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The suitability of natural and synthetic filter material for the removal of petroleum products from the aqueous media

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ABSTRACT

Petroleum products that have leaked into the natural environment are very hazardous. They are particularly dangerous for aquatic ecosystems. The major part of oils is carried into natural water bodies together with the surface outwashes from urban areas. One of the most inexpensive and easiest methods of the removal of oil contaminants from runoff is filtering. The authors of this study examined several synthetic substances (Duck and Reo-dry), and several vegetal materials (shredded stems of reeds, bulrush and lesser bulrush) and compared their filtration characteristics with the characteristics of the synthetic substance Fibroil now widely used for filtration. The authors of this study feel that the materials tested are suitable for the petroleum removal filter medium. Industrialists submit specifications of their products that were recorded under ideal conditions, at a low filtration rate. The authors studied filter material of natural origin as well as synthetic ones by simulating actual conditions that originate during heavy rains, i.e. at high filtration rates. In some aspects, low-cost vegetal material for the surface treatment of runoff performed better than the synthetic filters.

Keywords: Petroleum products; Removal; Filtration; Natural material

1. Introduction

Petroleum products (PP) that leak into the natural environment, cause significantly ecological harm: due to their toxicity, they impede the growth of plants and animals, pollute the environment and spoil its aesthetic appearance, and are difficult to remove and also takes a very long time to degrade in nature [1–3]. PP spilled onto the surface of the soil go deeper into subsequent soil layers, and contaminate the groundwater; thus today one of the most important environmental tasks is the suspension and reduction of the spread of PP in the geological environment and its subsequent pollution [3,4]. Petroleum contaminants usually get into water bodies through surface runoff, which can occur during accidental spills as well as normal rain or spring thaws when rainfall water washes away the hard surface coverings of urbanized areas. During strong rains, PP concentration in the surface runoff from parking areas, garages and petrol station areas can often amount to 50 mg/L or more [4–6]. It is important to prevent this pollution from

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spreading into the environment in a timely manner, in particular from getting into natural water bodies. To solve this problem oil traps are installed, which come in various forms, including settlement tanks, coalescent and sorption filters [7,8]. Environmentally, it is irrational to install oil traps everywhere where they are needed. They are expensive and take up space, and pumps are often needed for them. They must also be powered by electricity. Another concern is the unpredictability of the runoff. In the case of rainy periods, the flow of surface runoff is uneven. The pollutant concentrations of runoff are also constantly changing. There is also the issue of the traps themselves. A large amount of contaminated water gets into oil traps, and it can flow very quickly, which the oil traps may not be able to deal with. Another problem is that pollutants can fail to settle or be separated in the coalescent filter. Sorption filters can also get clogged. The simplest prevention method against PP pollution is the rapid filtration of contaminated water (from impervious surfaces), during which PP would be held in a filter filler. This is true for the countries which, such as Lithuania, are located in areas with high humidity (where there is an annual average rainfall of 650–750 mm). Heavy rains usually occur in the summer, but they do not last long, usually an hour and a half on average. Surface runoff pollution with oil (and other pollutants) that is generated during rainfall is the greatest at the beginning of the rainfall, thus it is of particular importance to clean up PP (or at least significantly reduce their concentration) and release them into open water bodies. Simple filtration columns with mediums that are replaced at regular intervals can be installed instead of expensive oil traps. This would also help to achieve the required treatment efficiency of oil-contaminated water and hydraulic conductivity [9,10]. It is desirable that the mediums of these kinds of filters that have filtered PP can be easily recoverable, as well as utilizable in various kinds of conditions, and their price would be lower than those commonly used in oil filters [11–15]. This article analyzes the suitability of using various materials for the removal of PP from surface runoff when filtration through them is carried out at relatively high rate (15 m/h), including patented sorbents available on the market such as Fibroil, Duck and Reo-dry, as well as plant-based materials available in Lithuania such as bulrush, cattails and reeds that could be used for PP filtration There is a variety of materials able to hold back the PP that was studied in the laboratory, but mostly low filtration rates are used: 0.3–0.4 m/h [16]; 0.5–1.5 m/h [13]; however, in reality the surface runoff is filtered at 10 m/h or more through the oil trap mediums during rain Therefore, the authors tested the selected filter mediums under the kinds of adverse conditions that occur in real life.

2. Methodology

2.1. Simulation of surface runoff flow

For carrying out the filtration tests, filtration equipment (shown in Fig. 1) was installed in the laboratory of VGTU's Department of Water Management.

Surface runoff scooped up from urban areas and delivered to the laboratory was poured into the first tank (1) and then supplied to a second tank (2) by a pump (12). A constant volume of liquid was kept in this tank so that the flow in the pipe (4) would be smoother. After turning on the valve installed at the bottom of the tank (2), the fluid flowed along a pipe (4) with a slope of 3˚. PP (diesel) was inserted into the fluid flowing out of the vessel (3) with a peristaltic pump at a rate where the initial (C_0) concentration of PP (there have been sampled) would form behind the screen (5) at the end of the pipe. Then the fluid was supplied to the cross-sectional area (0.005 m^2) of the filtering cylinder (6) at a continuous pace by means of the screen and filtered through a 20 cm medium layer. The filtration rate was regulated through controlling the opening of the valve and measuring the filtrate flow rates every 10 min. A filtration rate of 15 m/h (1.3 L/min) was chosen for the tests. For testing, the samples were poured into 0.5 L jars every 10 min before (inlet) and after (outlet) the filter cylinder operation, and the samples were measured for the level of contamination by PP and suspended sediment material (SM). The pressure losses that developed in the filtration cylinder were measured with a piezometer (10). Samples of surface runoffs from an impervious surface (paved roads in Vilnius during 2013 and 2014) were then brought to the laboratory. After the samples had been poured into a 100 L tank (Fig. 1), the PP and SM concentrations were measured. Runoff and diesel (PP) were added to the filtration plant by means of pumps at a constant flow rate, which allowed for the possibility to prepare mixtures with constant high PP concentrations (~0 mg/L). PP could not segregate onto the liquid surface; the formation of PP with high concentrations of real runoff was simulated. The filtration experiments lasted 210–220 min, which allowed the authors of this study to compare the characteristics of various fillers and the suitability of PP removal from the various kinds of surface runoff.

2.2. Filter mediums

Two types of materials were selected for experimental tests: (1) sorbents for PP absorption sold on

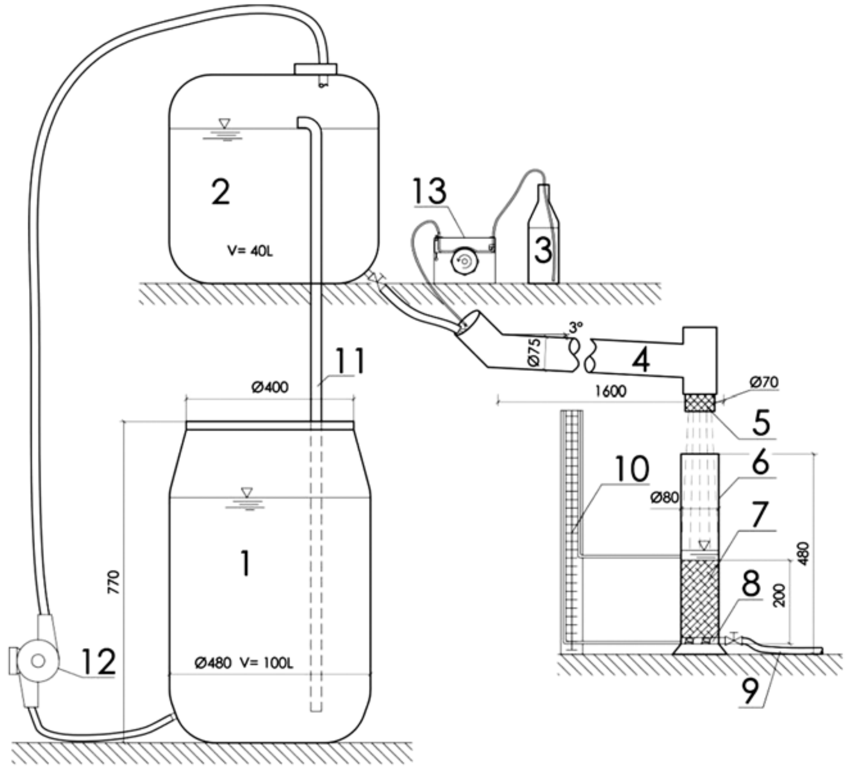

Fig. 1. Filtration equipment: 1—100-L tank for holding the runoff; 2—50-L tank; 3—vessel filled with diesel; 4—the pipe through which the runoff with injected PP is supplied to the filter; 5—screen; 6—filtration cylinder; 7—medium (filler); 8—rubble layer supporting filler; 9—flexible outlet hose for filtrate sampling; 10—piezometer; 11—overflow pipe; 12—pump; 13—pump for diesel dosing.

Fig. 2. Materials used as filter mediums: 1—Fibroil; 2—Duck; 3—Reo-dry; 4—reed stems (*Phragmites australis*); 5—bulrush stems (*Schoenoplectus lacustris*); 6—lesser bulrush stems (*Typha angustifolia*).

the market, and (2) plant stems, which are natural substances that are inexpensive, could keep PP in check, could be easily disposed of after their use, and would be environmentally friendly.

The first row of Fig. 2 shows the three materials (Fibroil, Duck and Reo-dry) that absorb PP well under static conditions. The sorption capacity of synthetic Fibroil is 8–14 g/g; the mineral-based Duck and cellulose-based Reo-dry absorb about 3–4 L/kg of diesel or oil (information that is indicated by the producers of these materials) [17]. Fibroil, Duck and Reo-dry are described as non-water absorbable and are used for the collection of liquid products that are lighter than water from the water surface. The second row (Fig. 2) shows natural vegetal materials: shredded stems of reeds, bulrushes and cattails. Before placing in the filter cylinder, plant stems were dried in the laboratory at room temperature (~20˚C) for two weeks and cut into pieces (1–2 cm).

The height of each medium in the cylinder was 20 cm. The sorbents sold on the market (Fig. 2, 1–3) were selected by the recommended overspread density of the sorbent. The weight of the Fibroil medium used in the experiment was 60, 113 g for Duck and 230 g for Reo-dry. The weight of the tested vegetal stems was determined by the volume it took up (which was lightly pressed together) and amounted to 100 ± 20 g. Each filtration experiment was carried out until the PP concentration in the filtrate reached 7 mg/L or until the filter became clogged. The blockage of the filter medium was reflected by the decrease in the filtrate flow rate and piezometer (Fig. 1) readings.

2.3. Analytical methods

Pollutant concentrations were calculated according to the formula (1). It is only possible to show the changes of the PP concentration in the samples, thus at present only the average concentration is presented in this study. The average concentration at typical points is the following:

$$C_{\text{aver.}} = \frac{1}{n} \cdot \sum_{i=1}^{k} \cdot c_i \cdot m_i \qquad (1)$$

PP removed from solution between any two sampling intervals:

$$q = (C_0 - C_2) \cdot (V_2 - V_1) \qquad (2)$$

Thus the amount of PP removed after n samples is:

$$q = (C_0 - C_1) \cdot (V_1 - V_0) + (C_0 - C_2) \cdot (V_2 - V_1) \cdots + (C_0 - C_n) \cdot (V_n - V_{(n-1)}) \qquad (3)$$

The concentration of suspended solids (SS) in runoff that had been brought in as samples and filtered were measured in the laboratory of VGTU Department of Water Management: runoff samples were filtered through glass fibre filters, and the sludge was dried to a constant weight at 105˚C temperature (LST EN 872:2005. Water quality. Determination of SS. Method by filtration through glass fibre filter).

The concentration of PP in the samples was measured at Eco-care laboratory, which is the laboratory of UAB Grinda a company specialized in rain sewage development that oversees the maintainance of Vilnius's streets. This laboratory is certified by the Environmental Protection Agency of the Republic of Lithuania. The concentration of PP was determined gravimetrically in accordance with the normative document LAND 90-2010 of the Republic of Lithuania. After the extraction of mineral oil from aqueous media with chloroform, the solvent was evaporated from the extract; the residue, dissolved in hexane, was passed through the chromatographic column filled with aluminium oxide-trapping polar hydrocarbons. The eluate was evaporated, and the residue was weighed.

3. Results and discussion

The pollution of collected surface runoff (composite sample) with PP and suspended SMs is shown in Table 1. After pouring these runoffs into the tank (1) (Fig. 1), part of the SS end up deposited on the bottom, while the PP rise to the liquid surface. However, after the insertion of the PP (Fig. 1, 3) into the pipe from the vessel, runoffs that had a similar amount of contamination formed behind the screen (inlet). The average indicators calculated according to the formula (1) are shown in Table 1.

At first, the hydraulic conductivity of the filter medium was tested and determined by the decreasing filtration rate. The results of the change of the following indicator in the course of the process are shown in Fig. 3.

As Fig. 3 shows, some of the filter mediums started clogging over time, which led to a decrease in the filtration rate. In terms of the speed that the mediums clogged, the Reo-dry medium clogged the quickest (filtration completed in 140 min); followed by the Duck medium (filtering completed in 170 min) and the cattail medium (filtration rate decreased to 7 m/h). The filtration through the Fibroil and reed mediums was similar. The reed mediums clogged marginally—the

Table 1
Average pollution rate of road runoff samples and influent concentration

	Road runoff		Influent concentration	
Sample taking points	[PP] (mg/L)	[SS] (mg/L)	[PP] (mg/L)	[SS] (mg/L)
1.	16.5	150	50.5	36.5
2.	20.1	176	52.0	42.4
3.	10.1	125	49.3	35.3
4.	2.0	145	51.2	39.8
5.	18.6	213	50.8	45.3
6.	20.7	167	50.5	42.7
7.	30.2	285	52.3	51.2

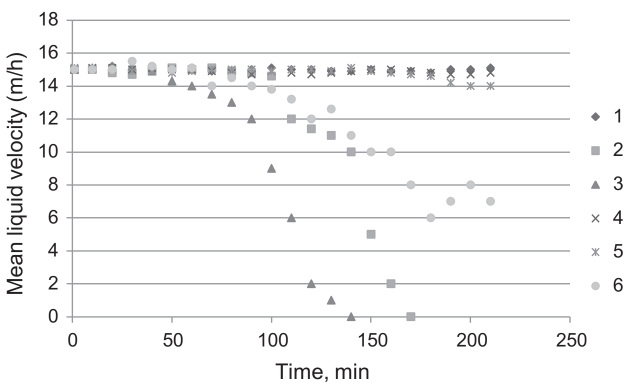

Fig. 3. Average liquid velocities, when C_0 ~50 mg/L. Series: 1—Fibroil; 2—Duck; 3—Reo-dry; 4—reed stems (*Phragmites australis*); 5—bulrush stems (*Schoenoplectus lacustris*); 6—lesser bulrush stems (*Typha angustifolia*).

filtration rate dropped to 14 m/h. Filtration is a compendium of complex phenomena, with the hydraulic resistance influenced by several factors: material density, sorption properties, hygroscopicity and the water absorption. The mediums clogged because they suspended PP and SM, which led to a decrease in the permeability of the filters. Research has shown that a reed medium is as good as a Fibroil medium in terms of its hydraulic conductivity; their hydraulic conductivity was the best from the six kind of material that was tested.

Fig. 4 shows the change of PP concentration in the filtrate samples over time. When the filter load was 65 mg PP/min, the PP concentration in all filtrate samples at the beginning of filtration (10 min after the start of the experiment) was less than 5 m/L.

The maximum permissible regulated average annual concentration of PP when releasing runoff directly into the environment is 5 mg/L (the maximum instantaneous concentration is 7 mg/L) [3]. Thus, all filter mediums are able to hold the PP and clean filtered runoffs to regulated indicators during the first 30 min. Over a period of time, the PP concentration in filtrate samples increased: primarily, the margin of 5 mg/L was exceeded in the cattail medium filtrate samples. Comparing the materials of the first medium and second medium it was observed that the first-type of filter mediums filtered the runoffs more effectively: an average of 2–6 mg/L of PP was left in filtrate samples (96–88% efficiency). Vegetal stems were less effective, with an average of 11–17 mg/L of PP was left in filtrate samples (78–66% efficiency). The difference in efficiency amounted to 18–22%. Surface runoff that is treated in this way can be released into the general sewage system, and then go into city treatment plants. In this case, the annual average concentration of PP regulated is 10 mg/L, and the maximum instantaneous concentration is 30 mg/L. Figs. 3 and 4 show that the filtration through a Reo-dry medium took only 140 min, with a ~3 mg/L concentration of PP left in the last filtrate samples. This medium clogged before it was able to fully use its sorption capacity. Manufacturers indicate that Reo-dry absorbs about 3–4 L/kg of diesel or oil under static conditions; during this experiment only a 0.04 g/g ratio was achieved. This research showed that the Reo-dry medium can be used for runoff filtration at a high speed (~15 m/h) only for a short time; however, it is done effectively. The Duck medium clogged after 160 min of filtration; however, the PP concentration in filtrate samples ranged from 0.5 to 6 mg/L. This medium can also be used for short time filtration.

Figs. 5 and 6 illustrate what amounts of PP were held in all filter mediums until they clogged or while the experiment was still going (220 min). In this case, the individual materials were compared with each other according to their efficiency to hold the PP, and not according to their ability to absorb them (g diesel/g filter medium) (as the masses used were made of different materials of different quantities).

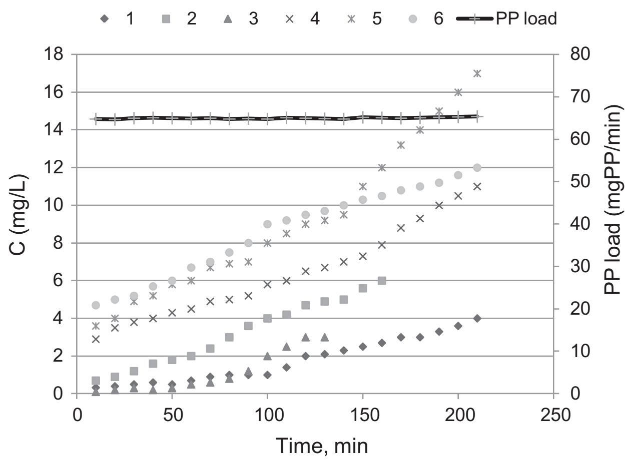

Fig. 4. The PP load and dynamics of PP concentration at the filter outlet. Series: 1—Fibroil; 2—Duck; 3—Reo-dry; 4—reed stems (*Phragmites australis*); 5—bulrush stems (*Schoenoplectus lacustris*); 6—lesser bulrush stems (*Typha angustifolia*).

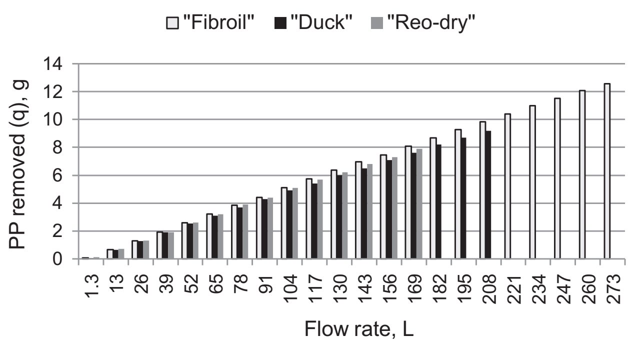

Fig. 5. PP removed from the liquid (first-type materials are tested).

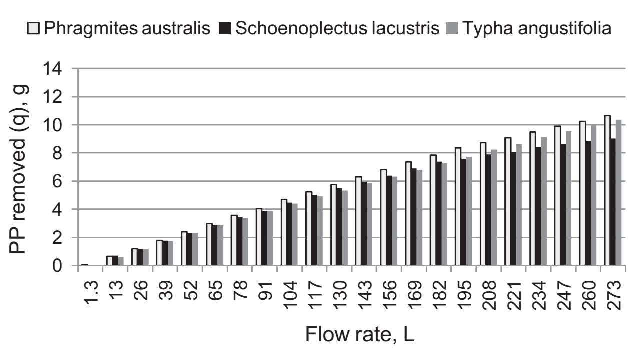

Fig. 6. PP removed from the liquid (second-type materials are tested).

As shown in Figs. 5 and 6, the maximum amount of PP was suspended by the Fibroil medium (12.6 g), less—by reed (10.7 g), cattails (10.4 g), Duck' (9.2 g), bulrush (9.0 g) and Reo-dry (7.9 g) mediums. Due to the different filter medium density and other properties (adhesion, absorbency, etc.), some of them held more SM (caused the clogging), others—PP. Table 2 shows the efficiency of SM suspension.

As Table 2 shows, all the filter mediums held part of SS. SM was effectively held by Reo-dry (97–98%) and Duck (88–94%) mediums; however, they became clogged first. All tested filter mediums reduced SM concentration up to regulatory requirements for the surface runoffs discharged into the natural environment (the maximum average annual concentration of SM discharging the runoff directly into the environment is 30 mg/L, the maximum instantaneous concentration is 50 mg/L) [3].

Based on the data obtained during the experiment, there was a sorption capacity that was estimated: 0.22 g/g for Fibroil, 0.08 g/g for Duck, 0.04 g/g for Reo-dry and about 0.11 g/g for plant stems. The sorption capacity is not very high compared to other sorbents, for example, the 3–6 g/g for natural fibres of wool; ~5 g/g for recycled wool-based nonwoven material; 0.19 g/g for sepiolite [15], and 2–3 g/g for roots and leaves of the *Pistia stratiotes* [18]. But most of the authors of this study consider that the sorption capacity is usually measured in sorbent under static conditions or low filtration rate when the sorption process is completed [15,16,18–26]. Many researchers observed a decrease in the sorption efficiency in experiments using an oil separator with the sorbents during the filtration [13,15,16,26]. The same workers investigated the treatment of stormwater using a detention pond and constructed filters [8]. The constructed filter system became clogged due to the cementation of the filter substrate. It was established that future systems for the treatment of runoff from urban roadways may include filtration through reactive porous media in order to increase the efficiency of the removal of dissolved contaminants [8].

The main focus of this work was on the runoff treatment and pollution concentration in filtrate. Based on the experience of scientists working in this field, this study was organized in such a way that it meets the following basic criteria [27]: a high treatment efficiency (cost–benefit ratio), a small space requirement and low maintenance. There is a potential for separators to remove other priority pollutants as a part of a "multi barrier" approach in order to meet higher environmental standards [7]. It is important to note that the vegetal mediums under the experiment conditions were suitable for the suspension of PP from the surface runoffs. Although their efficiency to hold PP is slightly lower than the sorbents sold on the market, plant stems are

Table 2
Average efficiency of mediums to hold suspended materials. SS concentration in inflow: C_0—from 36.5 to 51.2 mg/L

Medium	mg/L (%)	Filtration run (min)										
		20	40	60	80	100	120	140	160	180	200	220
Fibroil	C_f	5.6	5.8	6.1	6.5	7.0	7.6	8.3	9.2	10.2	10.7	11.2
	E (%)	86.8	86.3	85.6	84.7	83.5	82.1	80.4	78.3	75.9	74.8	73.6
Duck	C_f	2.3	2.5	2.8	3.2	3.5	3.9	4.3	4.7	–	–	–
	E (%)	94.2	93.7	93.0	92.0	91.2	90.2	89.2	88.2	–	–	–
Reo-dry	C_f	1.0	1.0	1.2	1.3	1.4	1.5	–	–	–	–	–
	E (%)	97.8	97.8	97.4	97.1	96.9	96.7	–	–	–	–	–
Phragmites australis	C_f	7.1	7.8	8.7	9.3	9.9	10.5	11.3	12.4	13.5	14.7	15.9
	E (%)	84.3	82.8	80.1	79.5	78.1	76.8	75.1	72.6	70.2	67.5	64.9
Schoeno-plectus lacustris	C_f	6.3	6.6	6.9	7.3	7.7	8.5	9.3	10.5	11.0	11.6	12.3
	E (%)	85.2	84.5	83.8	82.9	82.0	80.1	78.2	75.4	74.2	72.8	71.2
Typha angustifolia	C_f	1.2	1.5	1.6	1.8	2.1	2.6	3.2	4.0	4.7	5.5	6.8
	E (%)	96.7	95.9	95.6	95.1	94.2	92.9	91.2	89.0	87.1	84.9	81.4

characterized by lower hydrodynamic resistance. *Phragmites australis*, *Schoeno-plectus lacustris* and *Typha angustifolia* are plants growing in wet areas, swamps and lakeshores, so their stems are well-permeable. Their efficiency to absorb PP decreases with increasing duration of the filtration; however, a rain shower, the contamination of surface runoffs is usually the highest at the beginning of the shower [8]. Therefore, it is appropriate to use vegetal mediums during each rain event. It is not expensive to use reed, bulrush and cattails as naturally absorbing filter mediums for the removal of PP from surface runoffs. For example, the price of a medium made of these kinds of plant stems that amounts to 0.03 t for an experimental filter is only 1.8 Euro. The costs of most biomass sorbents only go for their collection and preparation. The use of the plans tested in runoff treatment in Lithuania is relevant not only economically, but also ecologically there are 2,830 lakes in Lithuania. Most of them are in the ripening and ageing stages. A lot of lakes disappeared due to land reclamation and eutrophication. The shores of lakes are plumed by a dense amount of reeds, rushes and cattails, which spoil the landscape. The shores become unsuitable for recreation and beaches, gradually turning into swamps. In addition, changes occur in their flora and fauna. Thus, what would be desirable is regular maintainance of the coastline, for example, through the harvesting of water plants, which could be used for absorbing petrolium products. The economic price of dry mass of tested plants is equal 0.06 Euro/kg. It is less than the number of low prices of natural sorbents: 0.15 Euro/kg for oakum, 0.12 Euro/kg for Reindeer moss, 1.46 Euro/kg for Coconut carbon [28].

It can be stated that there are enough production areas of reed, bulrush or cattails in all water bodies in Lithuania, and that the price for collecting and using them is low. There are ever more articles and publications about the using of natural organic sorbents for runoff treatment, as they are more acceptable than synthetic sorbents from an environmental point of view [12,14–16,21]. Today, for economic reasons, organic and inorganic natural products, such as rice straw, corn cob, peat moss, wool, cotton, cotton grass, cattail fibre, rice husk, sawdust, wood chips bark, bagasse, milkweed, zeolite, clinoptilolite and vermiculite are becoming increasingly important as alternatives to synthetic sorbents [10,11,16,18,19,24,25]. After they have absorbed PP, plant stems can be easily disposed of through combustion; however, sorbents sold on the market are more expensive: the price for 1 kg of Fibroil is 5.50 Euros, while for Duck and Reo-dry it is 2.90 Euro for 1 kg. The recovery price of these materials is 0.058 Euro per 1 kg. Moreover, Reo-dry and Duck sorbents can be safely used in filtration systems only for a short period of time (140–170 min in the case of a rain shower, as later they get clogged). These laboratory studies have shown that vegetal filter mediums work more reliably than Reo-dry and Duck sorbents at a high filtration rate (15 m/h). Plants have been found to be efficient for the removal of aquatic pollutants, especially PP. However, the practical utility of these materials should be studied on a commercial scale as well.

4. Conclusions

The suitability of different materials—both synthetic (Fibroil, Duck, Reo-dry) and those of a natural vegetal origin (bulrush, cattail and reed stems)–to remove PP from the liquid, when the filtration through them was carried out at relatively high rate (15 m/h) was studied during the laboratory experiments. The plant stems that were tested (*P. australis*, *S. lacustris* and *T. angustifolia*) removed PP from contaminated surface runoffs effectively (78–66%). The efficiency of plant stems to remove PP seemed to be lower than the sorbents sold on the market. There was a difference in their efficiency that ran from. There was an 18–22% difference in the efficiency of filtration between the first-type of material (the three sorbents currently sold on the market) and the second-type of material (the three vegetal-based materials). However, the plant stems that were tested were characterized by better hydraulic permeability, and could have been used longer than Reo-dry and Duck sorbents. Reo-dry and Duck sorbents can be safely used in filtration systems only for a short period of time. The assessment in the environmental and economic aspects showed that the tested plant stems (reeds, bulrushes and cattails) can be used in filtration schemes for surface treatment of runoff with PP.

List of symbols

$C_{aver.}$	—	average concentration at typical points (mg/L)
c_i	—	concentration of substances at typical points (mg/L)
m_i	—	probability at recurrence of concentration
n	—	number of measurements
k	—	number of different values of concentration
V_1 and V_2	—	are the respective volumes of liquid treated at the time the samples are taken (L)
C_0	—	influent concentration (mg/L)
C_2	—	effluent concentration (mg/L)

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