



Semi-technical scale research on constructed wetland removal of aliphatic hydrocarbons C7–C40 from wastewater from a car service station

Tomasz Bergier*, Agnieszka Włodyka-Bergier

Faculty of Mining Surveying and Environmental Engineering, Department of Environmental Management and Protection, AGH University of Science and Technology, al. Mickiewicza 30, Kraków 30-059, Poland, Tel. +48 12 617 47 57; Fax: +48 12 617 50 76; email: tbergier@agh.edu.pl (T. Bergier)

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ABSTRACT

Dynamic development of road infrastructure and a significant increase in the number of cars are important challenges making it necessary to rationally treat and manage stormwater polluted with oil derivatives. Ecological engineering can be a source of valuable solutions in this domain, which are in agreement with sustainability principles, friendly to environment and hydrological cycle, convenient and attractive to the users. The article presents the results of research on using constructed wetlands to remove oil derivatives, which have been conducted with the semi-technical scale experimental installation planted with *Phragmites australis*, treating stormwater from a car service station in Sosnowiec. The results and discussion focus on the concentration of individual aliphatic hydrocarbons, their sum, as well as the efficiency of their removal by the studied installation. Some additional parameters of analysed wastewater such as temperature, suspended solids, pH, conductivity and air temperature have been also presented and discussed. The condition of the reed population has been also evaluated and discussed. The concentration of total aliphatic hydrocarbons (C7–C40) in raw wastewater was on average 2,311.67 µg/L (121.08–17,664.02 µg/L) and much lower in treated wastewater –261.81 µg/L (77.09–532.56 µg/L). All measured hydrocarbons concentrations in wastewater flowing out of the experimental installation were much lower than the standard value, defined by Polish law for wastewater released to the environment or to the sewage system. Some relatively high values of the aliphatic hydrocarbons removal rate were also observed (averagely 67% for C7–C40 and 64% for C7–C30). The analysis of research results in relation to individual aliphatic hydrocarbons showed that most efficiently removed ones were hydrocarbon C20–C33 (on average around 80%), C34–C40 and C12–C19 with slightly lower efficiency (60–70%), and least efficiently—lightest hydrocarbons C7–C11. Common reed *P. australis* confirmed its high applicability in constructed wetlands which treat stormwater polluted with oil derivatives. Generally, the experiments confirm high potential of constructed wetlands to remove oil derivatives from wastewater.

*Corresponding author.

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1. Introduction

Searching for a rational method of stormwater treatment is one of the most urgent challenges of water management both in Poland [1] and worldwide [2,3]. As a result of intensive development of urban areas and surface sealing of urban catchments, a significant increase in the quantity of stormwater has been observed recently [4,5]. It is increasingly important to find a rational solution. Mixing stormwater with domestic sewage, which is very common in Poland, and then treating them in municipal wastewater treatment plants is an irrational solution since it causes many operating problems, environmental threats and negative changes in a water cycle [4,6].

The described problem is especially complicated in the case of run-off from the roads and car parking lots [3,7]. Due to the presence of specific pollutants, including petroleum compounds, it is necessary to pre-treat such a kind of wastewater before its releasing to the sewer system or environment. Disposing stormwater from urban catchments on an increasingly large scale brings some significant changes in a hydrological cycle; local retention is compromised; the amount of water in the local water cycle decreases; there are significant fluctuations in surface waters drainage. All of these phenomena contribute to the occurrence of extreme events (floods, draughts) [8,9].

In the countries where road infrastructure is well developed, the ecological engineering solutions are used to manage stormwater from such infrastructure [10,11]. In US, Scandinavia and many other countries constructed wetlands are used on a large scale for this purpose [2,12,13]. Due to their ability to treat stormwater *in situ*, they minimize costs and negative environmental effects caused by the management of that kind of wastewater. The most important factors which contribute to the growing popularity of constructed wetlands are high immunity of vascular plants to changeable hydrological conditions and high concentration of pollutants [14], including petroleum compounds [7]; high activity and diversity of wetland micro-organisms [15]; relatively easy and inexpensive maintenance; additional benefits resulting from their application (an increase in biodiversity, the aesthetic aspect). The hydrological aspect of this solution is also important. The local retention of stormwater increases its evapotranspiration and groundwater discharge, decreases the water run-off rate [1,16]. Thus, the

natural local hydrological cycle is restored, and local microclimate conditions are improved.

The authors of this article for many years have been studying the possibilities of applying constructed wetlands to treat stormwater polluted with oil derivatives. In the first phase of the research, they proved the high capability of constructed wetlands to remove oil derivatives in pot experiments [17–19], as well as high immunity of macrophytes (especially common reed) for this type of pollutants. The results of pot experiments and gained experience were used to design semi-technical scale experiments. They were carried out simultaneously on two experimental installations, which represent the most common sources of petroleum pollutants in Polish conditions. The first one was the experimental constructed wetland, which since 2010 had been treating a part of stormwater from the petrol station in Balice. A detailed description of this installation as well as the results of experiments and observations conducted, there were described in several publications [20–22].

The other experimental installation in semi-technical scale has been operating since July 2012 in Sosnowiec and treating stormwater from a car service station. The aim of this study was to describe and characterize this installation in the initial period of its operation. It is realized by presenting the concentrations of aliphatic hydrocarbons (C7–C40), observed in the wastewater flowing through this installation, as well as their efficiency of removal. In the study, the results of other parameters characterizing the analysed wastewater are presented and discussed, including: temperature, suspension solids, pH, conductivity.

2. Methods

2.1. Experimental installation

The semi-scale experimental constructed wetland installation is operating in a car service station in Sosnowiec. The parking lots of around 200 m², where the repaired cars are parking along with the staff and customers' cars, are equipped with the collecting system, which directs stormwater to the first tank of the experimental installation. Besides stormwater, the small amount of wastewater from the inside of the service station (mainly everyday washing of a floor) is also directed to the installation.

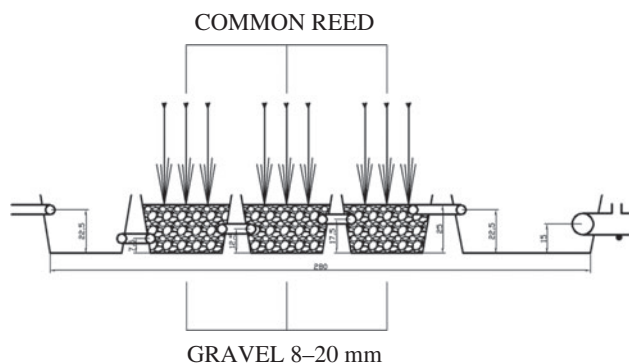


Fig. 1. The scheme of the experimental constructed wetland installation.

The installation (Fig. 1) consists of 5 tanks, which are identical rectangular plastic containers of 80 L capacity each. The first tank plays a role of a sedimentation and retention tank. Other three tanks constitute the constructed wetland part of the installation. They are connected to the first tank in series, and their filling is gravel (8–20 mm). In June 2012, the settlings of common reed (*Phragmites australis*) were planted on these three tanks. The decision to use common reed in the experiments was made based on the previous research and experience [17–19], and especially, its low sensitivity to the presence of oil derivatives. Treated stormwater, coming from the third tank of the constructed wetland part, is directed to the last tank. The area of each tank is about 0.4 m². Thus, the area of the whole installation is about 2 m², and its constructed wetland part—1.2 m². The described installation constitutes about 1% of the catchment it serves, which is consistent with the guiding principles of designing such types of sewage treatment plants, according to which this ratio should be 0.5–2.0% [12,13]. Stormwater is transported to the first tank of the installation through a PVC 110 mm pipe, and the connections inside the installation, between the tanks, are made of PVC 50 mm pipes.

Taking into the consideration the rate with which stormwater flows through this installation (see Section 3.2), the average retention time was 36 h for the whole installation, and 12 h for its wetland part. The average hydraulic loading rate was, respectively, 0.075 m/d and 0.125 m/d. However, due to the high variability of precipitation, the real values of retention time and the hydraulic loading rate could have been significantly different from the average values, and also highly changeable during the research period. The average hydrocarbons loading rate was 209,960 µg/d/m² (from 12,777 to 1,281,032 µg/d/m²) for hydrocarbons C7–C40, and 185 354 µg/d/m² (5,496–

1,219,202 µg/d/m²) for C7–C30. The hydrocarbons loading rates have been calculated only for the constructed wetland part of the experimental installation.

2.2. Experimental procedure

The samples from the installation described in the previous section were regularly taken from September 2012 to February 2013. The research period was chosen this way to compare the performance of constructed wetlands treating stormwater in warm and cold months, thereby to assess the possibility to apply this technology in Polish conditions.

Taking of samples was conducted every fortnightly, which made 11 measurement days (2 in each month, except for December, when there was only one measurement day). On each measurement day, two pairs of samples were taken: (1) raw wastewater (from the first tank of the installation) and (2) treated wastewater (from the last tank). In the research period described in this article, totally 42 samples were collected and analysed, among which 22 samples of raw wastewater, and 20 of treated wastewater (on 24 January 2013, it was impossible to collect treated wastewater samples because water in the last tank was completely frozen). Several additional parameters were measured during each experimental day. Before and after each sampling, the wastewater flow rate through experimental installation was determined (the amount of stormwater inflowing the installation's first tank was measured per time unit). The temperature of raw and treated wastewater, as well as the air temperature was also measured. For each measurement day, meteorological data were also collected, especially precipitation intensity and air temperature. These data were obtained from the Silesian University's meteorological station, located in Sosnowiec at Bedzinska Street, around 300 m far from the experimental installation.

2.3. Analytical methods

In all samples, the following parameters were analysed: the concentration of all individual aliphatic hydrocarbons from the range C7 to C40, total aliphatic hydrocarbons from C7 to C30 (TAH C7–C30), total aliphatic hydrocarbons from C7 to C40 (TAH C7–C40), suspended solids, pH and conductivity. The extraction of aliphatic hydrocarbons was conducted with a liquid–liquid method with n-pentane used as a solvent. Then, they were analysed with a gas chromatograph with a mass spectrometer (GC-MS)—Trace Ultra DSQ-II by Thermo. For the purpose of this research, capillary

column RxiTM-5 ms by Restek company was used (film thickness 0.5 µm; column length 30 m; diameter 0.25 mm) as well as helium as a carrier gas (flow 0.6 ml/min). The analysis of each sample lasted 90 min and the following temperature programme was used. The column was heated from 35°C (6 min) to 130°C (0 min) with a temperature increase 8°C/min, and then from 130°C (0 min) to 250°C (0 min) with a temperature increase 5°C/min and from 250°C (0 min) to 335°C (10 min) with a temperature increase 1°C/min. The detection limit for each individual analysed hydrocarbon was 0.02 µg/L. Both extraction and the analysis of aliphatic hydrocarbons were conducted according to the Polish Norm PN-C-04643: 1994. Suspended solids were analysed according to the Polish Norm PN-EN 872, conductivity—according to PN-EN-27888 and pH—PN-90/C-04540/01.

The presented experiments focused on the efficiency of hydrocarbons removal, and therefore, their concentration in raw and treated stormwater was measured. The hydrocarbons dynamic was not fully studied, that is their concentration in the gravel and in the plants was not measured.

3. Results and discussion

3.1. Total aliphatic hydrocarbons

The research results of total aliphatic hydrocarbons concentration are presented in Table 1. Average, minimum and maximum values are presented there, as well as medians for TAH C7–C40 and TAH C7–C30, both in raw and treated wastewater. Analogical sets of

Table 1
Total aliphatic hydrocarbons concentration and their removal efficiency in the experimental constructed wetland installation

	TAH C7–C40 (µg/L)	TAH C7–C30 (µg/L)
<i>Raw wastewater (n = 22)</i>		
Average	2,311.67	2,039.60
Median	1,268.87	905.93
Minimum	121.08	45.49
Maximum	17,664.02	17,398.51
<i>Treated wastewater (n = 20)</i>		
Average	261.81	197.20
Median	276.05	220.13
Minimum	77.09	32.96
Maximum	532.56	705.91
<i>Removal efficiency (n = 20)</i>		
Average	67%	64%
Median	69%	71%
Minimum	36%	26%
Maximum	98%	98%

statistical data were also presented for the removal efficiency of these two parameters.

As can be observed in Table 1, aliphatic hydrocarbons concentration in raw wastewater from analysed installation was characterized by very high changeability and relatively high values. In the samples from 21 February 2013, the highest concentration in raw wastewater was observed: for TAH C7–C30, it was 17,398 and 17,664 µg/L in the case of TAH C7–C40. It means that the concentration level of 15,000 µg/L was exceeded, which is the maximum concentration defined by the Polish law for wastewater disposed both to the municipal sewage system [23] and to environment [24]. It confirms the necessity to pre-treat such a type of wastewater. The values of the remaining parameters were lower than the standard ones; however, they were still relatively high. To compare, during analogical research carried out for wastewater from the petrol station, the maximum values were observed from 861.3 [20] to 6,177.3 µg/L [21], depending on the research period. When compared with the values reported in the general literature of the subject, the values observed during presented research in stormwater were similar to those observed in a run-off car park and on highways [3,25].

Aliphatic hydrocarbons concentrations in treated wastewater were significantly lower than in raw wastewater, and they also had much lower changeability. All measured TAH values were also much below Polish standards described in the previous paragraph.

Positive rating of the experimental installation operation effects were confirmed by relatively high removal efficiency of these parameters, which for C7–C30 was on average 67% (in several cases it was higher than 90%). For comparison, the average removal efficiency rates, observed during similar experiments conducted at the petrol station, were within 44–48% [20,21]. Thus, those values were significantly lower than the results reported in this study. Generally, in the literature, the hydrocarbon removal rates of up to 90% were reported [26]; however, there are a limited number of articles on this subject available in which a wide range of observed values of hydrocarbons removal rates can be found in them.

3.2. Additional parameters

As it was described in Section 2.3, apart from aliphatic hydrocarbons concentrations, some other parameters such as temperature, suspended solids, pH and conductivity were also measured in the samples. The average, minimum and maximum values of

Table 2

The basic wastewater parameters in the experimental constructed wetlands installation

	Wastewater temperature (°C)	pH	Conductivity (mS/cm)	Suspended solids (mg/L)
<i>Raw wastewater (n = 22)</i>				
Average	2.5	7.3	1.082	368.16
Median	0.0	7.2	1.024	50.32
Minimum	0.0	6.6	0.116	5.00
Maximum	13.4	8.1	2.930	2995.83
<i>Treated wastewater (n = 20)</i>				
Average	1.9	6.4	0.846	27.93
Median	0.0	6.1	0.911	28.61
Minimum	0.0	5.6	0.126	4.35
Maximum	13.7	7.3	1.201	66.25

these parameters as well as their medians were presented in Table 2.

The research was conducted in difficult atmospheric conditions—on most measurement days, the wastewater temperature was 0°C, and the air temperature—below zero. In the whole described research period, only on one day, the temperature was over 10°C. Despite such low temperatures, relatively high levels of oil derivatives removal were observed, as described in the previous section. The efficiency of constructed wetland treatment strongly depends on temperature [12,15], and therefore, even higher efficiency can be expected in warmer periods. The verification of this thesis will be possible after conducting a full research cycle to be continued on the described installation.

The range of wastewater flow rates through the experimental installation was 50–250 L/d. Due to the fact that the research was mainly conducted in winter, with air temperatures below zero, and the measured flow rate was relatively low and stable, it was decided not to include its detailed results in this article.

The suspended solids concentration demonstrated similar dependences as TAH, described in the previous section. In raw wastewater, high changeability of suspended solids was observed, and a few samples had very high levels of this parameter. In the case of five measurement days, in both samples of raw wastewater, the concentration of total suspension exceeded the level of 100 mg/L, which is the Polish standard for stormwater released to the environment [24]. The levels of suspended solids in treated wastewater were much lower than in raw wastewater (in all cases below the standard), and they also had much lower changeability.

The values of conductivity in raw wastewater also showed much higher changeability than these in treated wastewater. The decrease in this parameter was also observed, as a result of wastewater flow through

the experimental installation; however, the efficiency of the parameter removal was relatively small. The values of pH in raw wastewater were close to neutral—wastewater did not demonstrate any environmental risk in this respect. However, the fact that wastewater pH decreased significantly as a result of treatment by the experimental constructed wetland installation is unexpected and hard to explain.

3.3. Individual hydrocarbons

Fig. 2 shows concentrations of individual aliphatic hydrocarbons from the analysed group (C7–C40) in raw wastewater, Fig. 3—in treated wastewater, and Fig. 4—their removal efficiency by the experimental constructed wetland installation. The average, minimum and maximum values of each analysed parameter were presented in the corresponding figure.

The comparison of graphs in Figs. 2 and 3 allows one to observe some significant differences in the hydrocarbons concentration between raw and treated wastewater. It is in line with observations for total hydrocarbons concentrations, which were discussed in Section 3.1 of this article. The distribution of analysed hydrocarbons differs significantly between raw and treated wastewater. In raw wastewater, the largest share belongs to aliphatic hydrocarbons from C12 to C20, and relatively smaller is the number of the lightest hydrocarbons (C7–C11), even smaller of hydrocarbons C21–C34 and the smallest of the heaviest hydrocarbons (C35–C40). In treated wastewater, however, definitely the most numerous are the lightest hydrocarbons (C7–C10), and there is fewer hydrocarbons of slightly higher number of carbon atoms in a particle (especially C12–C13, C15) and the heaviest hydrocarbons (C39–C40). In particular, the last phenomenon is surprising since the hydrocarbons with largest particles should be most efficiently retained in

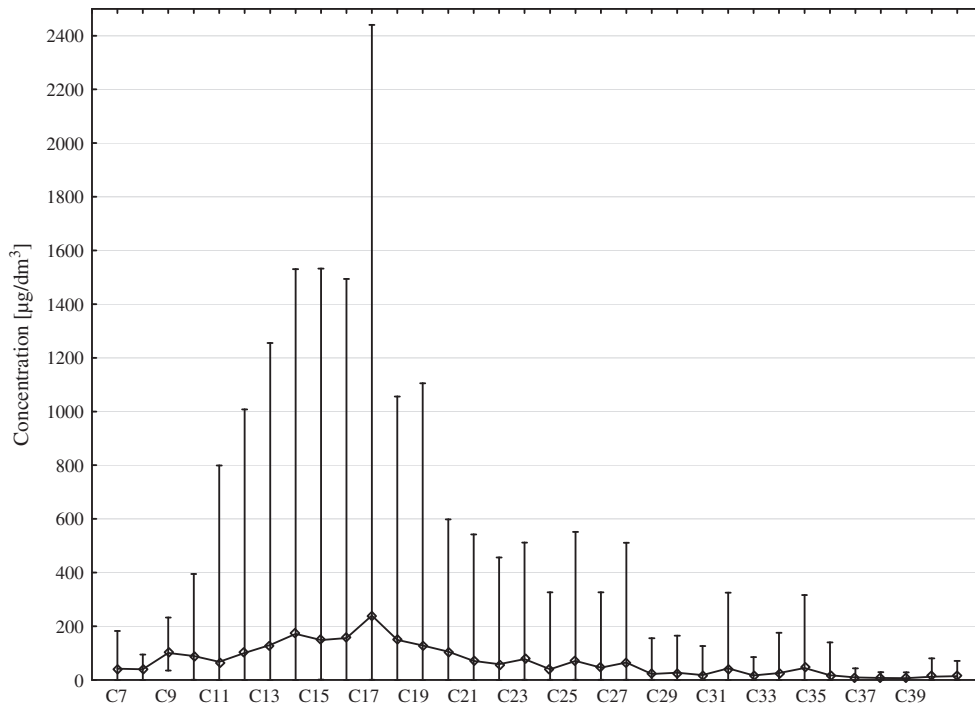


Fig. 2. The concentrations of individual aliphatic hydrocarbons in raw wastewater (average, minimum and maximum values are presented).

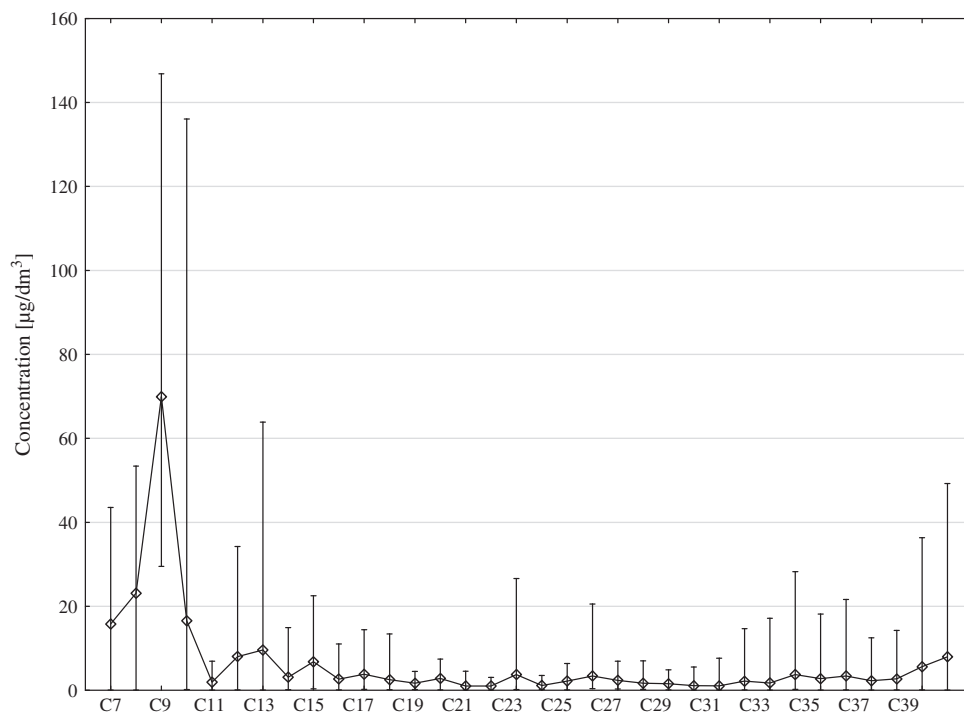


Fig. 3. The concentrations of individual aliphatic hydrocarbons in treated wastewater (average, minimum and maximum values are presented).

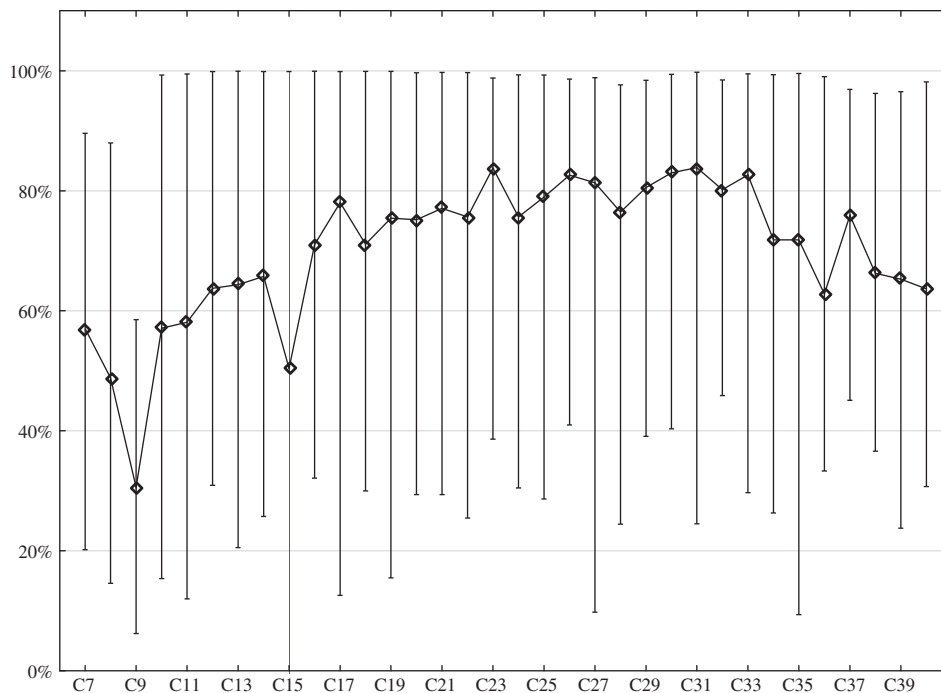


Fig. 4. The effectiveness of individual aliphatic hydrocarbons removal in the experimental constructed wetland installation (average, minimum and maximum values are presented).

constructed wetlands. High concentrations of lightest hydrocarbons are also quite surprising, in this case, however, it could be expected that the efficiency of their removal will be higher in warmer periods, when the temperatures of wastewater and air are higher and therefore these compounds, as more volatile, are more easily removed while flowing through constructed wetlands.

Fig. 4 complements and confirms the above observations—generally, hydrocarbons from the middle of the analysed group were most efficiently removed, especially the ones with the number of carbon atoms in the particle from C20 to C33, for which the removal rate is around 80%. The removal efficiency dropped with an increase as well as a decrease in the number of carbon atoms in hydrocarbon particle. For heavier hydrocarbons (C34–C40), the removal rate dropped to 60–70%. Similar values were observed for lighter hydrocarbons C12–C19 (except for C15, for which this parameter does not exceed 50%). Lightest hydrocarbons (C7–C11) are the least efficiently removed. As can be observed in Fig. 4, the values of removal efficiency parameter of individual hydrocarbons are very changeable. It results, most of all, from high changeability of hydrocarbon concentrations recorded for raw wastewater.

4. Conclusions

The concentrations of total aliphatic hydrocarbons in raw wastewater, obtained as a result of the research carried out, were on average 2,311.67 $\mu\text{g/L}$ (121.08–17 664.02 $\mu\text{g/L}$) for TAH C7–C40 and 2,039.60 $\mu\text{g/L}$ (45.49–17 398.51 $\mu\text{g/L}$) for TAH C7–C30. In treated wastewater on the studied experimental constructed wetland installation, these concentrations were much lower, respectively, 261.81 $\mu\text{g/L}$ (77.09–532.56 $\mu\text{g/L}$) and 197.20 $\mu\text{g/L}$ (32.96–705.91 $\mu\text{g/L}$). All measured TAH concentrations in wastewater flowing out of the experimental installation were much below the standard value, defined by the Polish law for wastewater released to the environment or to the sewage system.

Good efficiency of oil derivatives removal from wastewater by the examined constructed wetland is also confirmed by relatively high values of the removal rate of these compounds, which were on average 67% for TAH C7–C40 and 64% for TAH C7–C30. The analysis of research results in relation to individual aliphatic hydrocarbons showed that the ones most efficiently removed were hydrocarbon C20–C33 (on average around 80%), C34–C40 and C12–C19 with slightly lower efficiency (60–70%), and least efficiently—lightest hydrocarbons C7–C11.

Common reed *P. australis* confirmed its high applicability in constructed wetlands which treat stormwater polluted with oil derivatives. Despite the constant exposure to relatively high concentrations of these compounds in wastewater flowing through the surface and root part of plants, no negative changes or dying were observed in their state and condition. Based on the presented research results, it is impossible to determine the ratio of hydrocarbons removed directly by macrophytes (the TAH concentration was analysed in the influent and effluent, not in gravel filling nor plant tissue). However, the most important fact is that they survived in the experimental conditions and played an important role by creating the habitat for micro-organisms whose activity is commonly reported as the main mechanism of hydrocarbons removal in the constructed wetland [2,15,27].

The results and observations from the described research confirm high potential of constructed wetlands to remove oil derivatives from wastewater. They indicate that constructed wetlands are a valuable alternative to other solutions, and they should be considered in planning and designing systems and equipment used to manage wastewater polluted with oil derivatives.

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