropylbenzene), PAH (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, flouranthene, pyrene, benz(a)anthracene, chrysene), anions and cations, and heavy metals for the entire sampling period.

pH

Rainwater from the two sites was mainly acidic with average pH values ranging from 6.04 to 6.90 (exception pH = 7.09, sample No. 8, Armii Krajowej St.). Road runoff from the two sites was slightly basic with average pH values ranging from 7.04 to 7.45 (Fig. 2).

Heavy Metals

Heavy metal concentrations in the runoff samples were significantly higher than in the rainwater samples (Fig. 3).

Cd and Pb concentrations were higher by ten to one-hundred times, respectively. Zn concentrations in runoff water samples were comparable to rainwater samples. Higher concentrations of heavy metals were observed in precipitation samples and runoff samples during the day, which is connected with higher traffic intensity (Fig. 4).

Anions

The concentration of ions in runoff waters was higher than in precipitation waters (Fig. 5) and increased during the day, together with increasing traffic intensity. High concentrations of chloride ions determined in the rain and runoff water samples may be due to the location of the sampling sites in the coastal areas of the Baltic Sea (marine aerosols). In the case of runoff water samples, it may be additionally connected with the usage of road salts (NH_4Cl , CaCl_2 , MgCl_2) in order to prevent icing in a given area. Fluoride (0.040 mg/l) and phosphate ions (8.27 mg/l) were detected only in precipitation sample No. 5 (05.04.03). Their presence may be explained by the migration of pollutants transported by dust particles from the waste dump, which is situated in the Gdańsk Phosphate Fertilizer Plant. It is probable because the direction of the air mass inflow during that day was strictly north (arctic mass, atmospheric front) and phosphate plants are localized in that region. Fig. 5 also shows the concentration of SO_4^{2-} , NO_3^- , Cl^- ions in rain and runoff waters. It shows that SO_4^{2-} and NO_3^- ions are present in the form of dissolved salts (e.g. Na_2SO_4 , NaNO_3). pH value grows together with an increasing NH_4^+ ion concentration (e.g. sample No. 1, rain: $\text{NH}_4^+ = 0.150$ mg/l, pH = 6.64; runoff: $\text{NH}_4^+ = 0.811$ mg/l, pH = 7.04; sample No. 2, rain: $\text{NH}_4^+ = 0.057$ mg/l, pH = 6.40; runoff: $\text{NH}_4^+ = 0.231$ mg/l, pH = 6.84; sample No. 10, rain: $\text{NH}_4^+ = 0.039$ mg/l, pH = 6.62; runoff: $\text{NH}_4^+ = 0.365$ mg/l, pH = 6.74; and sample No. 16, rain: $\text{NH}_4^+ = 0.013$ mg/l, pH = 6.87; runoff: $\text{NH}_4^+ = 0.213$ mg/l, pH = 7.17). Whereas Fig. 5 D shows that when the Cl^- ion is higher, pH becomes lower. Similar results regarding the determination of selected cations and anions in road runoff was performed in Gdansk at the break of 1999/2000, which confirmed the proposition that road runoff in an urban agglomeration carries a large load of pollutants that contribute to the degradation of the environment [19].

PH and PAH

In all the analyzed samples of rain and road runoff, there was a confirmed presence of naphthalene, flouranthene, phenanthrene, anthracene, pyrene, and benzene, ethylbenzene, hexane, heptane, toluene, MTBE, xylene, nonane, butylbenzene and isopropylbenzene. PAH and PH concentrations in runoff samples exceeded the concentration of these compounds in rainwater samples (Fig. 6A & B). Research conducted in Madrid has proven that there exists a close dependence between an increase in urban traffic intensity and high concentration levels of compounds from the PAH group and simple hydrocarbons in urban runoff

waters [27]. The highest concentrations were recorded for MTBE (91% overall PH concentration in runoff for Armii Krajowej St.), benzene (18% overall PH concentration in runoff for Słowackiego St.), toluene (47% overall PH concentration in rain for Słowackiego St.; 66% overall PH concentration in runoff for Słowackiego St.). Their presence can be explained by emissions from vehicles through spills, evaporation from fuel tanks and emissions with exhaust gases following incomplete combustion of the fuel (regular, eurosuper, super premium unleaded and "Optimax" gasoline contain MTBE as an octane enhancer [28]).

Discussion and Conclusion

The research presented in this paper is a continuation of the preliminary studies on the chemistry of runoff waters in Poland described in an earlier paper [19]. The studies performed here showed that concentrations of measured analytes were higher in runoff waters than in precipitation waters. Higher pH values in runoff samples (pH > 7) result from a higher amount of base-forming factors (e.g. ammonium ions) in runoff waters.

The maximum concentration of ammonium ion (0.81 mg/l) was reported for a sample collected from Armii Krajowej St. (3,000 vehicles/hour). In comparison with the sample collected from Słowackiego St. (2,100 vehicles/hour), this value is more than two times higher. Similar pH results have been obtained by other authors [29-32]. Barbosa and Hvitved-Jacobsen [29] presented data concerning samples of road runoff which were collected at four different sampling sites, where pH values ranged from 5.9 to 7.2. pH values ranging from 6.3 to 7.9 were reported for 48 events by Legret and Pagotto [30]. However, Maltby [33] determined pH results as high as 8.23 in runoff water samples. The pH of rainwater is acidic, which is mainly due to a high concentration of SO_4^{2-} and NO_3^- components. The higher values of SO_4^{2-} compared to NO_3^- at all the sites indicates that SO_2 is more important than NO_x for the acidity of rainwater. The levels of sulphate ions in runoff waters varied from 11.9 to 139 mg/l in this study, compared to 60.4-270.9 mg/l in the UK [33], 4.9-520 mg/l in Norway [34] and 5-650 mg/l in the USA [35].

Concentrations of heavy metals in the runoff samples were similar to those reported elsewhere [33, 36-38]. Among heavy metals, Zn and Pb exhibited the highest concentrations, as expected for typical pollutants associated with vehicular traffic [39]. The main source of lead is fuel. However, it appears that only 5% of lead is removed by runoff water. The largest fraction may, therefore, disperse in the atmosphere or settle on the soil by the roadside. A relatively high level of Zn could be explained by the existence of metallic guardrails. According to Hewitt and Rashed [40], the main sources of zinc are wear and tear from tires and brakes and from the corrosion of galvanized safety barriers, or from an alternative material [29]. As for Pb and Zn, concentrations increase in winter

because of the use of chloride-based deicing salts, which generate supply and corrosion phenomena.

PH levels in runoff waters range from 160 to 340 $\mu\text{g/l}$ in France [34] and from 300 to 500 $\mu\text{g/l}$ in the USA [41], compared to 5–379 $\mu\text{g/l}$ in this study. PH concentrations in runoff water and precipitation samples are comparable, but there are some events when these values are even 20 times higher (sample no.4, 31.03.03). However, taking into account all events, the ratio between PAH concentrations in runoff water samples and PAH concentration in rain samples is not higher than 2. While Hewitt and Rashed [42] presented the information in their paper that from 1 to 8% of the selected PAH (phenanthrene, anthracene, fluorantene, pyrene), which were determined in runoff water samples, is deposited on the road or on nearby soil and grass surfaces, whereas the remaining 92–99% is dispersed away from the motorway. Finally, the PAH levels in runoff waters found in this study (0.55–0.68 $\mu\text{g/l}$) were similar to the levels found in England (0.36–7.06 $\mu\text{g/l}$ [40]) and in France (0.011–0.474 $\mu\text{g/l}$ [30]).

The obtained results enabled us to get information on the effect of mobile sources on the quality of road runoff. The research has confirmed the high level of road pollution as well as the necessity to restrict the emission of exhaust gases from vehicles.

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