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ASSESSMENT OF THE OZONATION IMPACT ON ADSORPTION EFFICIENCY IN SURFACE WATER TREATMENT

Organic substances in water, both of natural and synthetic origin, especially their share in water treatment by-products can pose a threat to drinkers. That is why adsorption, as a very effective process of dissolved organic compounds removal is commonly used in surface water treatment systems. For process design and optimization, mathematical models both mechanistic and statistics are created. The results of the investigation of granular activated carbon (GAC) bed adsorption in a pilot plant with a capacity of 3 m³/h have been presented. Two systems have been tested – without ozonation and with ozonation before GAC adsorption. The models of the kinetics of GAC adsorption capacity exhaustion, the model of minimal GAC bed depth (adsorption zone) for assumed process efficiency (\underline{C}/C_0), as well as the model of adsorptive equilibrium, the first model enables the determination of the isotherm parameters of the Freundlich type, the two other models are used for the calculation of GAC bed run time for the certain bed depth and assumed efficiency. It has been shown that in this case (water pollution, GAC type, pre-treatment) ozonation plays a minor role.

1. INTRODUCTION

Due to the increase in contamination of source surface waters with household and industrial wastewater and atmospheric precipitation water treatment systems more often make use of adsorption processes [1, 2]. This becomes all the more acute because of the increasing number of organic pollutants, particularly pharmaceuticals, cosmetics [3, 4], and pesticides, whose consumption is increasing worldwide [5, 6]. Apart from micropollutants, an ever-increasing amount of residential and industrial sewage containing organic substances of varying properties is being introduced into water [7–9].

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The adsorption process, as shown by Hyung and Kim [10] enables the removal of substances of low and medium molecular masses from water, which are susceptible to removal by coagulation to a low degree [11] with a simultaneous increase in the effectiveness of removing micropollutants [12–14] as well as disinfectant by-product precursors [15]. Additionally, in the case of adsorption on activated carbon beds, a spontaneous biofilm formation occurs on the surface of sorbent grains [16, 17] which enables simultaneous biodegradation [18]. Often a significant increase in organic substance removal takes place, including biodegradable dissolved organic carbon (DOC) [19], while also increasing bed lifetime. An increase in the effectiveness of biologically active beds in removing organic substances is also achieved by the preceding adsorption process with ozonation [14] which yields a change in the structure of organic substances in water towards one more susceptible to adsorption and biodegradation

The effectiveness of activated carbons is determined by the concentration and type of contaminants present in water, primarily their size, level of aromaticity, and degree of hydrophobicity [20]. Precisely, it is the presence of a very diverse mixture of organic substances differing in properties and the dissimilarity of these substances in different waters makes it difficult to choose the right activated carbon to ensure effective removal of organic substances. Therefore, it is common to use the modeling of the adsorption process by determining the adsorption isotherm, i.e., adsorption isotherm parameters and the most suitable model. For solutions containing specific substances, they are determined empirically, while for multicomponent solutions, substitute adsorption constants can be used [21].

Mathematical models that described the generalized course of adsorption in granular activated (GAC) beds in the line of surface water treatment without and with ozonation before adsorption have been presented in this paper. A comparative analysis of the efficiency of DOC removal in these lines has been made.

2. METHODS EMPLOYED IN THE STUDY

The study was carried out in flow water treatment systems without ozonation (I) and with ozonation (II), consisting of coagulation, sedimentation, rapid sand filtration, and adsorption (Fig. 1). The ozonation was performed prior to adsorption. Both systems were operated continuously with a throughput of $3 \text{ m}^3/h$.

The study was started after the adsorption filters (with bed height of 1.5 m) of both systems were filled with fresh activated carbon and ran for 238 days. The adsorption process was carried out on rapid filters filled with GAC (WG12), whose properties are shown in Table 1. The ozonation process was operated with a constant ozone dosage of $1.2 \text{ g O}_3/\text{m}^3$, generated by a BMT 803 BT ozone generator. The water bed contact time for both systems was between 17 and 19 min. Insignificant changes in the flow time through the beds resulted from constant system throughput.



- Fig. 1. Schematic of water treatment process without ozonation (upper) and with ozonation (lower): RMT – rapid mixing tank, FB – flocculation basin, Cl – clarifier,
 - $SF-sand\ filter,\ GAC-granular\ activated\ carbon\ filter,\ CWB-clear\ water\ basin$

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Parameter	Value
Bulk density, g/dm ³	470
Volatile substance content, %	1.50
Ash content, %	11.6
Specific surface area, m ² /g	968
Iodine number, mg/g	1014
CTC (adsorption CCI ₄), %	62.3
<0.5 mm subfraction content, %	0.0
Mechanical strength, %	97.3

Properties of WG12 activated carbon*

*Data from the Gryfskand Manufacturer.

The study was conducted for surface water with and without ozonation, pre-treated by coagulation, sedimentation, and rapid sand filtration, with the range of DOC level of 2-6 g C/m³, and the flow of 5 m/h. Water samples were taken once a day at different depths of the bed (0.6, 1.1, and 1.5 m). Dissolved organic carbon (DOC) concentrations were analyzed using the Shimadzu TOC analyzer.

The following parameters of the model have been defined:

• The kinetics of adsorptive mass accumulation in fraction of GAC bed depth

$$M_{\rm DOC} = f\left(t_c, H_{\rm GAC}\right) \tag{1}$$

• The minimal depth of GAC bed (adsorption zone)

$$H_{\min} = f\left(\frac{C}{C_0}\right)_{AS}$$
(2)

• The velocity of adsorption zone migration to the bottom of the GAC bed

$$V_M = f\left(\frac{C}{C_0}\right)_{\rm AS} \tag{3}$$

where C and C_0 are effluent and influent concentrations, respectively, H_{GAC} is the GAC bed depth, m, t_c GAC bed run time, d.

For the state of adsorption equilibrium in both lines, the parameters of Freundlich's isotherm have been defined.

3. RESULTS

DOC levels in the samples taken from different GAC bed depths are given in Tables 2 (without ozonation) and 3 (with ozonation).

Table 2

	H = 0.60 m		H =	1.10 m	H = 1.50 m		
Bed run time	$(m_{\rm GWA} = 77.8 \text{ kg})$		$(m_{\rm GWA} =$	= 142.6 kg)	$(m_{\rm GWA} = 194.5 \text{ kg})$		
[d]	C/C_{\circ}	$M_{\rm DOC}$	$C C_{0}$	$M_{\rm DOC}$	$C C_{0}$	$M_{\rm DOC}$	
	C/C0	[g]	C/C0	[g]	C/C0	[g]	
7	0.46	316.7	0.21	467.9	0.15	504.0	
14	0.61	599.2	0.41	882.0	0.24	987.9	
21	0.73	795.8	0.64	1169.3	0.40	1394.5	
28	0.74	952.0	0.55	1406.2	0.42	1741.4	
36	0.73	1186.2	0.47	1846.8	0.48	2218.5	
42	0.77	1370.5	0.65	2174.4	0.57	2568.4	
49	0.82	1515.0	0.73	2389.4	0.61	2852.3	
56	0.78	1637.6	0.73	2563.3	0.70	3064.0	
62	0.79	1744.9	0.70	2713.8	0.62	3234.7	
70	0.80	1872.6	0.70	2901.0	0.61	3435.7	
77	0.85	1962.5	0.84	3020.3	0.64	3631.4	
84	0.84	2037.3	0.72	3126.1	0.65	3807.0	
91	0.82	2117.9	0.76	3248.7	0.67	3968.3	
99	0.89	2193.7	0.79	3367.7	0.68	4139.2	

Relative DOC levels (C/C_0) and DOC mass accumulated (M_{DOC}) on different bed depths and bed run times (line without ozonation)

Table 2

	H = 0.60 m		H =	1.10 m	H = 1.50 m		
Bed run time	$(m_{\rm GWA} = 77.8 \text{ kg})$		(<i>m</i> _{GWA} =	= 142.6 kg)	$(m_{\rm GWA} = 194.5 \text{ kg})$		
[d]	C/C_0	MDOC	C/C_0	MDOC	C/C_0	MDOC	
105			o = 1		0.62		
105	0.78	2257.1	0.74	3455.5	0.63	4267.4	
112	0.77	2353.7	0.74	3567.2	0.66	4421.1	
119	0.80	2444.4	0.75	3672.2	0.67	4561.4	
126	0.87	2517.5	0.75	3784.8	0.67	4711.0	
133	0.84	2588.9	0.78	3900.7	0.70	4801.7	
140	0.88	2655.3	0.80	4001.5	0.73	4939.5	
147	0.88	2713.3	0.83	4093.9	0.77	5064.7	
154	0.91	2765.4	0.89	4163.6	0.85	5158.0	
161	0.93	2806.6	0.84	4232.5	0.77	5253.8	
168	-	2824.2	0.90	4298.9	0.79	5364.7	
175	0.87	2876.3	0.81	4371.1	0.79	5459.7	
183	0.92	2938.7	0.84	4475.8	0.82	5576.8	
189	0.90	2981.2	0.86	4547.1	0.81	5663.9	
196	0.92	3033.3	0.86	4632.8	0.82	5776.5	
203	0.93	3079.2	0.87	4716.0	0.81	5891.6	
210	0.92	3119.5	0.88	4782.4	0.84	5989.1	
224	0.93	3205.2	0.88	4925.2	0.87	6167.2	
238	0.93	3297.6	0.92	5056.3	0.87	6331.9	

Relative DOC levels (C/C_0) and DOC mass accumulated (M_{DOC}) on different bed depths and bed run times (line without ozonation)

Table 3

Relative DOC levels (C/C_0) and DOC mass accumulated (M_{DOC}) on different bed depths and bed run times (line with ozonation)

	H = 0.60 m		H = 1.10 m		H = 1.50 m		
Bed run time	$(m_{\rm GWA} = 77.8 \text{ kg})$		$(m_{\rm GWA} =$	= 142.6 kg)	$(m_{\rm GWA} = 194.5 \text{ kg})$		
[d]	$C C_0$	$M_{\rm DOC}$	$C C_0$	MDOC	C/C_{0}	$M_{\rm DOC}$	
	0/00	[g]	0/00	[g]	0/00	[g]	
7	0.55	270.5	0.22	450.2	0.11	509.0	
14	0.65	497.3	0.35	856.8	0.28	965.1	
21	0.77	688.8	0.49	1189.1	0.43	1332.2	
28	0.73	826.6	0.55	1451.2	0.41	1652.2	
36	0.81	1035.9	0.59	1811.2	0.49	2112.0	
42	0.86	1154.7	0.69	2070.4	0.59	2440.3	
49	0.69	1293.3	0.72	2266.1	0.91	2560.4	
56	0.72	1465.5	0.85	2391.3	0.63	2694.8	
62	0.83	1574.2	0.68	2500.0	0.65	2866.2	
70	0.81	1680.8	0.73	2673.8	0.61	3084.1	
77	0.79	1779.9	0.91	2762.0	0.64	3269.7	

Table 3

	H = 0.60 m		H =	1.10 m	H = 1.50 m		
Bed run time	$(m_{\rm GWA} = 77.8 \text{ kg})$		$(m_{\rm GWA} = 142.6 \text{ kg})$		$(m_{\rm GWA} = 194.5 \text{ kg})$		
[d]	C/C.	MDOC	C/C_0	MDOC	C C	MDOC	
	C/C_0	[g]		[g]	C/C_0	[g]	
84	0.81	1874.8	0.66	2862.8	0.63	3442.7	
91	0.80	1967.2	0.68	3021.6	0.56	3636.7	
99	0.83	2064.2	0.68	3191.5	0.60	3863.3	
105	0.81	2126.8	0.75	3293.7	0.63	4002.3	
112	0.79	2205.8	0.66	3414.7	0.60	4158.5	
119	0.84	2278.0	0.68	3546.6	0.62	4315.6	
126	0.82	2353.6	0.69	3683.5	0.64	4476.9	
133	0.85	2439.3	0.76	3813.7	0.69	4634.8	
140	0.86	2514.9	0.70	3943.1	0.62	4800.3	
147	0.87	2581.3	0.77	4075.0	0.69	4970.8	
154	0.94	2625.0	0.84	4166.6	0.79	5091.8	
161	0.91	2661.1	0.81	4249.8	0.71	5211.1	
168	0.91	2705.6	0.78	4349.8	0.70	5354.7	
175	0.85	2763.6	0.78	4454.8	0.76	5484.9	
183	0.91	2832.7	0.81	4571.0	0.77	5619.3	
189	0.96	2861.5	0.80	4659.6	0.77	5723.0	
196	0.86	2917.8	0.79	4783.9	0.75	5870.0	
203	0.91	2989.2	0.77	4908.2	0.79	6011.1	
210	0.88	3043.0	0.77	5019.9	0.78	6124.5	
224	0.90	3079.3	0.88	5219.8	0.78	6391.6	
238	0.95	3171.7	0.90	5355.9	0.88	6606.6	

Relative DOC levels (C/C_0) and DOC mass accumulated (M_{DOC}) on different bed depths and bed run times (line with ozonation)



→ H=0.7 m → H=1.1 m → 1.5 m

Fig. 2. Kinetics of DOC mass accumulation for line without ozonation



Fig. 3. Kinetics of DOC mass accumulation for line with ozonation

Time dependences of adsorptive mass accumulation (M_{DOC}) are presented in Figs. 2 (line without ozonation) and 3 (line with ozonation). The courses for M_{DOC} , g, shown in Figs. 2 and 3 can be described by the following equations:

without ozonation

$$M_{\rm DOC} = (90 + 207H_{\rm GAC})\sqrt{t_c}$$
(4)

• with ozonation

$$M_{\rm DOC} = (100 + 207H_{\rm GAC})\sqrt{t_c}$$
(5)



Fig. 4. Isoplane of GAC bed at a depth of 0.6 m (line without ozonation)



Fig. 5. Isoplane of GAC bed at a depth of 1.1 m (line without ozonation)



Fig. 6. Isoplane of GAC bed at a depth of 1.5 m (line without ozonation)



Fig. 7. Isoplane of GAC bed at a depth of 0.6 m (line with ozonation)



Fig. 8. Isoplane of GAC bed at a depth of 1.1 m (line with ozonation)



Fig. 9. Isoplane of GAC bed at a depth of 1.5 m (line with ozonation)

The isoplane courses of GAC columns with different bed depths are shown in Figs. 4–6 for the line without ozonation and in Figs. 7–9 for the line with ozonation. Based on the isoplane course, the time of adsorptive capacity exhaustion, and the maximum possible efficiency of the process at the beginning of the cycle have been determined. The bed depth that guarantees the assumed value of C/C_0 is described by the following equations:

• for the line without ozonation $H_{GAC, min}$, m, is

$$H_{\rm GAC,\,min} = \left(0.45 + 3.48 \frac{C}{C_0}\right)^{-1}$$
(6)

• for the line with ozonation $H_{GAC, min}$, m, is

$$H_{\rm GAC,\,min} = \left(0.33 + 3.83 \frac{C}{C_0}\right)^{-1}$$
(7)

The equations relating concentrations and velocities of their movement to the bottom of the bed V_M , m/h, are as follows:

• for the line without ozonation

$$V_{M} = \left(230 - 237 \frac{C}{C_{0}}\right) \times 10^{5}$$
(8)

• for the line with ozonation

$$V_{M} = \left(240 - 247 \frac{C}{C_{0}}\right) \times 10^{5}$$
(9)

with boundary condition $C/C_0 \leq 0.97$.

The run time of the GAC bed with the assumed depth to the moment in which a target adsorbate concentration is obtained is described by an equation:

$$t_{c(C/C_0)_{\rm AC}} = \frac{H_{\rm GAC, AS} - H_{\rm GAC, \min(C/C_0)_{\rm AC}}}{V_{M(C/C_0)_{\rm AC}}}$$
(10)

The analysis of the process in the state of adsorptive capacity exhaution (Figs. 2 and 3, and the state of equilibrium) enables one to determine the parameters of isotherms of the Freundlich type. These parameters are presented in Tables 4 and 5.

Table 4

Parameters of GAC bed in the state of equilibrium (line without ozonation)

H _{GAC} [m]	M _{GAC} [kg]	<i>tc</i> [d]	$\frac{C}{C_0}$	M _{DOC} [g]	$x = \frac{M_{\text{DOC}}}{M_{\text{GAC}}}$ $[g \cdot kg^{-1}]$	С	lnx	lnC
1.5	194.5	80	0.62	3582.2	18.42	2.48	2.91	0.91
1.1	142.6	70	0.68	2658.1	18.64	2.72	2.92	1.00
0.6	77.8	50	0.80	1514.6	19.47	3.20	2.97	1.16

Table 5

Parameters of GAC bed in the state of equilibrium (line with ozonation)

H _{GAC} [m]	M _{GAC} [kg]	<i>tc</i> [d]	$\frac{C}{C_0}$	M _{DOC} [g]	$x = \frac{M_{\text{DOC}}}{M_{\text{GAC}}}$ $[g \cdot kg^{-1}]$	С	lnx	lnC
1.5	194.5	80	0.65	3671.6	18.88	2.60	2.94	0.96
1.1	142.6	70	0.72	2741.7	19.22	2.90	2.96	1.06
0.6	77.8	45	0.80	1585.3	20.38	3.20	3.01	1.16

Equations of isotherms have the following forms:

• the line without ozonation

$$x = 14.3C^{0.27} \tag{11}$$

• the line with ozonation

$$x = 14.3C^{0.29} \tag{12}$$

As an example of the model application, the bed run time $t_{c (C/C_0)_{AC}}$ for the assumed efficiency $(C/C_0)_{AC}$ and bed depth $(H_{GAC, AS})$ have been calculated. For $C/C_0 = 0.5$ and the bed depth 2.0 m the bed run time is equal 58 and 57 days for the technological lines without and with ozonation, respectively. For $C/C_0 = 0.75$ and the bed depth 2.0 m the bed run time is equal 133 and 128 days, respectively.

4. CONCLUSIONS

• Mathematical description of phenomena in the course of GAC bed adsorption shows differences not significant from a statistical point of view in the efficiency of the process, and the capacity of adsorption for compounds measured as DOC in ozonated and non-ozonated water.

• The presented models apply to dissolved compounds with the small size of particles and high velocities of their diffusion.

• For this particular case of the Oława River water, after coagulation, sedimentation, and rapid sand filtration, the process of ozonation does not change the size of dissolved particles.

• The statics of adsorption for both lines are described by Freundlich isotherms with similar parameters.

• The need for ozonation before adsorption ought to be confirmed in a laboratory study.

• Ozonation introduced to the system of treatment periodically ought to be taken into consideration.

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