

## EFFECTIVENESS OF OIL DERIVATIVES REMOVAL FROM STORMWATER TREATED BY THE EXPERIMENTAL CONSTRUCTED WETLAND BEDS IN SEMI-TECHNICAL SCALE

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**Abstract:** In the paper, the research results on the removal of aliphatic hydrocarbons (C7 to C30) on constructed wetlands have been presented. The research has been realized on the semi-technical scale constructed wetlands, planted with reed *Phragmites australis*. The experimental installation is located on the filling station in Balice and treats the fraction of stormwater from this utility. The concentrations of total aliphatic hydrocarbons in analyzed stormwater were between 96.02  $\mu\text{g}/\text{dm}^3$  and 6177.33  $\mu\text{g}/\text{dm}^3$ , and from 47.55  $\mu\text{g}/\text{dm}^3$  to 5011.14  $\mu\text{g}/\text{dm}^3$  in effluent from the installation. The average total aliphatic hydrocarbons removal effectiveness was 48%, the values ranged from 19% to 81%. Hydrocarbons C14 to C18 were removed with the lowest effectiveness (26%–32%), the lighter hydrocarbons – with higher one (39%–68%), however the highest removal effectiveness were observed for the hydrocarbons with the highest carbon atoms numbers (from 51% for C20 to 92%–93% for C26–C30).

### INTRODUCTION

The elaboration of the rational method to treat stormwater is one of the most urgent challenges of the modern environmental engineering, both in Poland [16] and worldwide [12, 22]. As the result of urban development a significant increase of stormwater amount has occurred – its rational management is more and more important, and mixing it with municipal wastewater, widely used in Poland, is the outdated and inefficient solution. The problem is especially important and difficult in the case of stormwater from motorways and parking lots [9, 17] – this kind of runoff has to be pretreated before releasing to the wastewater network and to the wastewater treatment plant. It is caused by the oil derivatives present in this kind of wastewater. Oil-derivatives due to their widespread occurrence and high harmfulness both for human health and the environment are micro-pollutants of special importance. Aliphatic hydrocarbons are the group representing oil-derivatives, which is usually measured in stormwater and for which the limits are defined by environmental protection legislations. Total aliphatic hydrocarbons (TAH) concentrations observed in runoff from highways and parking lots reach up to 24  $\text{mg}/\text{dm}^3$  [22, 18],

while Polish law defines the maximum TAH concentration at the level of 15 mg/dm<sup>3</sup> for wastewater released both to the environment [15] and to municipal systems collecting wastewater [14]. For these reasons, stormwater from highways and parking lots has to be treated and oil-derivatives have to be removed from it. In countries with the developed highway infrastructure, constructed wetlands (CW) are more and more widely applied to treat this kind of runoff [18, 21]. Due to the complex interrelated processes of pollutants' removal, which occur in constructed wetlands [6], they guarantee effective and economic stormwater treatment in the place of its formation [17, 23]. As it has been reported by several authors, constructed wetlands have the potential to efficiently treat both run-off from highways and parking lots [12, 17] and other kinds of wastewater polluted with oil-derivatives [11, 5, 7, 8]. The research reports have also proved the capability of several species of macrophytes to function in an environment highly polluted with oil-derivatives [13, 20], as well as the activity of microorganisms (both aerobic and anaerobic) capable to digest oil-derivatives [18] and thus remove these substances from water.

To assess the possibility of using this technology in Poland, the author conducted a series of the pilot pot experiments and proved the high potential of constructed wetlands to remove TAH and other oil-derivatives from wastewater [1, 2, 3, 4]. The next phase of the research was to design and conduct the experiments in a semi-technical scale. The experimental installation constructed for these experiments has been described in the article, the obtained results have been also presented and discussed, particularly the effectiveness of TAH removal from stormwater.

### EXPERIMENTAL INSTALLATION

The semi-technical scale experimental constructed wetland installation is located next to the filling station in Balice, Olszanicka street. Beside the station on the southern direction, it is surrounded by: the stream with no name in the north, the International Airport Kraków-Balice in the west and A4 highway in the east. The scheme of the installation is presented in Figure 1.

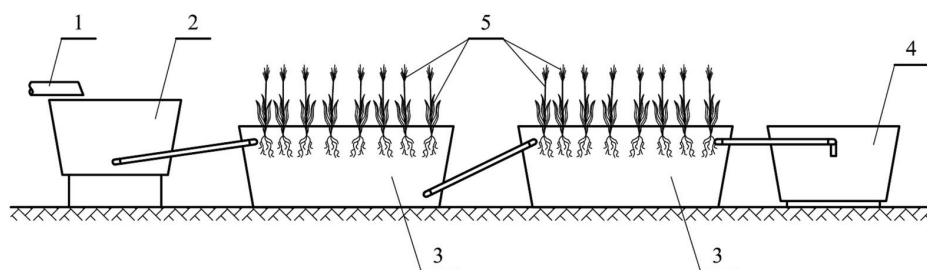


Figure 1. The scheme of the experimental installation: 1 – stormwater influent, 2 – sedimentation tank, 3 – CW bed, 4 – final tank, 5 – reed (*Phragmites australis*)

The main part of the installation consists the subsurface horizontal flow constructed wetlands in a form of two reed beds connected in series. Gravel, which has been used as the beds' filling, is characterized by diameter 8–20 mm and porosity 37%. These two beds were planted with *Phragmites australis*. Before the first constructed wetland, there is a preliminary sedimentation tank, from which pretreated stormwater flows to the first main

bed through a perforated pipe to guarantee the stable flow. Effluent from the second bed is stored in the final container – the last element of the experimental installation.

Stormwater is gathered from the parking lot which belongs to the filling station, from the square with fuel pumps and the access road, as well as from the roof of the building and the roofing of the pumps. The fraction of the total stormwater stream is directed to the experimental plantation with the prefabricated rain collecting channel ACO SELF, which is 50 cm long and has closed ends. Stormwater from the channel is transported to the preliminary sedimentation tank with the pipe (PP DN75) of 6.5 m length. The area of CW beds is about 0.6 m<sup>2</sup>, their total volume – about 0.16 m<sup>3</sup>, and the porous volume – about 0.06 m<sup>3</sup>. All four containers of the installation are the same heavy-duty plastic baskets of 90 dm<sup>3</sup> volume and 0.35 m height. They are connected with PVC-U pipes of 3/4" diameter. The catchment area, from which runoff flows to the experimental installation, is approximately 105 m<sup>2</sup>, the area of constructed wetlands (0.58 m<sup>2</sup>) is 0.55% of their catchment, which is relevant to the design guidelines [10, 19] for such objects defining the value of this fraction from 0.5% to 2.0%.

The installation was constructed between 10 and 22 May, 2010, and the reed cuttings were planted on 22 May, 2010. The experimental installation functions in a semi-technical scale, it is open all the time, not only for the periods of experiments, and when it rains, stormwater flows through the installation and is treated by it.

## METHODOLOGY AND TAH MEASUREMENT

The experiments were conducted on a semi-technical scale constructed wetland installation, which is described above. Five series of experiments were conducted on 30 May; 1, 12, 20 and 24 June, 2010. Every time two kinds of samples were taken: 1) influent to the sedimentation tank, and 2) effluent from the final tank. The samples taking process was conducted with respect of retention time (calculated from the flow rate, which is described below). On each of the experimental days, two such pairs of samples were taken. Prior to their chemical analysis, the samples were stored in tightly closed dark-glass bottles in a fridge, not longer than 48 h.

The rate of stormwater flow through the installation was also measured during each sample's taking. The precipitation intensity was also determined for each experiment, based on data measured by the meteorological station, which is situated in a distance of approximately 300 m from the experimental installation (Balice airport) and operated by IMGW (Institute of Meteorology and Water Management).

The main parameter monitored in samples taken from the experimental installation was the concentration of total C7-C30 aliphatic hydrocarbons (TAH). The concentrations of single aliphatic hydrocarbons (these with the even number of carbon atoms in a molecule) were measured too. These compounds were analyzed by the gas chromatograph with the mass spectrometer detector (GC-MS) Trace Ultra and DSQ-II by the Thermo, with helium as carrier gas (flow rate 0.6 ml/min). The Rxi<sup>TM</sup>-5ms capillary column by Restek was used (film thickness 0.5 μm; column length 30 m; column diameter 0.25 mm).

The aliphatic hydrocarbons were extracted with the liquid-liquid method with n-pentane, due to the Polish Standard PN-C-04643 and analyzed with GC-MS. The analysis of one sample took 90 minutes: the column was heated from 35°C (6 min) to 130°C (0 min) with the temperature rate of 8°C/min, from 130°C (0 min) to 250°C (0 min) with

the temperature rate of 5°C/min, from 250°C (0 min) to 325°C (0 min) with the temperature rate of 1°C/min and from 325°C (0 min) to 335°C (10 min).

## RESULTS AND DISCUSSION

The results of individual aromatic hydrocarbons concentrations have been presented in Table 1, the values for both influent (IN) and effluent (OUT) have been presented there.

Table 1. The average concentrations of the individual aromatic hydrocarbons in the analyzed samples

DAY	SAMPLE	Concentration of aliphatic hydrocarbons [ $\mu\text{g}/\text{dm}^3$ ]											
		$C_8$	$C_{10}$	$C_{12}$	$C_{14}$	$C_{16}$	$C_{18}$	$C_{20}$	$C_{22}$	$C_{24}$	$C_{26}$	$C_{28}$	$C_{30}$
30 May	IN	nd	1.33	24.95	60.69	34.15	29.78	30.44	28.50	29.76	41.23	48.99	51.80
	OUT	nd	0.46	10.37	26.52	16.07	11.04	4.89	4.29	nd	nd	nd	nd
1 June	IN	nd	3.05	4.13	11.70	12.04	3.64	1.93	1.06	6.41	1.94	2.11	nd
	OUT	nd	1.58	0.77	8.86	8.91	2.66	0.99	nd	nd	nd	nd	nd
12 June	IN	71.64	60.31	304.43	752.51	689.10	550.80	268.93	107.47	75.37	75.78	63.28	69.06
	OUT	27.38	44.33	217.68	600.48	666.17	549.55	266.57	63.93	26.03	18.36	11.52	13.58
20 June	IN	63.84	52.17	70.67	798.24	474.94	390.60	398.47	354.07	188.43	95.96	86.01	88.14
	OUT	15.76	19.00	64.29	717.02	32nd	151.55	70.82	38.86	20.56	9.29	7.00	4.93
24 June	IN	18.24	83.10	155.30	399.30	402.66	335.09	260.53	114.19	106.60	94.59	69.06	79.44
	OUT	6.24	34.02	124.56	316.73	302.77	310.76	158.39	45.09	11.53	5.75	4.79	5.84

nd – not detected

Each single cell in the table contains an average of analytical results of two samples taken during the experiments.

Table 2 presents the values of the hydraulic and meteorological parameters, which were measured in addition to the main parameters.

Table 2. Rates of stormwater flow in experimental constructed wetlands

	Flow rate [ $\text{dm}^3/\text{s}$ ]	Precipitation intensity [ $\text{mm}/\text{h}$ ]
30 May 2010	2.10	7.06
1 June 2010	0.05	0.15
12 June 2010	0.92	3.07
20 June 2010	0.33	1.10
24 June 2010	0.76	2.55

For each experiment, an average value of stormwater flow rate through the installation have been presented, as well as the average precipitation intensity for a period of samples taking. Despite the fact that the experimental constructed wetlands installation was designed regarding the guidelines and its area was 0.5% of the total catchment, it was overloaded with stormwater – the values of flow rates were far too high. The conclusion is that in Polish conditions this fraction has to be higher than 0.5%, thus the installation is going to be extended, two additional constructed wetlands beds are going to be added before the next phase of experiments.

Figure 2 presents the values of TAH in the samples of influent (IN) and effluent (OUT) from the experimental installation.

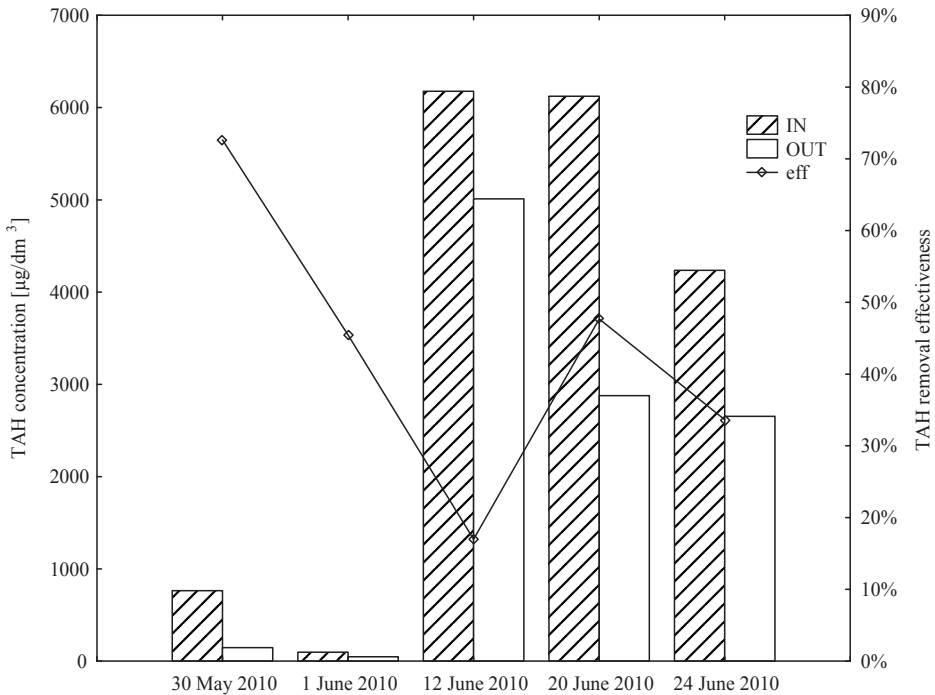


Figure 2. The total aliphatic hydrocarbons concentration in samples of stormwater from the experimental installation (IN – influent, OUT – effluent) and TAH removal effectiveness (eff)

There are also values of TAH removal efficiency presented in this Figure. TAH concentrations vary significantly in the samples from particular experimental days. The maximum TAH level in stormwater influent was observed on 20 June 2010 and was slightly higher than  $6100 \mu\text{g}/\text{dm}^3$ ; the lowest one was observed on 1 June 2010 and was close to  $100 \mu\text{g}/\text{dm}^3$ . In all samples (both in influent and effluent) TAH concentrations were lower than Polish standard, which is defined as  $15 \text{ mg}/\text{dm}^3$  [15, 14]. In all experiments, TAH concentrations observed in effluent were lower than these observed in influent, however TAH removal efficiency was different for particular experimental days, it varied from the level of 19% (12 June 2010) up to 81% (30 May 2010). Its average value was 49% and

it was lower than a value expected basing on the results of the pot experiments, in which the observed levels of this efficiency were above 98% [1, 2, 4]. It is also lower than values reported from other studies, in which the efficiency between 70% and 90% was observed [13, 21] both in laboratory and field experiments.

Figures 3–5 concern the individual aliphatic hydrocarbons, Figure 3 presents their concentrations in samples taken from influent to the experimental installation, Figure 4 – the same parameters for effluent, Figure 5 – the effectiveness of TAH removal by the tested installation.

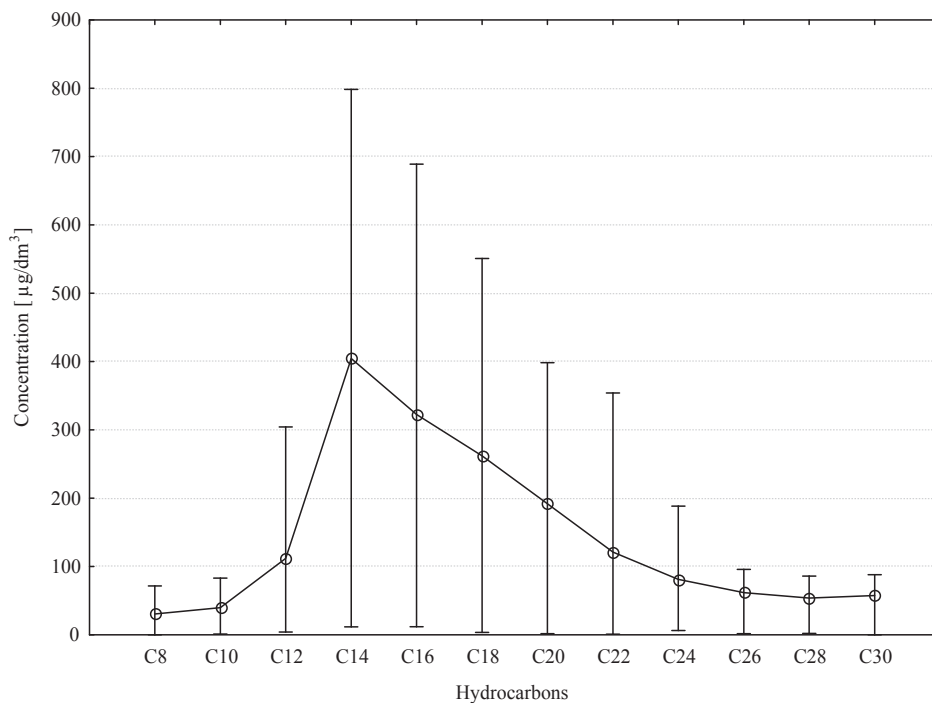


Figure 3. The concentration of individual aliphatic hydrocarbons in stormwater – an influent to the experimental installation (average, minimum and maximum values are presented on the graph)

For all of these Figures the same manner of presenting data has been used – there are average, minimum and maximum values presented. In the analyzed stormwater samples, the highest concentrations have been observed for alkanes with the carbon atoms number between 14 and 20, whereas the lowest – for the lightest hydrocarbons. The removal effectiveness for these substances shows the opposite tendency, the hydrocarbons C14 to C18 were removed with the lowest degree (26%–32%), the lighter hydrocarbons were better removed (39%–68%), however the highest removal effectiveness were observed for the hydrocarbons with the highest carbon atoms numbers (from 51% for C20 to 92%–93% for C26–C30).

It is difficult to define the strong relations between the initial concentration of pollutants and their removal effectiveness, as well as between it and the stormwater flow rate. There are several possible reasons for that difficulty, one of them is the unusually

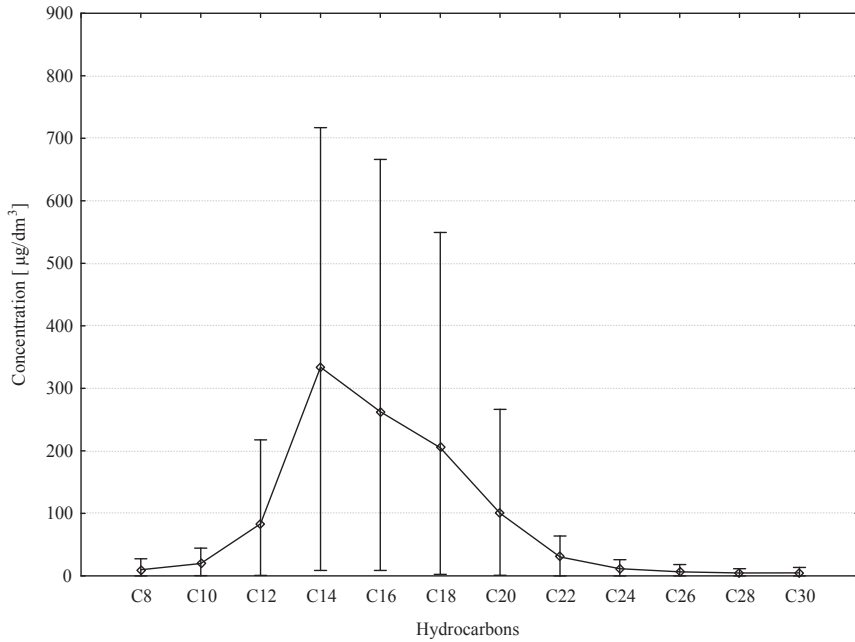


Figure 4. The concentration of individual aliphatic hydrocarbons in stormwater – an effluent from the experimental installation (average, minimum and maximum values are presented on the graph).

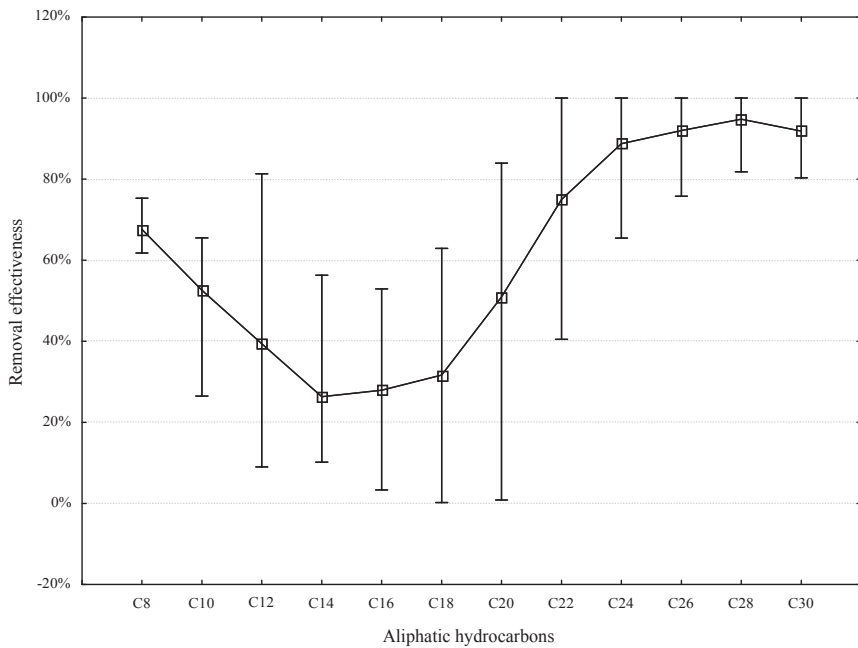


Figure 5. The effectiveness of individual aliphatic hydrocarbons removal by the experimental installation (average, minimum and maximum values are presented on the graph)

high precipitation intensity in months when experiments were conducted, this resulted in a high amount of run-off stormwater flowing through the system and their relatively low pollution. The other possible reason is the fact that it was the first season of these field experiments, thus the reed population and the constructed wetland microcosms had too short time to mature and develop. This research is going to be continued in next seasons and these mechanisms and relations are going to be further tested and explained.

During and after experiments, any kind of negative changes in the reed population have not been observed. Plants were resistant to the oil-derivatives influence, generally to the conditions of the experimental installation and to the constant influence of stormwater from the filling station.

## CONCLUSIONS

The research experiments have proved the potential of constructed wetlands to treat stormwater and to protect the environment against oil-derivatives, which are consisted in this kind of wastewater.

The TAH concentrations observed in stormwater from the filling station ranged from 96.02  $\mu\text{g}/\text{dm}^3$  to 6177.33  $\mu\text{g}/\text{dm}^3$  in influent to the experimental installation, and from 47.55  $\mu\text{g}/\text{dm}^3$  to 5011.14  $\mu\text{g}/\text{dm}^3$  in effluent from it. The average TAH removal effectiveness was 48%, the particular values ranged from 19% to 81%.

All tested stormwater samples showed a similar distribution of the individual aliphatic hydrocarbons; the alkanes with the carbon atoms number between 14 and 20 dominated, ones with the higher number of the carbon atoms had relatively lower share, and the lightest alkanes were the smallest fraction. Hydrocarbons C14 to C18 were removed with the lowest effectiveness (26%–32%), the lighter hydrocarbons – with higher one (39%–68%), however the highest removal effectiveness was observed for the hydrocarbons with the highest carbon atoms numbers (from 51% for C20 to 92%–93% for C26–C30).

Reed *Phragmites australis* is a plant resistant to oil derivatives influence – any kind of negative changes have not been observed during the experiments.

The constructed wetlands are the promising alternative for the conventional treatment methods, and they should be considered as an valuable option in designing and constructing the systems to treat motorways and parking lot runoff contaminated with oil-derivatives. There is an urgent need to form the design guidelines adopted to Polish situation, which would allow designers to plan and construct such utilities on technical scale.

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#### EFEKTYWNOŚĆ USUWANIA ZWIĄZKÓW ROPOPOCHODNYCH ZE ŚCIEKÓW DESZCZOWYCH PRZEZ EKSPERYMENTALNE ZŁOŻA HYDROFITOWE W SKALI PÓLTECHNICZNEJ

W artykule przedstawiono wyniki badań nad usuwaniem węglowodorów alifatycznych (C7 do C30) przez złożo hydrofitowe. Badania zostały przeprowadzone z wykorzystaniem oczyszczalni hydrofitowej obsadzonej trzcina pospolitą *Phragmites Australis*. Oczyszczalnia w półtechnicznej skali została zbudowana w okolicy

stacji benzynowej w Balicach i oczyszcza część ścieków deszczowych powstających na jej terenie. Obserwowane wartości stężeń sumy węglowodorów alifatycznych w ściekach deszczowych wynosiły od 96,02  $\mu\text{g}/\text{dm}^3$  do 6177,33  $\mu\text{g}/\text{dm}^3$ , a w ściekach oczyszczonych od 47,55  $\mu\text{g}/\text{dm}^3$  do 5011,14  $\mu\text{g}/\text{dm}^3$ . Średnia efektywność usuwania sumy węglowodorów alifatycznych wynosiła 48%, poszczególne wartości zmieniały się od 19% do 81%. Alkany od C14 do C18 były usuwane z najniższą skutecznością (26%–32%), lżejsze węglowodory – z wyższą (39%–68%), jednak najwyższą efektywność zaobserwowano dla węglowodorów z największą liczbą atomów węgla w cząsteczce (od 51% dla C20 do 92%–93% dla C26–C30).