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STUDY OF THE POSSIBILITY OF USING LEAF LITTER TO CLEAN UP WATER ENVIRONMENTS FROM PETROLEUM PRODUCTS

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ABSTRACT

We studied the sorption properties of tree foliage samples growing in urban anthropogenic landscapes in relation to petroleum products. As sorption material we chose leaf litter of apple-tree (Lat. Mālus), birch (Lat. Bétula), oak (Lat. Quércus), and aspen (Lat. Populustremula). The physical and chemical characteristics (moisture, ash content, and bulk density) of the fallen leaves samples were determined. Atomic-emission determination of the content of various elements in acetate-ammonium extracts of selected leaf samples was carried out to evaluate the safety of their use as sorption material. The leaves of an apple tree growing along a busy highway were chosen as an object for evaluation of metalaccumulation capacity, and samples of leaves were taken in spring and autumn periods. To assess the background content of elements, samples of apple-tree leaves were also taken away from anthropogenic sources of impact. It was found that the content of heavy metal ions in the leaves of apple-tree under intensive anthropogenic load does not exceed the established standard requirements, respectively; the material in question can be used as a sorbent for purification of water environments from various pollutants. Experiments on purification of model solutions containing oil products were carried out under static and dynamic conditions. It was found that all the leaf samples under consideration possess sorption properties towards oil products to a different degree: the minimum adsorption capacity of aspen leaves - 0.42 mg/g under static and 0.17 mg/g under dynamic conditions, the maximum adsorption capacity of oak leaves - 0.73 and 0.28 mg/g respectively.

Keywords: leaf litter, water treatment, petroleum products, adsorption, sorption material.

1. INTRODUCTION

Currently, one of the significant problems is the pollution of water bodies. Among the main pollutants of water bodies, oil and oil products (OP) play a special role. Oil gets into the aquatic environment and undergoes complex processes of transfer and transformation, the course of which depends both on the properties of oil itself and on environmental conditions. Spreading of oil and OP is accompanied by their dissolution, emulsification, chemical and photochemical oxidation and breaks normal functioning of aquatic ecosystems [1].

When oil gets into the water, usually immediately a surface film is formed. Formation of oil films prevents the flow of oxygen and sunlight in the water column [2], resulting in various violations of the physiological activity of hydrobionts, leading to their death and further breakage of food chains. Ultimately, the water body becomes unsuitable for use for domestic and drinking consumption [3].

To date, many different methods of treatment of water bodies polluted with oil and OP are proposed [4-6]. Among the available methods the adsorption method is most often used for water purification from OP and in recent years more often production and consumption wastes are considered as sorption materials (SM) [7-10]. In order to improve the physicochemical and sorption properties of the above-mentioned SM various modification options are proposed, including physical and chemical effects (heating, plasma, ultrasound, microwaves) and treatment with chemical substances [11-15].

Researchers study the sorption properties of components and wastes of woody plants [16-18].

Relatively high sorption capacity, low cost and developed raw material base allow considering the above materials as promising sorbents for removal of oil and OP from water environments.

The presented work investigated the possibility of using leaves of woody plants (apple, birch, oak, and aspen) in their native form as sorption materials for the purification of aqueous media from OP.

2. METHODS

Every year in the fall, the municipalities of the Republic of Tatarstan (Russian Federation) collect the fallen leaves by their public utilities. Dozens of tons of biomaterial are removed from the environment, burned or buried in landfills. It is generally believed that trees growing in large settlements are potential accumulators of pollutants (including heavy metal ions) and further use of waste and components of woody plants can cause some harm to the environment.

As part of the presented work, we conducted studies on the possibility of using leaf litter as a SM for the extraction of petroleum products from aqueous media. As SM we chose the leaf litter of apple (Lat. Mālus), birch (Lat. Bétula), oak (Lat. Quércus), and aspen (Lat. Populustremula). Sampling of fallen leaves was conducted in the fall of 2020. The initial phase of the study examined the physicochemical characteristics of the leaf samples. The leaves, dried at room temperature, were ground in a laboratory ball mill to a powdery state, after which humidity, ash content, and bulk density of the samples were determined in accordance with standard methods [19].

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In order to estimate the metal-accumulating ability of tree leaves, atomic-emission determination of 25 elements content in acetate-ammonium extracts of apple-tree fallen leaves samples growing along a busy highway was performed. Leaf samples were collected during the spring and fall periods. Samples of apple-tree leaves away from anthropogenic sources of influence were also taken to assess the background content of elements. Determination of elements was carried out on an atomic emission spectrometer with inductively coupled plasma "Agilent 720 ICP-OES" [20].

Calculation of the content of elements determined by the standard software device by comparing the obtained data on the analyzed sample with the data calibration graphs, taking into account the sample taken for analysis of the sample, as well as a blank sample. The content of analyzed elements in the sample in mg/kg (X) was calculated by the formula (1):

$$X = \frac{X_p \cdot V}{m} \qquad (1)$$

where X_p - mass concentration of the element in the analyzed solution (mg/kg), found from the calibration chart, mg/dm^3; m - mass of the sample, kg; V - volume of the analyzed solution, dm^3.

Diesel fuel (DF) was used as model solutions in the study of NP adsorption. Model OP solutions were obtained by intensive mixing of DF and distilled water. After stirring, the solution was sedimented and separation of the aqueous and organic phases was carried out. The content of OP was determined by infrared spectroscopy on the analyzer of oil products "KN-3" in accordance with the standard method [21].

The study of OP adsorption was carried out in the static mode. We determined the concentration of dissolved components of DF after 2 hours of contact with samples of chopped leaf weight of 1 g with 100 ml of a model solution containing oil products with a concentration of 10 mg/l in a neutral medium at 20 ° C.

The purification efficiency (R, %) of the model solution was calculated by formula (2):

$$R = \frac{C_0 - C_1}{C_0} \cdot 100 \qquad (2)$$

where C_0 - initial concentration of OP, mg/dm^3; C_1 - final concentration of OP, mg/dm^3.

Adsorption capacity of leaf samples (A, mg/g) was calculated by formula (3):

$$A = \frac{(C_0 - C_1) \cdot V}{m} \qquad (3)$$

To evaluate the sorption properties of leaf samples under dynamic conditions, adsorption was performed on a laboratory filtration unit by passing model solutions containing OP with a concentration of 1.9 mg/dm^3 with a given flow rate through SM-filled glass columns 150 mm long, 20 mm in diameter with a perforated bottom at room temperature. SM layer height was 80 to 90 mm, SM mass was 5 g, model solution volume was 1 dm^3. According to the results of the experiments, the purification efficiency and adsorption capacity of the sorption materials under consideration were calculated by formulas 2 and 3. To compare the sorption capacity of the studied materials activated carbon of "BAU-A" brand was used [22].

3. RESULTS AND DISCUSSIONS

Woody plants can act as a natural universal filter capable of protecting the environment from pollution, because they extract and concentrate various elements in their tissues, so they are used to identify the level of accumulation of heavy metal ions (HME) as one of the sources of anthropogenic pollution [23]. The possibility of using components and wastes of woody plants as SM for the extraction of various pollutants from aquatic environments is also widely studied [24]. However, one of the obstacles to the use of the latter for adsorption of pollutants from aqueous media is the prejudice that they are the main accumulator of HME.

In the presented work, in order to assess the safety of further application, the metal-accumulating ability of leaves of urban tree plantations was investigated and the possibility of their use as a sorption material for OP removal from water environments was assessed. The leaves of apple, birch, oak, and aspen were chosen as the samples under study. At the initial stage, physico-chemical properties of dried and crushed leaf samples were determined. The results are presented in Table-1.

Table-1. Physicochemical properties of leaf samples.

Index	Index value			
	Apple tree	Birch	Oak	Aspen
Humidity, %	10,8	8,9	9,1	9,5
Tamped density, g/sm^3	0,17	0,19	0,21	0,19
Ash content, %	1,14	1,21	1,23	1,16
Cellulose content, %	30 - 40			
Lignin content, %	15 - 30			
Hemicellulose content, %	15 - 40			
Tannin content, %	0,8 - 2			

According to the results obtained, the bulk density of leaf samples varies from 0.17 to 0.21 g/cm^3. In contrast to the wood itself, leaves have a fairly high ash content (1.14 - 1.23 %), due to the increased content of lignin. Also, according to the results of the analysis of literary sources [25], Table-1 shows quantitative data on

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the content of chemical compounds included in leaves - cellulose, lignin, hemicellulose and tannins.

The next stage of the work was to establish the metal-accumulating ability of leaves. Apple-tree leaves were chosen as a test sample. Leaf samples were taken in spring and autumn periods at seven control points along a busy highway (T. 3 - T.9), as well as to assess the background content of elements at two control points away from anthropogenic impact sources (T. 1 - T.2). The content of 25 elements in acetate-ammonium extracts of

apple-tree leaves was determined by the method of atomic emission spectroscopy. The results are presented in Tables 2 and 3.

According to the results of studies, it was found that by autumn, the content of macronutrients (calcium, magnesium) in apple-tree leaves samples in terms of dry weight increases, as well as the concentration of trace elements (strontium, barium) increases. The content of aluminum, iron and boron decreases, selenium, vanadium, silicon - remains approximately at the same level.

Table-2. The content of elements in the acetate-ammonium extract of apple leaves (spring time) in terms of dry weight.

Element	Content, mg/kg								
	T. 1	T. 2	T. 3	T. 4	T. 5	T. 6	T. 7	T. 8	T. 9
Al	1,70	1,90	5,35	11,40	4,70	9,30	4,90	4,00	3,10
Ba	0,57	0,52	2,18	3,08	5,11	1,28	6,93	3,72	6,69
Be	<0,01								
Cd	<0,01								
Co	<0,01								
Cr	<0,01								
Cu	0,43	0,52	1,21	1,64	15,0	2,09	3,18	6,23	2,94
Fe	11,2	19,4	18,5	30,3	22,9	25,5	16,5	15,3	16,2
Mn	15,3	46,6	30,0	41,2	72,4	66,6	21,8	34,4	30,5
Mo	<0,05								
Ni	0,31	0,27	0,75	0,82	0,86	0,94	1,54	1,08	0,96
Pb	<0,05								
Sb	<0,25								
Se	1,40	1,30	1,90	1,75	2,70	2,30	3,30	4,80	1,85
Si	20,3	20,1	26,2	37,8	33,4	37,4	43,6	24,1	25,1
Sr	3,50	1,70	8,50	6,50	16,85	5,25	13,60	11,75	5,95
Ti	<0,05								
V	2,35	2,80	4,55	4,50	5,40	3,65	3,45	5,45	4,10
Zn	14,5	14,7	16,8	18,1	26,7	23,2	34,8	24,2	26,0
Ca	9250	8200	13650	11900	16600	12050	19850	20100	10500
B	2,95	3,10	5,40	10,20	5,70	5,95	6,45	13,30	5,25
Mg	1935	1780	2550	2580	3095	2205	2045	3120	2605
Ag	<0,25								
Tl	<0,25								
As	<0,25								

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Table-3. The content of elements in the acetate-ammonium extract of apple leaves (autumn time) in terms of dry weight.

Element	Content, mg/kg								
	T. 1	**T. 2**	**T. 3**	**T. 4**	**T. 5**	**T. 6**	**T. 7**	**T. 8**	**T. 9**
Al	0,95	0,80	1,90	3,45	1,80	1,55	1,60	3,90	2,25
Ba	2,82	2,11	14,6	15,3	7,83	9,66	7,58	12,3	11,7
Be	<0,01								
Cd	<0,01								
Co	<0,01								
Cr	<0,01								
Cu	0,64	0,72	1,36	3,11	3,42	3,85	4,21	2,66	4,58
Fe	8,15	6,22	13,7	16,3	17,1	19,9	13,0	9,85	12,5
Mn	26,8	27,3	55,4	68,1	62,9	75,3	98,7	62,6	54,3
Mo	<0,05								
Ni	0,34	0,31	0,79	0,87	0,81	0,99	1,38	1,23	1,08
Pb	<0,05								
Sb	<0,25								
Se	0,35	0,25	0,67	2,05	2,00	1,95	3,95	3,65	0,60
Si	19,7	18,0	26,1	44,0	32,9	37,2	48,4	20,6	31,5
Sr	3,40	2,75	18,5	14,6	22,7	9,65	30,5	29,1	14,4
Ti	<0,05								
V	1,60	1,25	3,15	3,90	3,35	4,55	5,60	6,90	3,85
Zn	13,9	14,5	19,0	18,8	27,3	24,5	36,6	26,2	31,0
Ca	10150	11200	26400	18350	20800	15250	29850	31000	18350
B	1,60	1,35	1,95	3,10	4,20	2,05	1,45	3,15	2,50
Mg	1075	1195	2935	3840	3510	2970	3075	3730	3260
Ag	<0,25								
Tl	<0,25								
As	<0,25								

Assessment of the level of chemical pollution was carried out by the indicators developed in the conjugate geochemical and hygienic studies of the environment of large settlements [26]. Such indicators are the concentration factor of a chemical element (K_c) and the total pollution index (Z_c). Kc determined as the ratio of the real content of the element in the sample (C) to the maximum allowable concentration of the element (MPS):

$$K_c = \frac{C}{MPS}$$

(4)

Z_c was also calculated for each control point, reflecting the effect of exposure to a group of elements:

$$Z_C = \sum_1^n K_{ci} - (n-1)$$

(5)

where K_{ci} - concentration factor of the i-th element in the sample; n - number of elements taken into account [26].

The results of calculations of K_c of some elements are shown in Tables 4 and 5.

Table-4. Concentration coefficient of chemical element in apple leaves (spring time).

Element	Concentration coefficient of chemical element (K_c)								
	T. 1	**T. 2**	**T. 3**	**T. 4**	**T. 5**	**T. 6**	**T. 7**	**T. 8**	**T. 9**
Cu	0,09	0,11	0,24	0,33	2,98	0,43	0,65	1,23	0,57
Fe	0,22	0,39	0,37	0,61	0,46	0,51	0,33	0,31	0,32
Mn	0,11	0,33	0,21	0,29	0,52	0,48	0,16	0,25	0,22
Ni	0,62	0,53	1,49	1,63	1,72	1,87	3,08	2,16	1,92
Zn	0,26	0,27	0,30	0,33	0,49	0,42	0,63	0,44	0,47
B	0,07	0,08	0,14	0,26	0,14	0,15	0,16	0,33	0,13

Table-5. Concentration coefficient of chemical element in apple leaves (autumn time).

Element	Concentration coefficient of chemical element (K_c)								
	T. 1	**T. 2**	**T. 3**	**T. 4**	**T. 5**	**T. 6**	**T. 7**	**T. 8**	**T. 9**
Cu	0,12	0,15	0,28	0,61	0,73	0,79	0,83	0,54	0,92
Fe	0,16	0,12	0,28	0,33	0,34	0,41	0,25	0,20	0,25
Mn	0,20	0,21	0,42	0,48	0,43	0,55	0,72	0,47	0,38
Ni	0,68	0,61	1,59	1,74	1,62	1,98	2,75	2,46	2,15
Zn	0,25	0,26	0,35	0,34	0,50	0,44	0,67	0,48	0,56
B	0,04	0,03	0,05	0,08	0,11	0,05	0,04	0,08	0,06

Table-6. Cleaning efficiency and adsorption capacity of leaf litter samples in relation to petroleum products in static and dynamic conditions.

Sample	Cleaning efficiency, %		Adsorption capacity, mg/g	
	static conditions	**dynamic conditions**	**static conditions**	**dynamic conditions**
Apple tree	41	49	0,51	0,19
Birch	49	57	0,62	0,24
Oak	58	64	0,73	0,28
Aspen	32	43	0,42	0,17
BAU-A	92	98	2,36	0,95

It was established that in spring, MPS exceedences for copper and nickel were observed in the control points along the highway. In autumn, there are no exceedances of MPS for copper and for nickel.

For each control point Z_c was calculated, reflecting the effect of exposure to a group of elements. According to the results of calculations Z_c established that the level of pollution by the studied elements is permissible. It should be noted that, in general, in samples of leaves growing away from anthropogenic sources, the content of elements is significantly lower than in points along a busy highway. Thus, even under conditions of intensive anthropogenic load, the content of elements (including HME) in apple-tree leaves does not exceed MPS, and, accordingly, the leaf litter under consideration can be used as a potential media for purification of water environments from various pollutants.

The next stage of the research was to establish the possibility of using leaf samples for the extraction of OP from aqueous media. To evaluate the sorption properties of leaf litter of apple-tree, birch, oak and aspen in relation to OP, experiments on purification of model solutions containing OP were carried out. In the study of adsorption in the static mode, the residual concentration of OP in the model solution containing 10 mg/dm^3 OP was determined after 2 h of contact with samples of crushed leaves. Under dynamic conditions, the model solution containing 1.9 mg/dm^3 OP was passed through glass columns filled with leaf litter. The results of the experiments are presented in Table-6. For comparison of sorption properties of the studied materials the table also contains data on commercial sorbent of "BAU-A" brand.

Experiments on purification of model solutions with the use of crushed leaf litter showed that the efficiency of purification from OP varies from 32 to 58%

in static conditions and from 43 to 64% in dynamic mode. The maximum value of an indicator of adsorption capacity is observed at the crushed leaf decay of oak, which is 0.73 mg/g in static and 0.26 mg/g - in dynamic conditions. Thus the spent researches allow to consider the leaf litter as a safe material for OP extraction from water environments. To improve the sorption properties of the considered SM it is advisable to carry out further modification by various physical and chemical methods.

SUMMARY

The paper reflects the results of studies on the physicochemical and sorption properties of leaf litter of apple, birch, oak, and aspen in its native form. Samples of fallen leaves were collected in an urbanized urban area in late autumn. After appropriate sample preparation, the physical and chemical characteristics of the fallen leaves were determined. According to the results obtained, ash content of leaf samples ranged from 1.14 to 1.23 g/cm^3, bulk density - 0.17 to 0.21 g/cm^3. The metal-accumulating capacity of leaves of apple trees growing along a busy highway was determined to establish the safety of further application as SM. Leaf samples were taken in the spring and fall periods. The method of atomic emission spectroscopy determined the content of 25 elements. According to the results of calculations of concentration coefficients (K_c) and the total pollution index (Z_c), it was found that the level of pollution by the studied elements is acceptable. To evaluate the sorption properties of leaf litter of apple-tree, birch, oak and aspen in relation to OP we conducted experiments on the purification of model solutions containing OP in static and dynamic conditions. The carried out researches have shown that all considered samples of leaf litter in various degree possess sorption properties in relation to OP. The minimum adsorption capacity of aspen leaves was 0.42 mg/g under static and 0.17 mg/g under dynamic conditions; the maximum adsorption capacity of oak leaves was 0.73 and 0.28 mg/g, respectively.

Thus, the conducted experiments allow considering the leaf litter of apple-tree, birch, oak and aspen as a safe SM for adsorption of OP from water bodies. To improve the sorption properties of the above materials it is advisable to carry out further modification by various physical and chemical methods.

4. CONCLUSIONS

The possibility of using leaf litter in its native form to purify aquatic environments from petroleum products was investigated. The leaves of apple-tree, birch, oak and aspen were chosen as objects of research. It has been established that even in conditions of intensive anthropogenic load the content of ions of heavy metals and other elements in the samples of leaf litter does not exceed the established standards, and, accordingly, the considered samples of leaves after appropriate sample preparation can be used as potential sorption materials for treatment of water environments from oil products.

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