Original Research

Physico-Chemical and Ecotoxicological Characterizations of Urban Storm Water Runoff

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Abstract

Suspended solids, petroleum hydrocarbons (C_{14}-C_{28}) (PH), benzo(a)pyrene (B(a)P), heavy metals (Pb, Cd, Zn, Cu, Hg), and pH were analyzed in storm water runoff samples collected at five sites in different areas of Vilnius city during three rain episodes in May-June, and at one site characterized by intensive traffic conditions during six rain episodes in May-October 2007. The concentration of xenobiotics investigated in samples was highly variable, depending on the sampling site and rainfall intensity. The evaluation of the toxicity of storm water runoff samples to rainbow trout was undertaken. Alterations of fish biological parameters (mean growth, white blood cell count) revealed significant differences in the toxic effects of storm water runoff samples depending on the runoff discharge point and chemical composition. The most significant decrease in the growth of rainbow trout fry was induced by storm water runoff, with the highest concentrations of benzo(a)pyrene and petroleum hydrocarbons.

Keywords: chemical composition, fish, storm water runoff, toxicity, xenobiotics

Introduction

Investigations of urban water cycle during recent decades have shown that pollution of urban water is associated not only with industrial and municipal wastewaters, but also with storm and snowmelt runoff. Urban runoffs can wash pollutants from the roofs of buildings, vehicles, ground surfaces of residential, industrial, and commercial regions, plus their waste dumping sites increasing their concentrations in surface waters and bottom sediment. They also can infiltrate into deeper layers of soil, impacting ground water resources [1-3]. Storm water studies have been widely performed and developed in the USA since 1970 [4-6], and later in industrially developed EU countries [7, 8]. In the majority of observations it was determined that urban storm water runoff is a significant pollution source to natural water basins. Moreover, it has been indicated that the major pollutants in storm water runoff are adsorbed to suspended solids (SS), which are not usually investigated in connection with their chemical composition, and which can cause uncertainties in quantification of storm water runoff pollution [4, 9]. The results of different investigations encourage evaluation of storm water runoff pollution in different locations with different urbanization levels, landscapes, and climatic conditions [10, 11]. The studies of storm water runoff pollution are only at their initial stage in Lithuania, and they are mainly directed toward analysis of common water parameters (pH, BOD, SS) [12].

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Furthermore, the literature data suggest that the majority of compounds transported by storm water runoff are known to be toxic and usually expose ecological systems to mutagenic and carcinogenic risks [1, 8]. This can cause a problem with natural water system pollution from discharge of urban storm water. Currently, the toxic effect of urban storm water runoff has been studied insufficiently. The toxicity of some individual chemicals has been investigated using mostly algae and aquatic plants [8, 13, 14], while studies of the effect of polluted storm water runoff on different species of fish are scarce [15, 16]. Fish are widespread and economically valuable water inhabitants that could be exposed to a complex mixture of contaminants in the natural water bodies that, in turn, may alter adaptive or protective reactions of fish [17].

The purpose of this study was to determine the physicochemical composition of Vilnius storm water runoff and to assess the load of Vilnius city outflow systems by dissolved and particle-bound xenobiotics. Additionally, evaluation of the toxicity of storm water runoff samples to the fish, rainbow trout (*Oncorhynchus mykiss*), was undertaken.

Experimental Procedures

Sampling Sites

The sampling of storm water runoff was performed in Vilnius (403 km^2 area, ~600,000 inhabitants) at five sites of specific urban characteristics from the common drainage system with storm water runoff discharge to the basin of the Neris River. The chosen sampling sites No. 1 and No. 2 are mostly characterized by high density of residents, developed commercial activity, and high intensity of traffic (3,000-5,000 veh./h), while three other sites (3, 4, and 5) are mostly landscaped with several industrial objects and enterprises.

Characteristics of sampling points and discharge of runoff:

1. Area of Antakalnis traffic intersection (residential area with intensive traffic (3,500-4,000 veh./h), area of drainage − 20.7 ha, annual discharge – 52,992 m^3/year;
2. Area of Geležinio Vilko and Saltoniškių Street crossing (commercial area in the city center with traffic intensity over 5,000 veh./h), area of drainage − 41.9 ha, annual discharge – 107,264 m^3/year;
3. Vilkpedė (industrial and commercial area), area of drainage – 33.4 ha, annual discharge – 85,504 m^3/year;
4. Liudvinavas (suburb with chemical storage), area of drainage – 13.9 ha, annual discharge – 35,584 m^3/year;
5. Vaidotai (suburb with substantial railway sidings), area of drainage − 10.0 ha, annual discharge – 25,600 m^3/year.

The annual discharge volume of runoff was calculated according to the average annual precipitation depth in Vilnius (h=640 mm) and the coefficient of infiltration of 0.6 by equation:

$$V = F\,(1 - 0.6)\,h{\cdot}10 \qquad (1)$$

...where V is the annual storm water runoff volume (m^3/year), F is the catchment area (ha), h is the average annual depth of precipitation (mm).

Sampling and Analysis

The samples of storm water runoff were collected at five sampling sites in the city during three rainfall episodes on 2 and 29 May and on 26 June, 2007. Additionally samples were collected in area No. 1 (characterized by intensive traffic conditions) during three rain episodes on 2, 4, and 12 October, 2007. Some samples were replicated, totally 25 samples of storm water runoff were collected. Each runoff sample consisted of no less than three grab samples taken with the interval of ten minutes during the first hour of rainfall due to the fact that the first flush of rainfall is mainly polluted [18]. After collection, the samples were sealed in pre-cleaned jars and stored at 4ºC in darkness until analysis. The intensity of rainfall was observed continuously while sampling of the runoff at site No. 1 by measuring the precipitation amount per unit of time. Prior to analysis, the runoff samples were separated into solid and liquid phases. The concentrations of xenobiotics in the runoff were determined by the methods used for pollutant analysis in water media and SS, and were described in detail [19]. The methods applied to the determination of xenobiotics are approved by international intercalibration or standardized according to regulation of the Ministry of Environment of the Republic of Lithuania [20, 21].

For analysis of heavy metals, storm water runoff was filtrated through ashless filters (DP 145090 ALBET). Heavy metals such as Pb, Cd, Cu, Zn, Hg, B(a)P, and PH were analyzed in the water of runoff (soluble form) and in SS of runoff (insoluble form). Concentrations of heavy metals were determined with a Zeeman/3030 atomic absorption spectrophotometer (detection limit for Cd 0.05 µg/l; for Pb, Cu, and Zn 0.5 µg/l). Mercury measurement was performed using a "Gardis" mercury analyzer (detection limit 0.001 µg/l) [21]. For analysis of B(a)P and PH, SS were filtrated through "Filtrak" paper filters for finest sediments No. 90, Ø 11cm (German production). The concentrations of B(a)P were determined with a Spectrometer DFS-12 (detection limit 0.0001 µg/ml) [19]. PH concentration (C_{14}-C_{28}) was determined by infrared (IR) spectrometry (IR spectrophotometer AN-1, detection limit 10 µg/l). pH was determined by potentiometric method with a PerHecT Meter.

Toxicity Test

Storm water runoff samples collected at four sampling sites (No. 1, No. 2, No. 3, and No. 4) in Vilnius on 2 May were tested on rainbow trout fry for 21 days. Rainbow trout (Oncorhynchus mykiss) juveniles were obtained from the Žeimena Hatchery (Lithuania) and maintained in holding tanks of about 3,000 l capacity supplied with flow-through artesian aerated water. The toxicity test was undertaken under semi-static conditions with five groups of fish (ten

Table 1. Ranges and average concentrations of xenobiotics in storm water runoff.

Xenobiotics	Soluble form				Insoluble form				Ratio
	Min.	Max.	Average	*C.V., %	Min.	Max.	Average	*C.V., %	C_s/C_{ins}
Cd, [μg/l]	0.03	0.86	0.18	133	5.3	17.5	9.5	45.9	0.018
Pb, [μg/l]	0.50	6.20	1.77	96.6	24.4	68.2	47.1	38.5	0.038
Cu, [μg/l]	7.10	36.0	14.3	73.4	15.3	70.2	34.9	56.6	0.40
Zn, [μg/l]	12.0	129	54.1	60.8	49.6	488	174	88.8	0.31
Hg, [μg/l]	0.02	0.06	0.04	25.0					
B(a)P, [μg/l]	0.004	0.018	0.01	50.0	0.04	4.00	0.47	108	0.021
PH, [mg/l]	0.15	2.67	0.83	109.6	0.92	82.3	22.9	164	0.04
SS, [g/l]					0.048	3.64	0.92	129	

*C.V. − coefficient of variation

individuals in each group) according to the standard method with minor alterations [22]. During this study, fish were fed on commercial DANA FEED fish food *ad lib*. Artesian water of high quality was used as control water. The average hardness of control water was approximately 284 mg/l as $CaCO_3$, pH ranged from 7.6 to 7.8. Concentrations of parameters studied in control water constituted: Pb – 0.7 μg/l, Cd – 0.05 μg/l, Zn – 2 μg/l, Cu < 0.05 μg/l, Hg – 0.007 μg/l, B(a)P – 0.003 μg/l, PH – 0.03 mg/ and, SS – 0.7 mg/l.

Control water and urban storm water runoff in the aquaria were renewed three times a week. The fish body weights were determined gravimetrically at the start and at the end of exposure periods. Blood was sampled from each group of fish and blood smears were prepared. Smears were stained with May-Grünwald-Giemsa and Romanovsky stains and in smears 200 leukocytes were classified according to different counts [23]. The temperature of water in the aquaria was maintained at 10±0.5ºC, and the oxygen concentration in the aquaria ranged from 8 to 10 mg/l. The reliability of these data was evaluated using Student's t-test at P≤0.01 and P≤0.05.

Results and Discussion

Concentrations of Pollutants in Storm Water Runoff

Due to the variety of factors influencing the pattern of concentrations, it is difficult to systematize the concentration values in various areas of Vilnius. Therefore, the data obtained from five sampling sites during the whole period of the investigation are presented as ranges and average values of pollutant concentrations in the territory of the city (Table 1).

The data presented in Table 1 indicate that the concentrations of the principal xenobiotics in soluble and insoluble forms in the city varied in wide intervals. The highest concentrations of Cd, Pb, Zn, and OP were determined at the Antakalnis road intersection (site No. 1) during rainfall on 2 October, in the center of the city (site No. 2) and in the area of Liudvinavas (site No. 4) on 12 October. The high concentrations of Pb and PH were also determined in insoluble form in the area of Vaidotai on 12 October, when the number of antecedent dry days before the rainfall was predominant (4-8 days). The greatest variability of data was determined for the soluble form of minor xenobiotics (C.V. ranged between 25% and 133%) and for insoluble form of PH (C.V.=163.6%). The variation of the PH concentration at different sites of the city is related to traffic, vehicle repair enterprises, and parking sites where leakage of PH on the ground surface is possible. Their washout by storm water runoff may drastically influence the pattern of the PH concentration in the city.

Distribution of pollutants between soluble and insoluble forms showed that relatively high amounts of Cd, Pb, B(a)P, and PH were found in solid phase (>90%), while Cu and Zn were determined up to 70% in this phase. The C_s/C_{ins} ratio indicating pollutant distribution in soluble and insoluble forms varied in the interval of 0.018-0.40. It was lowest for Cd and B(a)P and highest for Cu and Zn. The values of pH of storm water runoff varied in the range of 7.4-8.7, and this lightly alkaline medium reduced solubility of heavy metals in storm water runoff and inhibited their release from SS [24]. The concentration of SS in storm water runoff exceeded its concentration prescribed for outflows of treatment plants (25 mg/l). Therefore, the concentrations of investigated heavy metals, PH, and B(a)P usually exceeded their maximum permissible concentration (MPC) for the water-receivers. Besides, the concentrations of investigated pollutants were very similar to those detected in Paris, where high concentrations of Cd (0.1-32 μg/l), Cu (3.0-247 μg/l), and Zn (57-38,061 μg/l) were determined in storm water runoff from roofs, yards and streets of the city [7]. A high concentration of SS in urban storm water runoff from the city drainage system allows us to admit that the generation of pollutants during storm water runoff results from the erosion of the city soil where historical accumulation of atmospheric deposition and local leak-

Table 2. Dependence of xenobiotic concentrations on rainfall intensity.

Date of sampling	Rainfall intensity	Cd	Pb	Cu	Zn	B(a)P	PH	SS	Antecedent dry days
	[mm/h]	[μg/l]					[mg/l]	[g/l]	
2007.05.02	0.6	4.80	19.0	12.6	37.5	0.19	2.80	0.17	2
2007.05.29	3.4	10.4	32.7	35.1	49.6	1.60	8.33	1.45	1
2007.10.02	4.8	17.5	58.5	70.2	134	1.24	12.2	2.25	4
2007.10.12	5.9	12.5	46.8	55.6	367	1.80	14.2	1.66	8
2007.10.04	6.7	8.33	68.3	64.7	188	4.00	10.8	3.64	2
2007.06.24	10.2	13.1	64.1	66.1	272	2.92	11.7	3.22	3
Correlation coefficient		0.468	0.914	0.895	0.726	0.853	0.882	0.862	

age of pollutants was observed [4, 19]. The highest concentrations of traffic-related metals, PH, and B(a)P were determined in the bulk deposition and soil in residential and commercial (central) areas of Vilnius, which probably were impacted by street density, courtyards, and automobile parking squares [25, 19]. High concentrations of Pb, Zn, and PH determined at site Nos. 1 and 2 with the developed traffic confirmed the suggestion that street runoff could be the main source of the annual pollutant loads entering the Neris River and other city outflows.

Dependence of Pollutant Concentrations and Rainfall Intensity

During the experiment, it was observed that the concentrations of SS and the insoluble forms of xenobiotics in storm water runoff depended on rainfall intensity. This dependence of insoluble forms of the xenobiotics and SS at sampling site No. 1 during six rainfall episodes is presented in Table 2. With an increase of rainfall intensity, the rapid increase in SS and xenobiotics was noticeable in storm water runoff samples. The correlation coefficients indicated that the increase of soluble and insoluble forms of the xenobiotics depended on the intensity of the rainfall with a confidential reliability of 98%. This relationship is expressed more significantly for the SS, Pb, Cu, B(a)P, and PH, and less so for Cd and Zn. Consequently, the variation of xenobiotic concentrations presented in Table 2 may be based both on traffic development and on rainfall intensity.

Estimation of Xenobiotic Loading for Vilnius Water Outflow System

According to the concentrations of xenobiotics (Table 1) and the storm water runoff annual discharge, loading factors for the outflow system of Vilnius (area ~403 km^2) were calculated and evaluated (Table 3). The amount of pollutants which may be accumulated in storm water runoff and discharged into the urban water cycle can be presented in a decreasing order: SS>PH>Zn>Cu>Pb>Cd>B(a)P. The controversial data of pollutant loading by storm water runoff depending on washout objects (building sidings and roofs, automobiles, auto brakes and tires, oil leakage, lands of different use, etc.) are presented in literature sources. Our data of heavy metal concentrations are very close to those determined for different land uses by Wong et al. (Pb loading 0.6 kg/ha yr and Cu loading 0.2 kg/ha yr) and for urban oil leakage determined by Davis et al. (48 l oil/ha yr) [2, 26]. The annual load of the Vilnius water outflow system from storm water runoff discharge is from a few to a few hundred times larger for the insoluble form of xenobiotics than that for the soluble form. If the average load of water-related PH is 85.5 tons/year, then the load for the particle-bound PH could be on average 2,360 tons/year. Furthermore, recent investigations have shown that the SS are distinguished by sorption properties, especially for PH, which are sorption activators for other types of xenobiotics [27]. Subsequently,

Table 3. Loading of xenobiotics for Vilnius outflow system (tons/year).

Xenobiotic	Xenobiotic form	Minimum	Maximum	Average
Cd	soluble	0.003	0.088	0.018
	insoluble	0.55	1.80	0.98
Pb	soluble	0.053	0.638	0.182
	insoluble	2.51	7.03	4.85
Cu	soluble	0.73	3.71	1.47
	insoluble	1.58	7.23	4.11
Zn	soluble	1.24	13.3	5.60
	insoluble	5.10	29.7	17.9
B(a)P	soluble	0.001	0.002	0.001
	insoluble	0.004	0.296	0.087
OP	soluble	15.4	274	85.5
	insoluble	94.7	8.47	2.36
SS		4.94	271	74.2

Table 4. Effects of storm water runoff on the growth of rainbow trout.

Runoff samples	Weight at start of exposure, g	Weight at end of exposure, g	Significant difference (t)	
			#	with control
Site No. 1	0.83±0.05	1.16±0.10	2.95*	1.48
Site No. 2	0.79±0.04	1.02±0.06	3.19*	3.47*
Site No. 3	0.92±0.05	1.15±0.06	2.95*	2.06*
Site No. 4	0.95±0.05	1.08±0.06	1.66	2.82*
Control	0.88±0.06	1.34±0.07	4.7*	

Difference between weight of fish at start and at end of exposure.
* Values of parameters are significantly different (P≤0.05).

highly polluted suspended particles, owing to their ability to accumulate pollutants over a prolonged period, must be separated from the runoff prior to the storm water runoff being discharged into receivers of the city outflows (e.g. rivers or sewage systems).

Evaluation of Storm Water Toxicity

The total concentrations of xenobiotics in liquid and solid phases in storm water runoff samples for evaluation of storm water toxicity were analyzed from respective site Nos. 1, 2, 3, and 4. Simultaneously, the control tests with rainbow trout were performed in the aquaria with artesian aerated water. The concentrations of heavy metals in samples varied within wide ranges. Pb in storm water samples ranged from 0.7 to 6.2 μg/l, Cd ranged from 0.35 to 0.98 μg/l, Zn from 18.6 to 429 μg/l, Cu from 1.6 to 39.5 μg/l, and Hg ranged from 0.05 to 0.53 μg/l. The concentration of Zn was higher (4-fold) in sample No. 2 and the concentration of Cu was strongly elevated (3-fold) in the first two samples (Nos. 1 and 2) as compared to MPC of these metals for the water-receivers. The concentration of B(a)P ranged from 0.28 to 0.53 μg/l, the concentration of PH ranged from 3.72 to 3.91 mg/l, and those in the first three samples of storm water runoff exceeded MPC of these xenobiotics for the water-receivers. Runoff sample No. 4 was relatively clean. The values of pH of storm water runoff studied varied in the range of 7.4-8.3.

No mortality of rainbow trout fry both in control individuals and in fish exposed to urban storm water runoff was registered in the toxicity study. However, a substantial negative effect of the polluted runoff on fish was observed in differences in their growth. Though all exposed and control individuals were fed on the same food in the same amount, their weight at the end of the experiment was different (Table 4). The weight of fish exposed to runoff sample Nos. 1, 2, and 3 at the end of exposure increased significantly (P≤0.05) as compared to that at the start of exposure. However, the growth of fish treated with runoff sample Nos. 2 and 3 was significantly lower as compared to controls (P≤0.05). The weight of fish treated with runoff sample No. 4, however, did not change at the end of exposure, and possibly the growth of some fish was even inhibited. The effect of toxic properties of sample Nos. 1, 2, and 3 on the growth of fish fry was related largely to the high amounts of PH and B(a)P. Petroleum hydrocarbons are practically insoluble in water (solubility in water 0.01μg/l), but they are highly lipophilic and can be accumulated successfully by living organisms, especially in fish liver and gills. It is generally accepted that oncogenesis is most likely to occur in the tissue exposed to polyaromatic hydrocarbons for a long time when certain amounts of these substances are accumulated in fish tissues [28]. The chemical analysis of runoff sample No. 4 indicated relatively low amounts of the studied contaminants; however, an adverse effect on the growth of fish was observed. We admit that the runoff from sampling site No. 4, characterized as a site of chemical storage, contained some other toxic substances that were not analyzed in this study for technical and financial reasons. According to a report on dangerous substances in the aquatic environment of Lithuania [29], Vilnius wastewater contains organic compounds such as phthalates, organic chlorinated compounds, and detergents, etc. Chemicals used in industry and agriculture that can be found in storm water runoff in very low (μg and lower) concentrations after long exposure could induce adverse effects on aquatic organisms and disturb the immune responses in fish [30, 31]. Furthermore, the adverse effect of urban storm water runoff on fish was confirmed by changes in haematological parameters. A statistically significantly higher proportion of juvenile lymphocytes was found in the blood of fish exposed to runoff sample Nos. 1 (2.24±0.3%) and 3 (2.44±0.7%) than that in the controls (0.86±0.1%) (P≤0.05). Additionally, the neutrophile count was increased in the groups of fish exposed to runoff sample Nos. 2 and 3 (8.3±0.9% and 7.2±1.2%, respectively) as compared to control (5.8±0.9%). These elevations of lymphocyte and neutrophile count demonstrated disturbances in both specific and non-specific immune mechanisms in fish, which may result in impaired resistance of fish to diseases.

Conclusions

Chemical analysis of storm water runoff in five areas of Vilnius city during different rainfall episodes showed that the concentrations of SS, heavy metals (Cd, Pb, Cu, Zn, Hg), PH, and B(a)P were different and varied in a wide range depending on the pollution of the sampling site and rainfall intensity. Most of the xenobiotics were determined in storm water SS. Insoluble forms of Cu and Zn made up to 70% of their total amount measured in storm water runoff, while that of Pb, Cd, PH, and (B(a)P) made up over 90%. Therefore, the loading of city outflows can differ by three orders of magnitude if the pollution from both forms of runoff is evaluated.

The evaluation of the toxicity of urban storm water runoff samples to rainbow trout fry indicated differences in the toxic effects of runoff water to fish. The most significant adverse effects on the growth of fish fry were induced by storm water runoff, with the highest concentrations of B(a)P and PH. The elevation of selected haematological parameters (juvenile lymphocyte and neutrophile count) demonstrated the disturbances in both specific and non-specific immune mechanisms of fishes. Such disturbances may result in the impaired resistance of fish to adverse environmental effects.

The findings of our study indicate that greater attention should be paid to the control and treatment of urban storm water runoff, discharge of which into natural water basins can cause negative ecotoxicological consequences.

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