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## Research of ozonation process of the biologically pre-purified municipal wastewater

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The paper considers the problem of post-treatment of municipal wastewater in Odessa using ozone. A significant place is given to the effect of the breakdown of toxic substances and important carcinogens. Based on theoretical studies, it was concluded that biological wastewater treatment does not guarantee a significant effect of the neutralization of many toxicants. The authors hypothesize the significant advantage of ozonation for the disposal and disinfection of wastewater compared to other methods, for example, chlorination. An important place is occupied by the search for optimal ozone treatment parameters. In laboratory conditions, data for mathematical modeling and optimization have been obtained. It has been established that the best treatment effect is achieved for the main wastewater pollutants, especially for synthetic surface-active substances (90 %) and BOD<sub>5</sub> (65 %). Moreover, the processing time is 15 minutes at a dose of ozone of 20–25 mg/l. The concentration of total nitrogen decreases the least (20 %). It was found that a significant effect of disinfection by *E. coli* during ozonation is achieved by treating wastewater for 4 min at a dose of 3 mg/l of ozone. In a mixture of wastewater and natural water, this effect is achieved in 24 hours. The author's hypothesis of the gentle action of ozonation products for the widespread aquatic culture of *Chlorella pyrenoidosa* is confirmed. A significant effect of destruction under the influence of ozone of carcinogens was observed. It was found that up to 82 % of benzpyrene in wastewater is destroyed within 16 minutes at a dose of 15 mg/l. The authors conclude that ozonation is a universal method for the purification of biologically treated municipal wastewater, which has a significant effect on the destruction of many toxicants and pathogenic microflora, without the presence of ozonation products that inhibit the development of natural aquatic cultures. Further research should focus on finding low cost ozone synthesis methods.

**Key words:** ozone, municipal wastewater, wastewater characteristics, wastewater treatment, wastewater ozonation, 3,4 benz (a) pyrene, *E. coli*, wastewater disinfection, modeling of the ozonation process, optimization of ozone regimes.

### Introduction

Urban sewage often comes after treatment with low water exchange capacity. In cities where the industry is a developed, biological treatment in urban wastewater treatment plants is in many cases insufficient in order to effectively remove residual contaminants. The concentration of these substances at the point of discharge of wastewater often exceeds the limit values for reservoirs of economic, drinking and cultural purposes. This is why the deep purification of biologically treated wastewater is a vital process for the environment. This fact started to be of particular importance when among the pollutants are

identified heavy oxidizing synthetic surfactants (OSSs), petroleum products, dyes, carcinogens, etc of particular importance are blastomogenic or carcinogenic compounds, among which polycyclic aromatic hydrocarbons are particularly prominent because of their widespread distribution. Medium is the most dangerous gasoline. The essential sources of pollution of the reservoirs of benzpyrene are municipal wastewater, atmospheric air, production processes accompanied by the spillage of petroleum products and the like.

In addition, the wastewater contains pathogenic microflora (Delzell et al., 1994; Rusanova & Ovechkina, 2002; Ivanko & Bidenko, 2012; Dai et al., 2013).

Additional treatment of biologically treated municipal wastewater can be carried out by chemical reagents, most often chlorine and its derivatives, physical (thermal, electrical, electromagnetic, etc.), chemical reactions (sorption, etc.) and other methods (Ivanko & Bidenko, 2012).

In Ukraine, chlorination is most commonly used in urban wastewater treatment plants. This process is quite simple to implement, but is associated with the formation of organochlorine compounds that are harmful to the human body. Significant inconveniences of chlorine use are related to adherence to special safety rules at all stages of the process (White, 1992; Rusanova & Ovechkina, 2002; Kim et al., 2006; Delzell et al., 2008; Li et al., 2013; Dai et al., 2013).

Chlorine dioxide has some advantages over chlorine in the purification process, in particular a higher bactericidal effect and a lower residual concentration in wastewater. However, the concentration and number of by-products of treatment in this case is higher than when using chlorine (Petrenko et al., 2007; Otterholm & Jadesjo, 2000; Vaezi et al., 2004; Solovieva & Maliuchenko, 2005).

Many researchers say that ozonation, despite some economic disadvantages, is a promising way to additional cleaning. Ozone is able to react under mild conditions with many organic, organo-organic and inorganic compounds. Thermodynamically, these reactions can lead to complete oxidation that is to the formation of water, carbon monoxide and higher oxides of other elements. An obstacle to complete oxidation is the low reaction rates at the final stages.

The standard redox potential of ozone in an acidic medium is 2.07 V, the product of the interaction of ozone with water of a hydroxyl radical is 2.8 V, which is the main cause of ozone activity against various kinds of water pollution, including microorganisms. The chemistry of many of these processes has been studied in great detail (Davis, 1981; Ivanova et al., 1985; Naidenko et al., 1985; Romanenko et al., 1987; Alekseev, 2002; Gehr et al., 2003; Kuznetsov, 2008; Grynevych et al., 2008; Rodríguez et al., 2008; Silva et al., 2010; Ushchenko et al., 2011; Tripathi et al., 2011; Lazarova et al., 2013; Yusuphuzhaeva, 2017). Ozone has high bactericidal and virucidal activity (Ivanova et al., 1985; Kuznetsov, 2008). In (Naidenko et al., 1985), the parameters of sewage ozonation are proposed, which give a high oxidizing effect in an alkaline environment. For achieving good effect, the authors propose to use ozone at doses of 450–550 mg/dm<sup>3</sup> and the consumption of ozone for domestic wastewater 300 g/m<sup>3</sup>. The processes occurring in the treatment of water with ozone are direct reactions with dissolved compounds, their decomposition to secondary oxidizers, such as highly reactive OH and HO<sub>2</sub> radicals, the formation of additional secondary ozone oxidizers, which react with other impurities, such as the formation of ozone oxidizes bromide ions, sequential reactions of these secondary oxidants with dissolved water pollutants. Reactions involving intermediates such as hydrogen peroxide, an extremely active hydroxyl radical moiety, play a major role.

The main role in these processes is played by hydroxyl radical – an extremely strong oxidizing agent that oxi-

dizes organic compounds by separation of a hydrogen atom:  $HRH + HO \cdot - HR \cdot + H_2O$ . As a result, organic radicals are formed that initiate the chain reactions of oxidation. Sulfur-containing impurities (synthetic detergents, hydrogen sulfide, sulfur dioxide, rhodanides) in water make up a significant proportion in the spectrum of pollutants. All of these compounds are oxidized by ozone. The most actively interact with ozone those in which the molecule has a double bond C = S. Oxidation of impurities in the treatment of wastewater with ozone does not exhaust the entire spectrum of its action. Inhibiting the growth of bacteria, viruses, algae and other aquatic organisms, ozone acts as a disinfectant.

Davis (Davis, 1981) notes the effect of rapid decomposition of ozone in wastewater and offers a multi-stage treatment scheme. In this case, there is annihilation of ascarid eggs at ozone doses of 209–357 mg/m<sup>3</sup>. The effect of the destruction of helminths during ozonation for 90 min is noted in studies (Romanenko et al., 1987).

Toxicological studies of the safety of water ozonation began to develop intensively only in the 80-ies of the last century. This is explained by the difficulty of developing the methodology for such studies, the need to evaluate the toxicity of a very large number of organic impurities, taking into account their mutual influence in different combinations, the influence of the environment and the like.

The analysis of these and other published works shows a great practical experience of ozone application, mainly for wastewater disinfection.

However, the effect of ozone disinfection on many chemical compounds should be noted.

It is important to take into account the possible negative impact of the decomposition products of these substances on the water bodies of the reservoirs, which receive treated municipal wastewater. Along with toxicological, hygienic studies are conducted on the safety of the use of ozonators. It is necessary to take into account the performance of the installation on ozone, the degree of excess of the MPC of ozone and nitrogen oxides in the air of the working area, the migration of substances into the air from the material of the ozonator body, adverse physical factors: noise, vibration, radiation, etc. (Rodríguez et al., 2008; Lazarova et al., 2013).

Thus, for the practical application of ozonation for wastewater treatment, a more detailed study of the contamination of major pollutants and the choice of modes that provide a high and lasting effect are required.

### Purpose and task of the research

The purpose of the work is to clarify the parameters and establish the feasibility of ozonation of urban wastewater that has undergone biological treatment for deep purification and disinfection.

The purpose of research is to be achieved by solving the following issues:

1. Establishing basic dependencies on ozone treatment to reduce biological and chemical oxygen demand (BOD and COD).
2. Establishment of optimal sewage ozonation parameters.

3. Determination of the main dependencies of wastewater disinfection during ozone treatment.

4. Assessment of the degree of toxicity of the decomposition products of pollutants under the influence of ozone.

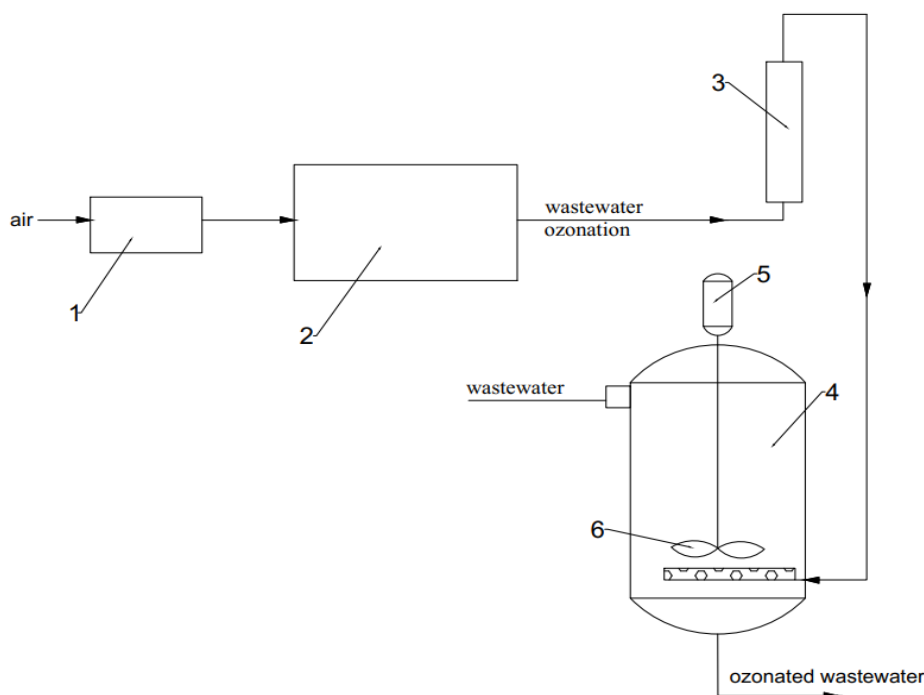
5. Determination of the effect of neutralizing carcinogenic compounds in sewage ozonation.

### Materials and methods

Samples of biologically treated sewage treatment plants in Odesa and the waters of the Khadzhibeiv estuary served as objects of research.

In biologically treated wastewater the concentrations of the major contaminants, namely, suspended solids,

BOD5, CBS, nitrogen, phosphates, phenols, OSSs and color were determined (Lurie, 1984). The concentration of E. coli was determined by (Ineshina & Gomboeva, 2006). The biological mass of *Chlorella pyrenoidosa* was determined using Goryaev's camera (Portnaia & Saltanov, 2015). Biological and microbiological research was carried out at the Odesa National Medical University. Benzpyrene in wastewater samples was determined by (PNA F 14.1:2:4.186-02). Wastewater ozone treatment was carried out at a laboratory facility equipped with an Oz-2 ozonizer with a capacity of up to 3 g/h of ozone (manufactured by Aqua, Ukraine). The scheme of the laboratory installation is shown in Fig. 1.



**Fig. 1.** Scheme of an experimental plant for ozonation of wastewater: 1. Compressor, 2. Ozonizer, 3. Rotameter, 4. Mixer, 5. Electric motor, 6. Mixer.

The air was supplied to the ozonator by means of a compressor, and then through a rotameter into a mixer with a mechanical stirrer. In the ozonator was the formation of ozone from the oxygen contained in the air. Ozonized air was fed into the tank with sewage, which was continuously mixed with a mechanical stirrer to distribute ozone uniformly over the volume of water. The ozone flow rate was monitored using a rotameter. The ozone dose was determined by the air flow rate and the ozone concentration, taking into account the mixer operating capacity. Stirring was performed at a frequency of 20 min<sup>-1</sup>. The time of ozone treatment was controlled by a timer. The air was pumped into the ozonator by means of a compressor and then through a rotameter into a mixer with a mechanical stirrer. Ozone dosage was determined by air flow and ozone concentration. Stirring was carried out continuously at a frequency of 20 min<sup>-1</sup>. The time of ozone treatment was controlled by a timer. Ozonated wastewater was analyzed for contaminants. The results

were used to optimize the ozonation process. The ozonized sewage was mixed with the water of the Hadzhibeiliman, which served as a natural reservoir to determine the behavior of *Clorella*. The mixing ratio was 0.25.

### Results and discussion

Mathematical processing of these researches and their planning led to the realization of a full factorial experiment. The results became the basis for optimization by method ascent steep.

Previous researchers have found that a significant effect of COD neutralization and SSOs concentration is achieved at an ozone concentration of wastewater of 18-20 mg/l at a duration of about 12.5 minutes.

The experiment planning matrix is presented in Table 2. The data for the matrix design are shown in Table 1.

**Table 1**  
Data for the matrix of the full factor experiment 2<sup>2</sup>

| Factor             | COD            |                  | Concentration SSS |                  |
|--------------------|----------------|------------------|-------------------|------------------|
|                    | Duration, min. | Ozone doze, mg/l | Duration, min.    | Ozone doze, mg/l |
| Code               | X <sub>1</sub> | X <sub>2</sub>   | Z <sub>1</sub>    | Z <sub>2</sub>   |
| Main level (0)     | 12.5           | 20               | 12.5              | 18               |
| Variation interval | 5              | 5                | 5                 | 5                |
| Upper level (+)    | 17.5           | 25               | 17.5              | 23               |
| Lower level (-)    | 7.5            | 15               | 7.5               | 13               |

**Table 2**  
Experiment planning matrix 2<sup>2</sup>

| Factor                           |                                  |                                  |  | Reduction effect |                   |
|----------------------------------|----------------------------------|----------------------------------|--|------------------|-------------------|
| X <sub>0</sub> (Z <sub>0</sub> ) | X <sub>1</sub> (Z <sub>1</sub> ) | X <sub>2</sub> (Z <sub>2</sub> ) | X <sub>1</sub> X <sub>2</sub> (Z <sub>1</sub> Z <sub>2</sub> ) | COD              | SSO concentration |
| +                                | +                                | +                                | +  | Y <sub>1</sub>   | Y <sub>2</sub>    |
| +                                | -                                | -                                | +  | 37.5             | 82.2              |
| +                                | -                                | +                                | -  | 18.3             | 81.8              |
| +                                | +                                | -                                | -  | 36               | 88.5              |
|                                  | 2.5                              | 7.1                              | -1.75  | 25.8             | 60.8              |
|                                  | -6.8                             | 7                                | 3.9  |                  |                   |

The result is mathematical expressions

$$Y_1 = 29.65 + 2.5 * X_1 + 7.1 * X_2 - 1.75 * X_1 * X_2$$

$$Y_2 = 78.3 - 6.8 * Z_1 + 7 * Z_2 + 3.9 * Z_1 * Z_2$$

Where Y<sub>1</sub>, Y<sub>2</sub> – COD reduction effect and SSO concentration accordingly.

The steep ascent method determines the optimal values of ozonation parameters for deep sewage treatment for other contaminants. The results of the calculations are presented in Table 3.

**Table 3**  
Effect of reduction of wastewater pollution in the course of ozonation

| № | Indicator             | Concentration, mg/l |       | Ozone doze, mg/l | Duration, min | Reduction effect, % |
|---|-----------------------|---------------------|-------|------------------|---------------|---------------------|
|   |                       | Initial             | Final |                  |               |                     |
| 1 | Suspended items       | 20                  | 7     | 25               | 15            | 65                  |
| 2 | BOD <sub>5</sub>      | 16                  | 5.6   | 25               | 15            | 65                  |
| 3 | COD                   | 80                  | 50    | 20               | 15            | 40                  |
| 4 | Nitrogen              | 20                  | 16    | 20               | 15            | 20                  |
| 5 | Phosphates            | 5                   | 5     | -                | -             | -                   |
| 6 | Phenol                | 0.05                | 0.03  | 12.5             | 5             | 30                  |
| 7 | SSO                   | 1.3                 | 0.13  | 15               | 20            | 90                  |
| 8 | Coloring <sup>o</sup> | 123                 | 56    | 15               | 15            | 54                  |

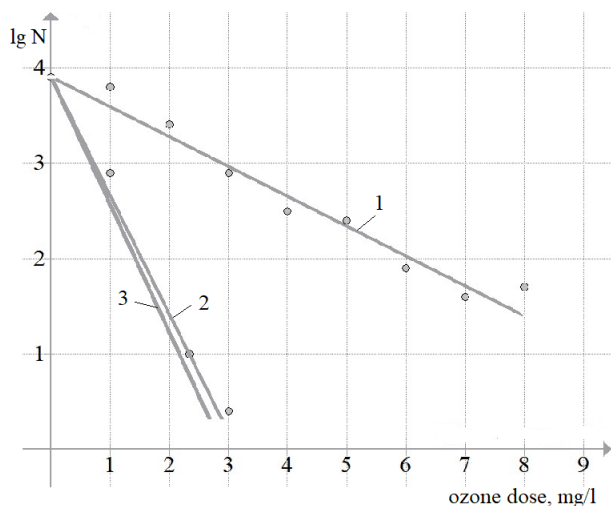
The ozonation parameters for the different pollution rates are slightly different. However, given the complex effect of ozone on pollutants at the same time, the optimal values should be considered ozone dose of 20–25 mg/l wastewater with treatment duration of 15 minutes. However, these parameters do not guarantee the effect of phosphate neutralization. The effect of ozonation on the total nitrogen bridge is the least studied.

In order to treat the wastewater, they were treated for 2–8 minutes at doses of ozone 0.8–8 mg/l. The test culture in the experiments served the bacteria *E. coli*. The results of the experiments are presented graphically in Fig. 2. The number of viable microflora after treatment was evaluated by the logarithm of the number of cells. According to the data in order to achieve a significant bactericidal effect, the dose of ozone is about 3 mg/l with a process duration of 4 minutes. Lower doses and increased ozonation time do not produce the desired result. The determined parameters can be considered as calculated in

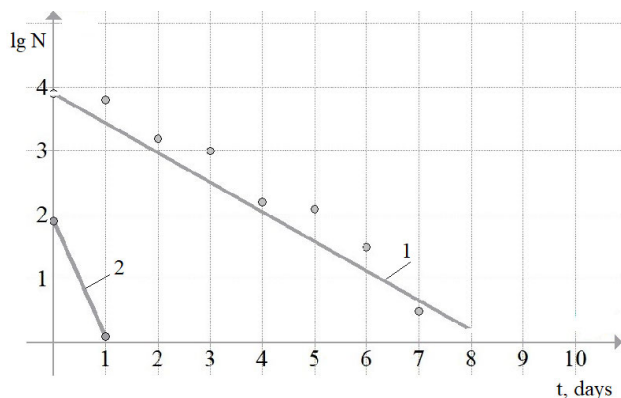
the development of ozone disinfection regimes of biologically treated municipal wastewater.

It is known that the consequences can be important in any treatment method for the treatment of wastewater. It is extremely important, for example, to influence the biota of the reservoir, which receives the treated wastewater. These consequences include the degree of inactivation of the *Escherichia coli* and the potential for restoration of its growth during long-term aeration.

To determine the effects identified the wastewater was mixed with the water of the Hadzhibei liman in proportion, which approximately corresponds to the actual reset conditions. Under laboratory conditions of gas exchange without artificial aeration, the logarithm of the number of viable *E. coli* cells was determined. Exposure of samples of biologically treated sewage with reservoir water and the same mixture with ozonized sewage produced the results, which are shown in Fig. 3.



**Fig. 2.** Dynamics of wastewater disinfection by ozonation (Test culture *E. Coli*): 1 –  $\tau=2$  min; 2 –  $\tau=4$  min; 3 –  $\tau=6$  min.



**Fig. 3.** Dynamics of wastewater disinfection in a mixture of treated and ozone reservoirs (Test culture *E. coli*): 1 – biologically treated wastewater + reservoir water; 2 – ozone sewage + reservoir water.

The results of the experiments prove that in the mixture of ozonized wastewater and natural water, the active destruction of *Escherichia coli* is achieved within the first 24 hours. Control experiment with non-ozonated biologically treated wastewater (curve 1) gave the same effect only after 7 days.

A known fact is the oxidation of ozone by many organic compounds contained in wastewater. The degrada-

tion of these compounds can lead to the accumulation of products with unknown or little-known degree of danger and chemical structure.

Thus, the effectiveness of ozonation will not be fully determined without taking into account the hygiene component.

The toxicity of treated water is detected by various tests using bacteria, laboratory animals, molecular structural studies. The analysis of the results of toxicological studies suggests that ozone treatment can eliminate the mutagenic activity of the source water, do not affect this property, slightly increase it. However, the mutagenic activity in this case remains lower than the chlorination of the same samples. With respect to the toxicity of such possible ozonation products as peroxides, epoxides and unsaturated aldehydes, their presence in water has not been established because these compounds are prone to biodegradation and decompose rapidly as they pass through the water distribution network. Possible toxicity of ozonation products can, for example, be determined by the behavior of the microflora and microfauna of the reservoir.

*Chlorella pyrenoidosa* is a widespread in natural ponds. If taken as a test culture the biomass growth rates, to can be determine the toxicity of compounds that enter the reservoir with ozonized sewage. In the presence of negative impact, the inhibition of test culture development should be investigated. The dynamics of biomass accumulation of *Chlorella pyrenoidosa* are shown in table 4.

The results of the experiments confirm that, after ozonation, inhibition of growth of *Chlorella pyrenoidosa* was not detected in any of the samples of the mixture of wastewater with natural. For example, all samples of the mixture under laboratory conditions under natural light the concentration of microalgae cells, which was determined in the Goryaev chamber for 10 days steadily increased from  $10^8$  to  $10^9$  cells/ml. This fact indicates that the products do not show marked toxicity to the selected test culture. Other species may need additional research.

The results of experiments confirm that sustained stimulation of *Chlorella pyrenoidosa* growth occurs after ozonation. This fact indicates that the ozonation products do not show marked toxicity to the selected test culture. Other species may need additional research.

**Table 4**  
Dynamics of biomass accumulation of *Chlorella pyrenoidosa*, mcg/l

| Exposure time, days | Control (reservoir water) | Ozone sewage + reservoir water |
|---------------------|---------------------------|--------------------------------|
| 0                   | 99.3                      | 95.5                           |
| 5                   | 95.3                      | 103.5                          |
| 10                  | 134.2                     | 141.6                          |
| 15                  | 229.5                     | 239.5                          |
| 20                  | 362.3                     | 370.5                          |

Many chemical compounds contained in urban wastewater have a carcinogenic effect. In this sense, the multi-nuclear aromatic hydrocarbon 3,4 benzapyrene (BP), harmful effect which is confirmed by numerous

experiments on animals, can be considered a universal indicator of environmental pollution. Today, BP concentration is a kind of indicator for all polycyclic aromatic carbohydrates in the environment. This is due to its very

high resistance. BP is found wherever such hydrocarbons are identified. Carcinogens contained in different objects of the aquatic environment in different concentrations,

depending on the degree of general contamination of water bodies.

The results of experimental studies of the destruction of BP in the sewage ozonation are shown in [table 5](#).

**Table 5**

Experimental data on the effect of reducing the concentration of BP

| Indicator | Concentration, mg/l |       | Ozone doze, mg/l | Duration of treatment, minute | Neutralization effect, % |
|-----------|---------------------|-------|------------------|-------------------------------|--------------------------|
|           | Initial             | Final |                  |                               |                          |
| Example 1 | 0,5                 | 0,16  | 3                | 4                             | 68                       |
| Example 2 | 0,4                 | 0,10  | 6                | 8                             | 74                       |
| Example 3 | 0,4                 | 0,08  | 9                | 12                            | 78                       |
| Example 4 | 0,5                 | 0,09  | 12               | 16                            | 81                       |
| Example 5 | 0,6                 | 0,11  | 15               | 16                            | 82                       |

Obviously in order to effectively reduce the concentration of carcinogenic compounds, wastewater should be treated for at least 4 minutes with a minimum ozone dose of 3 mg /l. if the dose of ozone is increased to 12 mg/l, while increasing the duration of treatment, the effect of BP neutralization is 80–81 %. Interestingly, the additional increase in dose and duration had little effect on the residual BP concentration (sample 5).

### Conclusions

Laboratory studies have confirmed the feasibility of ozonation of biologically treated wastewater to neutralize pollutants and disinfect. The main dependencies of the ozonation process were determined by the methods of mathematical planning of experiments and optimization. For major pollutants the optimal ozone treatment parameters are a dose of ozone of 20–25 mg/l for a duration of 15 minutes.

In this case, it is possible to achieve a reduction of contamination rates for COD by 40%, BOD<sub>5</sub> by 65%, suspended solids by 65%, SSO by 90%, phenols ha 40%.

It is established that ozonation leads to effective disinfection of wastewater at a dose of 3 mg/l ozone for 4 minutes. Ozonation products have been found to be non-toxic to typical *Chlorella pyrenoidosa* water bodies. Studies of several wastewater samples containing 0.4–0.6 µg/l benzpyrene after ozonation showed a 68–82 % destruction effect. Carcinogens depending on processing parameters. Further studies on the prospect of municipal wastewater ozonation may relate to seeking to reduce the economic performance of the treatment process.

### Conflict of interest

The authors declare that there is no conflict of interest.

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