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The disinfection of air in premises, especially in the presence of people, is an important element of the system for the prevention of infectious diseases. A promising solution for this is to fill the premises with an antimicrobial chemical agent that would quickly neutralize pathogens directly at the moment they enter the air. It is advisable to use hypochlorous acid HOCl as such an agent. But traditional methods of administering HOCl into the air by aerosolizing its solutions may be accompanied by a number of risks that can be prevented by using gaseous HOCl. Therefore, it is relevant to develop evaporation devices that would take into account the specifics of chlorine-active compounds and determine the influence of various factors on the release of gaseous HOCl from them into the air being treated. During the study, a design of a device was developed in which the transfer of HOCl into the air is carried out by contact of the latter with a foamed working solution of sodium hypochlorite NaOCl. The HOCl concentration in the treated air mainly depends on its concentration in the working solution, which, in turn, is determined by the pH of the solution and the total content of free chlorine in it. Additionally, the influence of the air and working solution temperatures, the volume of the working solution, the power of air flows and other technological factors on the air HOCl concentration was studied. When using an electrochemically generated approx. 1000 mg/l NaOCl solution with pH 8.50–8.60 at 20 °C, it is possible to maintain the total chlorine concentration in the air flow with a capacity of 50 m³/h at a level of about 0.30 mg/m³ for a long time. Simple design, multifunctionality and the fundamental possibility of combining the processes of electrochemical synthesis of HOCl and its immediate administering into the air open up broad prospects for using the developed installations for continuous disinfection of premises

Keywords: hypochlorous acid, sodium hypochlorite, active chlorine, antimicrobial agents, air disinfection, gas emission, evaporation devices, infection prevention

DETERMINATION OF THE PATTERNS OF HYPOCHLOROUS ACID RELEASE FROM FOAMED SODIUM HYPOCHLORITE SOLUTIONS INTO THE AIR IN AN EVAPORATOR OF SPECIAL DESIGN

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1. Introduction

Recent events related to the Covid-19 pandemic have demonstrated that their prevention, rather than treatment, is the most affordable and effective strategy for minimizing the negative socio-economic consequences of such phenomena [1]. Many studies emphasize the need for widespread implementation of disinfection and antiseptic measures to combat the spread of infectious diseases [2]. Regular treatment of surfaces with chemical disinfectants of various classes (oxidizers, metal salts, surfactants, etc.), as well as the use of sanitizers, makes it relatively easy to minimize the transmission of infection by contact [3]. However, it is known that some dangerous diseases (various types of influenza, tuberculosis, measles, etc.) are transmitted by an aerogenic mechanism [4]. In this

case, infection is possible both directly by inhaling air infected by a human carrier, and subsequently by contact, when microbial/viral aerosol settles on various surfaces. Therefore, air purification from pathogenic microorganisms is an important and at the same time complex technological task.

2. Literature review and problem statement

There are many approaches to air purification. The most common is the equipment of premises with supply and exhaust ventilation systems with filter elements that physically retain viruses and microorganisms [5]. Various polymers are mainly used as filters [6]. Some of such filters can often additionally have microbicidal properties, for example, due to the presence

of immobilized peroxy [7] or N-halamine [8] groups. However, for such a system to be effective, its power must be high, which requires significant energy costs and, as a rule, can be achieved only in small special-purpose rooms (surgical operating rooms, microbiological laboratories, etc.). In addition, regular regeneration or replacement of filters is necessary. Another way to disinfect air is its treatment with UV radiation, which is carried out both with the help of open lamps and lamps isolated in the housing of special devices – bactericidal recirculators [9]. The ozonators are most effective in this case [10]. However, ozonation of sufficient power cannot be carried out in the presence of people, and recirculators process only a small portion of air at a time. Currently, no method solves the problem of transmission of infection from person to person during close contact. Therefore, the most promising technology seems to be filling the entire volume of the room with a certain chemical disinfectant, which would neutralize pathogens immediately at the moment of their appearance in the air and would be harmless to humans.

As such a compound, hypochlorous acid (HOCl) deserves the most attention [11]. Its advantages are a wide spectrum (it is active against viruses, bacteria, fungi and prions) and a high rate of antimicrobial action, the absence of resistance of microorganisms to it, as well as high tolerability due to its endogenous nature, namely, participation in the process of phagocytosis in the formation of an immune response to various types of intoxication and inflammation [12]. Another important factor is the simplicity and cheapness of its production. Hypochlorous acid, together with other forms of active chlorine (hypochlorite ion ClO^- and molecular chlorine Cl_2), is part of a large-scale (according to various estimates, 3.5–4.0 million tons per year) industrial product – sodium hypochlorite (NaOCl), which can be obtained from available raw materials by chemical (dissolution of molecular chlorine in a sodium hydroxide solution) or electrochemical (electrolysis of sodium chloride solutions) synthesis [13]. The HOCl content in these solutions depends on their pH [14]. The combination of the above factors determines the widespread use of HOCl in medicine for antiseptic purposes [15], treatment of skin diseases and wounds [16], infectious diseases [17], etc., along with other chlorine-active drugs (NaOCl [18], N-chlorotaurine [19, 20] and other N-chloramines). Its use for air disinfection is also actively studied. Thus, studies show good results against airborne viruses [21] and bacteria [22, 23]. However, in this context, the technology of administering HOCl into the air and its hardware design are of great importance.

Currently, the main method of air treatment with HOCl is aerosol spraying of NaOCl solutions of various compositions. Being a volatile compound, HOCl evaporates from the surface of aerosol droplets, realizing its antimicrobial properties [23]. However, this approach has a number of significant drawbacks: the humidity in the room increases sharply, and the accompanying technological impurities (mainly sodium chloride, chlorate and hydroxide) contaminate surfaces, can have a toxic effect, are corrosive and pose a danger to the operation of electrical devices. It is much safer to introduce HOCl into the air in a gaseous state. A number of studies demonstrate the high efficiency of this disinfection method. Thus, it has been shown that even at a concentration of less than 100 ppm, gaseous HOCl is able to quickly kill bacteria both in the air [24] and on nutrient media [25]. At the same time, biochemical studies prove the low inhalation toxicity of such a procedure. In the research [26] on the model respiratory tract, it was proven that HOCl concen-

trations below 500 ppb do not lead to deviations in the viability of human nasal and bronchiolar epithelial cells even with prolonged exposure. A study on rats demonstrates [27] that at high (up to 5 mg/m^3 in terms of total chlorine) concentrations, gaseous HOCl after 4-hour exposure does not lead to the death of laboratory animals and can induce some antioxidant enzymes. However, toxicological studies on humans have not been conducted yet. The transfer of HOCl from solution to the gas phase is possible in forced-air evaporation systems of various types. In general, these are devices in which the air flow to be treated contacts a developed surface wetted with the working solution, from which the evaporation of volatile components occurs. For example, commercially available air purifiers of Venta type [28] and similar ones are suitable for such purposes. The principle of operation of such evaporators is based on the creation of a developed solution/air interface surface by rotating wettable polymer disks, which are partially immersed in the working solution and blown with air using a fan. However, the use of NaOCl as a working solution in such devices has a number of disadvantages. First, this is due to the high carryover of water, which entails a rapid concentration of the solution and a sharp increase in humidity in the room being treated. Second, all the working mechanisms of such evaporators are located in close proximity to a rather aggressive solution, which creates a risk of corrosion of metal components. In addition, such systems are quite technologically complex, and their integration into supply ventilation systems is difficult. There are devices in which, instead of rotating disks, porous or mesh bodies are used to ensure more intensive HOCl evaporation, which are wetted with a chlorine-active solution and blown with air [24, 25]. But they provide constant emission parameters only in flow systems, which are constantly supplied with a fresh working solution with specified characteristics (temperature, concentration, pH, flow rate, etc.). In accumulative systems, active chlorine quickly decomposes on a developed evaporation surface, which leads to difficult-to-predict changes in the transition of HOCl to the air. In addition, none of the studies conducted using evaporators of various designs describes the dependence of the release of gaseous HOCl (HOCl (g)) into the air on the composition and properties of its solutions, aerodynamic characteristics of the process, etc. Therefore, there is a need to develop specialized devices for treating rooms with HOCl (g) and studying related processes.

3. The aim and objectives of the study

The aim of this study is to establish the patterns of HOCl transition from foamed NaOCl solutions into the air. This will allow in the future to create devices suitable for use as elements of a complex disinfection system, and with their help to develop mechanisms for maintaining the effective concentration of HOCl (g) in the indoor air to ensure maximum antimicrobial effect in the absence of toxicity. To achieve the goal, it was necessary to solve the following tasks:

- to develop a design and create a working prototype of an evaporation device with forced air circulation and a working solution foaming system, which would allow to study the maximum number of factors affecting the emission of volatile components from their solutions;

- to study the dependence of the HOCl (g) concentration in the air processed in the developed evaporator on the properties of the working NaOCl solution and the technological operating parameters of the device.

4. Materials and methods

4.1. Object and hypothesis of the research

The object of this study is the emission of HOCl (g) into the air, which is carried out when the latter is brought into contact with a NaOCl solution in an evaporator of a special design. The main hypothesis of the study is that the air concentration of HOCl (g) when using such a device can be varied in a wide range. This is achieved both by changing the parameters of the working solution (active chlorine content, pH, temperature, etc.), and by regulating the properties of the air flows being processed. It was assumed that the patterns obtained for a NaOCl solution of a certain concentration can be extrapolated to solutions with a different concentration of active chlorine. Therefore, to simplify the experiment, the main study was carried out using a working solution of approximately 1000 mg/l NaOCl.

4.2. Working solutions of NaOCl and their analysis

To obtain working solutions with a free chlorine concentration of less than 1.0 g/l, a commercially available preparation SEKOBREN (Kyiv, Ukraine) was used, which is a highly pure electrochemically generated NaOCl solution on a background of 8 g/l NaCl, synthesized using a special technology and approved by the Ministry of Health of Ukraine, including for aerosol air disinfection [29]. To prepare more concentrated solutions, a commercially available 20 % NaOCl solution (PCC Rokita, Poland) was used, which was added to SEKOBREN in the calculated amount. To dilute NaOCl solutions, solutions of NaCl in water purified by reverse osmosis were used so that the NaCl content in the diluted solutions remained equal to that in the original solutions. The free chlorine concentration in working solutions was determined by the standard iodometric titration [29]. To acidify the working solutions, a minimum amount of 30 % hydrochloric acid HCl was used. To saturate the working solution with carbon dioxide CO₂, a chemically pure gas from a cylinder was used. The acidity of the solutions was controlled by a pH meter ADWA AD-131 (ADWA Instruments, Hungary).

To calculate the ratio of molar fractions of active chlorine components (hypochlorite ion ClO⁻, HOCl and molecular chlorine Cl₂) in the working solution, the well-known Pourbaix diagram [14] (Fig. 1) was used.

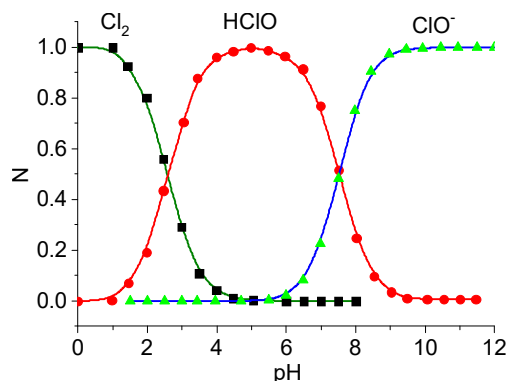


Fig. 1. General diagram of molar fraction of Cl₂, HOCl and OCl⁻ distribution in the studied NaOCl solutions

4.3. Method for determining the concentration of HOCl (g) in air

The HOCl concentration in the air was determined using the method described in work [27]. The method is based on sampling air by bubbling a given volume of it through the

absorption solution of taurine, which results in immediate formation of N-chlorotaurine. The total chlorine concentration in absorption solution was determined photometrically using an eXact Micro 20 photometer with the corresponding eXact Strip Micro Total Chlorine DPD 4 reagent strips (Industrial Test Systems, Inc., USA). Hereinafter, total chlorine is understood to mean the sum of free (HOCl and OCl⁻) and combined (N-chloramines of various structures) chlorine. The concentration of total chlorine in the air was calculated using the formula (1):

$$C_{TC} = \frac{(C_{AS} - C_{blank}) \times V_{AS}}{1000 \times V_{air}}, \quad (1)$$

where C_{TC} – total chlorine concentration in the air, mg/m³; C_{AS} – colorimetrically determined total chlorine concentration in the absorption solution, mg/l; C_{blank} – apparent total chlorine concentration in the “blank” sample, mg/l (by “blank sample” let’s mean the initial taurine absorption solution, analyzed photometrically before passing air from the installation through it); V_{AS} – volume of absorption solution, ml; V_{air} – volume of air sampled for analysis, m³. Since our experiments exclude the possibility of the formation of volatile Cl₂, and NaOCl is not volatile, the amount of total chlorine in the air can be considered equal to the amount of released HOCl (g). All experiments were repeated under identical conditions at least in triplicate.

4.4. Initial requirements for the prototype of the evaporation device

When developing the design of an evaporation device for transferring HOCl from solution to air and a detailed study of all factors affecting the emission process, the following requirements were taken into account:

- 1) the absence of metal parts that are in direct contact with the working NaOCl solution or are exposed to intense exposure to its aerosol;
- 2) manufacturing from chemically and corrosion-resistant materials;
- 3) a minimum of rotating and rubbing parts;
- 4) the possibility of easy integration into ventilation systems;
- 5) low noise level;
- 6) low energy consumption and low cost of components;
- 7) the possibility of varying the volume and temperature of the working solution, air temperature and air flow power in a wide range.

4.5. General method for conducting research on the process of HOCl (g) emission into the air during the operation of the developed evaporation device

The required volume of HOCl working solution, brought to a given temperature, with a predetermined concentration of active chlorine and pH, was poured into the evaporator container (Fig. 2). The mass of the evaporator container at the beginning of the experiment and the height of the working solution column in it were recorded. The solution temperature was maintained constant throughout the experiment using a heater. Then, the air flows were turned on in the installation, controlling their power, temperature, and CO₂ content. If necessary, the CO₂ concentration was increased by mixing gas from a cylinder into them. In most experiments, the power of air flow 2 was 50 m³/h, and flow 4 was 250 l/h (Fig. 2), but in some cases they were changed

according to the goals of the experiment. During the experiment, the pH of the working solution was constantly monitored, and its mass was also calculated periodically (at least once an hour). At the end of the experiment, the residual active chlorine concentration in the working solution was determined. Samples of air treated in the evaporator were taken at intervals of 10–20 minutes and analyzed for total chlorine content according to the method given in section 4.3. Thus, the influence of all main chemical and technological factors on the HOCl emission into the air was analyzed, namely:

1) the free chlorine concentration and the pH of the working solution, which affect the content of the volatile component in it;

3) the temperature of the working solution: its increase affects the solubility of HOCl and its evaporation rate;

4) the height of the column of the working solution: it was assumed that a significant contribution to the C_{TC} is made by air bubbles that come out of the bubbler and are saturated with HOCl as they move to the surface of the solution. The degree of saturation depends on the time from the formation of a bubble at the bottom of the container to its rise to the surface, i.e., other things being equal, on the height of the column of the working solution;

5) volumetric flow rate of air pumped by the compressor through the bubblers: with its increase, firstly, the amount of HOCl carried to the surface inside the bubbles increases, and secondly, the surface area of the working solution increases due to the formation of a larger amount of unstable foam;

6) volumetric flow rate and temperature of the air supplied through the inlet pipe, which affect the intensity of evaporation;

7) CO_2 concentration in both air flows, which, when absorbed by the working solution, are theoretically capable of changing its pH and, accordingly, the content of volatile HOCl in it.

5. Results of studies of the patterns of HOCl (g) emission into the air from foamed solutions in a designed installation

5.1. Development of a prototype evaporation device

The general scheme of the developed evaporation device is shown in Fig. 2.

The main element of the unit is a heat-insulated parallelepiped-shaped container 1 with an internal size of 29×16×28 cm, made of opaque polypropylene and hermetically sealed with a removable polypropylene lid. Polypropylene ensures stability of the chemical composition and concentration of the NaOCl working solution 2. At the bottom of the container there is a panel 3 with removable tubular chemically resistant ceramic bubblers 24 cm long, the number of which can be changed from two to six. The bubblers are connected to an external compressor 4 and ensure dispersion of the air flow pumped by it in the working solution by creating bubbles. The rate of this air flow is regulated in the range of 20–250 l/h using a rotameter 5 with a tap so as to minimize aerosol formation. Next to the bubblers on panel 3 there is a programmable electric heater with a PID thermostat in a chemically resistant plastic housing, which allows maintain-

ing a constant temperature of the working solution with an accuracy of 0.5 °C. The container is located on scales, which allows monitoring its mass loss due to the removal of volatile components with an accuracy of 1.0 g. Two polypropylene flanges are placed in the removable lid of the container. The first flange is equipped with a vertical inlet pipe 6 with a diameter of 100 mm and a length of 1.50 meters. The second flange is equipped with an outlet pipe 7 with a diameter of 100 mm and a polypropylene 90° pipe elbow. Through the inlet pipe 6, air is fed through the HEPA filter 8 via variable speed fan 9 with a duct heater 10, which allows maintaining its set temperature, into the container 1. The CO_2 content in the air is monitored by an AZ 7530 detector (AZ Instrument Corp., Taiwan), and its humidity is monitored by a Walcom HT-9600 (Walcom International Industry Limited, China). A Tenmars TM-4001 hot-wire anemometer sensor (Tenmars Electronics Co., LTD, Taiwan) can be introduced into the inlet pipe 6 through a sealed opening, allowing monitoring the temperature and speed of the blown air. In addition, the pipe 6 has a polypropylene outlet through which filtered air with a given temperature can be discharged into compressor 4. The outlet pipe 7 is equipped with a low-resistance polypropylene drip-eliminator filter 11, preventing the aerosol of the working solution from being carried out, as well as with nozzles that allow monitoring the air speed in it and taking air samples. It is possible to lower or raise pipes 6 and 7, adjusting the distance from their edge to the working solution surface. The outlet pipe 7 supplies air to an intermediate polypropylene chamber 12, equipped with a nozzle for taking air samples and a 100 mm pipe that discharges air from it into the laboratory exhaust ventilation system. The container lid also has a sealed opening for periodic monitoring of the pH and temperature of the working solution. Sampling of the working solution is possible via tap 13.

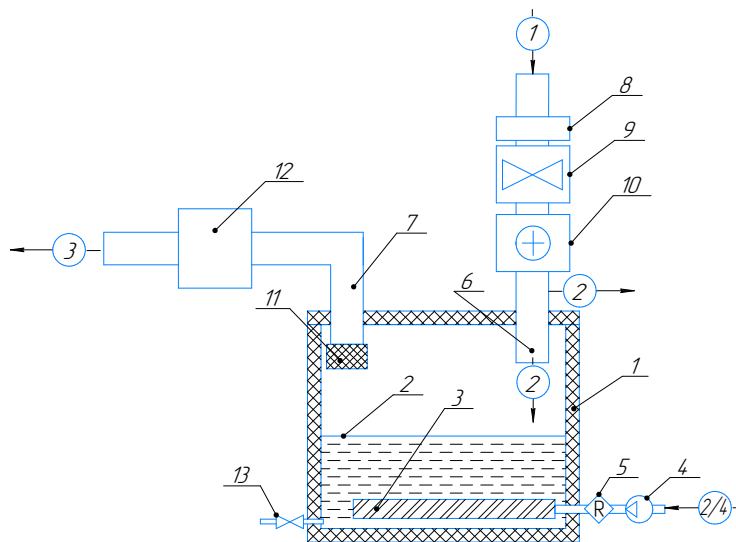


Fig. 1. Schematic diagram of the installation for studying the influence of various factors on the HOCl emission into the air. Elements: 1 – insulated container; 2 – working solution of NaOCl; 3 – panel with bubblers and heater with PID thermostat; 4 – air compressor; 5 – rotameter with tap; 6 – inlet pipe; 7 – outlet pipe; 8 – HEPA filter; 9 – fan; 10 – air heater; 11 – drip-eliminator filter; 12 – intermediate chamber; 13 – tap for sampling working solution. Flows: 1 – incoming air; 2 – HEPA-filtered and heated incoming air; 3 – air enriched with HOCl; 4 – mixture of flow 2 and CO_2 from the cylinder (in specific experiments)

The operating principle of the designed evaporator is as follows. Air, pumped by a compressor, is fed to the bottom of a container with a NaOCl working solution, and is dispersed by bubblers. As the bubble rises from the bubbler to the surface of the working solution, HOCl evaporates inside it. Upon reaching the surface, the bubble bursts, releasing the accumulated HOCl and increasing the phase boundary. At the same time, air pumped by a fan through the inlet pipe mixes with HOCl-enriched air and is discharged through the outlet pipe into the intermediate chamber. In this experiment, the chamber is necessary for a more accurate determination of HOCl in the air leaving the pipe after it is completely filled and a certain equilibrium concentration is established. The camera also prevents the HOCl-enriched air to enter the laboratory and being sucked into the installation with incoming flows. In addition, in further research, the chamber can serve for toxicological and microbiological research.

5.2. Influence of pH of NaOCl working solution, its concentration and technological parameters of the installation on HOCl emission into the air

To study the emission of HOCl from low-concentration NaOCl solutions, SEKOBREN with a concentration of 1020 mg/l and an initial pH of 9.52 was used in the first stage. The sample of 5 l of such a solution was poured into the container of the device, so that the height of the liquid column, equal to the path of the air bubble from the bubbler to the surface, was 11.0 cm. The temperature of the working solution and both air flows was maintained at 20 °C. The air velocity pumped by the fan was 0.48–0.52 m/s, which corresponds to a volumetric flow rate of about 55 m³/h. The compressor pumped air through the bubblers with a volumetric flow rate of 250 l/h. The CO₂ content in the air was 550–600 ppm. Immediately after starting the unit, the pH of the working solution began to decrease: after 30 minutes it was 9.00, after 60 minutes it reached 8.84, and after 120 minutes – 8.70. Over the next two hours, the pH decreased to 8.56 and then remained practically unchanged. Obviously, this is due to the absorption of CO₂ from the air. When the concentration of CO₂ in the bubbled air increases to 2000 ppm, this pH level can be reached after 20 minutes, and with continued bubbled air, even lower values can be achieved. However, when the supply of CO₂ through the compressor is stopped, the pH of the solution begins to increase and eventually reaches 8.50–8.55, even when the unit is not operating.

Thus, it was impossible to maintain the acidity of the solution at the initial level of about 9.50 without additional buffering. Therefore, the experiment was repeated using the same initial solution, but quickly saturated with CO₂ to a stable pH of 8.55. The free chlorine concentration the start of the experiment practically did not change and was 1010 mg/l. All other parameters remained unchanged. With continuous operation of the installation under such conditions for three hours, the concentration of total chlorine in the air of the outlet pipe was 0.30±0.07 mg/m³ (10 air samples were taken), and the pH of the working solution at the end of the experiment reached 8.60. The same air HOCl concentration was established in the intermediate chamber. The loss of mass of the working solution during this period was 150 g, and its concentration at the end of the experiment was 1026 mg/l. Thus, the calculated difference between the increase in the free chlorine concentration due to the removal of water (by 30 mg/l) and its decrease due to the removal of HOCl (by 9 mg/l)

is 21 mg/l. This practically coincides with the experimentally determined difference between the initial (1010 mg/l) and final (1026 mg/l) concentrations of the working solution, which is 16 mg/l. This means that in this exposure the material balance is carried out satisfactorily. The discrepancy is associated with the decomposition of free chlorine and, probably, with the error of the photometric determination of various forms of active chlorine at such low concentrations in the air and high in the solution. The experiment was repeated three times under the same conditions, and the deviations of all the listed parameters did not exceed 10 %.

The main studies on the influence of technological parameters of the installation were conducted using the above-mentioned CO₂-saturated working NaOCl solution.

To study the effect of the working solution column height on the HOCl (g) emission, 1 l, 2 l, 3 l, 4 l, 5 l and 5.5 l of the working solution were sequentially poured into the container. The height of its column was in the range from 1.4 to 12.4 cm. Under conditions similar to those described above, it was found that at the minimum height of the column, the C_{TC} was 0.12±0.02 mg/m³. But starting from a solution volume of 2 l (column height of 4.1 cm) it reached a value of 0.30 mg/m³ and did not change further. Thus, it can be concluded that the C_{TC} of approximately 0.30 mg/m³ here corresponds to the maximum saturation of the air bubble with its constant lifetime and size.

On the next stage, the effect of the volumetric flow rate of the air bubbled by the compressor on the process of HOCl (g) emission was studied. The sample of 5 l of the working solution were investigated under the same conditions with a compressor volumetric flow rate of 125 l/h (half of that described above) and without bubbling. In the first case, the C_{TC} was 0.27±0.05 mg/m³, and in the second – 0.12±0.03 mg/m³. Additional experiments showed that changing the number of bubblers, as well as replacing them with those with a different porosity, at the same volumetric air flow rate did not lead to a change in C_{TC} by more than 10 %.

Separately, the above experiment was carried out at a fixed volumetric velocity of the bubbling air at the level of 250 l/h, and at different volumetric flow rates of air supplied through the inlet pipe: 10 m³/h, 25 m³/h, 50 m³/h and 110 m³/h. The total chlorine concentration in the air at the outlet in all cases was almost the same and was about 0.30 mg/m³.

To study the effect of air flow temperatures, the flow rates of the air flows, which are bubbled and pumped by the fan, were maintained at 250 l/h and 55 m³/h, respectively. At the same time, their temperatures were changed, which were 20 °C, 25 °C, 30 °C and 40 °C separately. It turned out that this parameter does not have a significant effect on the C_{TC} , which remained close to 0.3 mg/m³. At the same time, increasing the temperature of the working solution has a more significant effect on the HOCl emission. In the same experiment, at a constant air flow temperature of 20 °C, increasing the temperature of the solution from 20 °C to 25 °C leads to an increase in C_{TC} by 20 % (to 0.37±0.06 mg/m³), and when heated to 30 °C – by 36 % (to 0.41±0.09 mg/m³).

The CO₂ content in the incoming air flows also affects the HOCl air concentration. As already noted, increasing it in the bubbling air allows to quickly lower the pH of the working solution and fix it at a level of approximately 8.50–8.60. However, despite the constancy of the acidity of the solution, during the operation of the unit for three hours, a change in the CO₂ content in the air pumped by the fan leads to a change in the C_{TC} . Thus, an increase in the CO₂ content

from 600 ppm to 800 ppm reliably leads to an increase in the HOCl concentration by 10–12 % (on average to 0.34 mg/m³). At a concentration of 1200 ppm this increase is about 33 % (C_{TC} reaches 0.40 mg/m³).

Given the above-described dependencies, it was interesting to investigate how the parameters of the working solution and the air being treated would change during continuous operation of the installation. For this purpose, 5 l (5038 g) of CO₂-saturated 1020 mg/l NaOCl solution with pH 8.55 at 20 °C were poured into the evaporator, continuously bubbling 250 l/h and pumping 50 m³/h of air with a temperature of 20 °C and a CO₂ content of 550–650 ppm. The experiment lasted 48 hours. The loss of the working solution was 50–60 g/h, and at the end of the experiment its mass was 2450 g. Throughout the entire time, a gradual increase in the pH of the working solution was observed: after 24 hours it was 8.85, and after 48 hours – 9.07. This is associated with the removal of the weak acid HOCl and the concentration of the solution, which entails an increase in the concentration of alkaline NaHCO₃ and NaOCl. At the same time, unexpectedly, the total chlorine concentration in the air almost did not change for 36 hours and was, as before, about 0.30 mg/m³. Only after that it began to show a slight tendency to decrease, reaching 0.20–0.24 mg/m³ after two days. At the end of the experiment, the working solution concentration was 1240 mg/l. At the same time, the increase in free chlorine concentration due to water evaporation should have been on average about 1070 mg/l, and its decrease due to HOCl removal – 144 mg/l. I.e. the resulting final concentration of the solution should have been about 1950 mg/l, which is significantly higher than the one actually obtained. This indicates that when the installation is operated under the specified conditions, not only is free chlorine emitted into the air, but also its intensive decomposition in the solution occurs.

To study the HOCl emission from a NaOCl solution with an initial pH of 7.5, 5 l of a working CO₂-saturated solution of 1020 mg/l NaOCl were used. Its pH was adjusted to 7.52 with a minimum amount of HCl. The process conditions were maintained the same as in the corresponding main experiment at pH 9.52. The experiment was carried out for 3 hours. The first air samples showed an air total chlorine concentration of 1.38±0.17 mg/m³. During further experiment, an increase in pH was observed, more pronounced than when using a solution with pH 8.55. After 90 minutes of operation, the acidity of the solution reached 7.75, which immediately affected the HOCl emission into the air, the concentration of which decreased by approximately 14 %. Changing the air flow rate pumped by the fan, as in the previous case, did not lead to a change in C_{TC} . At the same time, its temperature has a noticeable effect: when it increases to 40 °C, the HOCl concentration in the air increases by approximately 12 %. The effect of the temperature of the working solution is also pronounced: at 30 °C, the total chlorine concentration in the air is 31 % higher than at 20 °C. In the absence of bubbling, the emission of chlorine into the air decreased by approximately 3.2 times. Interestingly, the change in the concentration of CO₂ in this case had no effect on the HOCl emission. Another difference from the solution with pH 8.55 is the greater influence of the height of the column of the working solution. The C_{TC} reaches its limit value of about 1.4 mg/m³ only starting from a solution volume of 4 l (column height 8.8 cm). At the same time, when using 1 l of the working solution (column height 1.4 cm), C_{TC} was only 0.18±0.02 mg/m³.

When acidifying the 1020 mg/l NaOCl solution to pH 5.50 under operating parameters similar to those described above, a much faster increase in its pH was observed than in the previous cases. After 20 minutes it reached 6.04, and after 30 minutes – 6.35. The concentration of total chlorine in the air, measured in the pH range 5.50–5.70, is approximately 2.3 mg/m³. Further accurate analysis of such a solution under organized conditions was impossible.

Data on the influence of various factors on the total chlorine concentration in the treated air for working NaOCl solutions with pH 8.55 and 7.52 are summarized in Table 1.

Table 1

Generalized data on the influence of pH of the working NaOCl solution and process parameters on the HOCl emission into the air

Variable technological parameter		Total chlorine concentration in air (C_{TC}), mg/m ³	
		Working NaOCl solution with pH 8.55	Working NaOCl solution with pH 7.52
Temperature of the working solution, °C	20	0.30±0.07	1.38±0.17
	25	0.37±0.06	1.61±0.15
	30	0.41±0.09	1.81±0.14
Temperature of the air flows, °C	20	0.30±0.07	1.38±0.17
	30	0.32±0.08	1.50±0.12
	40	0.33±0.06	1.57±0.15
Working solution column height, cm	1.4	0.12±0.02	0.25±0.10
	4.1	0.28±0.04	0.68±0.12
	6.5	0.30±0.04	1.11±0.14
	8.8	0.31±0.3	1.38±0.17
Flow rate of air forced by the fan, m ³ /h	10	0.33±0.07	1.41±0.12
	25	0.30±0.07	1.38±0.17
	50	0.32±0.09	1.42±0.15
	110	0.33±0.10	1.43±0.13
Flow rate of bubbling air, l/h	no bubbling	0.12±0.03	0.42±0.12
	125	0.27±0.05	1.18±0.15
	250	0.30±0.07	1.38±0.17
Content of CO ₂ in the air, ppm	600	0.30±0.07	1.38±0.17
	800	0.34±0.06	1.36±0.14
	1200	0.40±0.07	1.39±0.12

To study the effect of the free chlorine concentration in the working solution on the total chlorine concentration in the treated air, a series of NaOCl solutions of different concentrations were prepared at the same acidity level. The working volume of the working solution was 5 l, the temperature of the solution and both air flows was 20 °C, the volumetric flow rates of the air pumped by the compressor and fan were 250 l/h and 55 m³/h, respectively, with a CO₂ content in the air of 600–650 ppm.

The results obtained for the diluted solutions of the CO₂-saturated SEKOBREN with different free chlorine concentrations at pH levels of 8.55 and 7.50 are presented in Table 2.

The accuracy of the photometric method did not allow reliable determination of the emission from a 128 mg/l NaOCl solution at pH 8.55. Overall, the data in Table 2 indicate a consistent decrease in the total chlorine concentration in the air with a decrease in the concentration of the working solution.

To study the HOCl release from more concentrated solutions, a NaOCl solution of 10,280 mg/l was prepared by adding the calculated amount of 20% “chemical” NaOCl concentrate to SEKOBREN. The resulting solution turned out to be, accordingly, 10 times more concentrated than the basic SEKOBREN. The pH level of such a solution was 12.45. When it was treated with air in the evaporator, a gradual decrease in pH was also observed, caused by the absorption of CO₂. After 2 hours of operation, its value reached 9.65, and the stable level was about 9.25, which is significantly higher than for SEKOBREN. Obviously, this is due to the higher concentration of free alkali and, accordingly, the formed sodium bicarbonate. The concentration of total chlorine in the air was 0.91±0.11 mg/m³. By the time the pH stabilized, there was a noticeable decrease in the concentration of free chlorine in the solution to 8800 mg/l, which indicates its low stability under such conditions. By increasing the CO₂ concentration in the bubbling air to 2,500–3,000 ppm, the pH of the solution, as in the case of SECOBREN, can be reduced further. But it also shows a tendency to rapidly increase both during operation of the unit and during storage in it due to CO₂ desorption. When a NaOCl solution with a concentration of 10,280 mg/l was acidified with HCl to pH 8.55, the C_{TC} was 2.65±0.22 mg/m³.

Data on HOCl emission from NaOCl solutions of various concentrations at pH 8.55, obtained by acidification and dilution of a rapidly CO₂-saturated 10280 mg/l NaOCl solution, are given in Table 3.

The data, as in the case of SEKOBREN, clearly indicate that a decrease in the free chlorine concentration in such working solutions causes a strictly proportional decrease in the concentration of total chlorine in the air treated by them.

located outside the housing and can be installed at a distance from it, even outside the room. This protects it from the effects of an aggressive environment and will potentially reduce the indoor noise level during operation. The installation allows not only to saturate the air with HOCl, but also to effectively absorb particles suspended in it, including those of microbiological origin, due to their contact with the foamed surface of the solution. The presence of inlet and outlet pipes allows such a unit to be used both for processing indoor air in recirculation mode and for supplying air to the premises from the outside, performing the function of supply ventilation. The evaporator can also be built into the exhaust ventilation system, which is important, for example, in infectious departments of hospitals to prevent contaminated air from entering the outside. Also, such a unit theoretically allows combining the processes of direct synthesis of HOCl (by membraneless electrolysis) and its simultaneous introduction into the air. In this case, it is possible to avoid the constant addition of fresh NaOCl solution, using the NaCl solution remaining in the container to recreate the required concentration of free chlorine by electrolysis. Finally, the low resistance to air flow, simplicity of design, low cost of components and low energy consumption of such units make them economically attractive. Thus, the use of evaporators of the specified design can fully ensure the effective use of HOCl (g) for disinfection of premises.

The results of the studies showed that the most significant contribution to the intensity of HOCl transfer into the air from the working solution is made by the acidity of the latter. As seen from Fig. 1, the target HOCl, which is the most powerful antimicrobial agent, prevails in the pH range of 2.5–7.5. However, at pH below 5.0, Cl₂ is present in the solution and, accordingly,

in the saturated vapor above it, which makes it impossible to treat the air with such a mixture in the presence of people due to its higher toxicity, irritant effect and pungent odor. In the pH range of 5.0–9.5, the only volatile component of the solution is HOCl. Its concentration decreases with increasing pH, which will lead to a decrease in the required disinfection effect. At pH>9.5, hypochlorous acid is practically absent in the solution. Thus, maintaining the pH of the solution in the evaporator within the limits of not lower than 5.0 and not higher than 9.5 is necessary both from the point of view of disinfection efficiency and to ensure the safety of the treated air. The acidity level of

the working solution during the operation of the unit depends on many factors: the initial pH level, which is determined by the presence of free alkali, the degree of hydrolysis of NaOCl and the absorption of CO₂ from the air. It is also necessary to take into account the intensity of HOCl removal, concentration of the solution due to water evaporation and the decomposition of free chlorine. That is why special attention is paid in the work to the study of working solutions with different pH in the developed device.

One of the key factors in the change in the pH of working solutions during evaporator operation is the absorption of CO₂ from the air. This is especially noticeable when using basic SEKOBREN with a pH of 9.5. In this case, the absorption of

Table 2
Emission of HOCl into air from electrochemically generated NaOCl solutions with different free chlorine concentrations

Concentration of free chlorine in the working solution, mg/l	Total chlorine concentration in air (C _{TC}), mg/m ³	
	For working solution with pH 8.55	For working solution with pH 7.50
1020	0.30±0.08	1.38±0.15
510	0.22±0.05	0.75±0.10
255	0.14±0.05	0.39±0.12
128	was not measured	0.22±0.05

Table 3
Emission of HOCl into the air from highly concentrated NaOCl solutions with different free chlorine content

Concentration of free chlorine in the working solution, mg/l	10,280	7,100	5,140	2,570	2,040	1,285
Total chlorine concentration in treated air, mg/m ³	2.65±0.22	1.80±0.17	1.35±0.16	0.73±0.13	0.56±0.11	0.38±0.09

6. Discussion of the results of the study of the HOCl (g) emission into the air in the developed evaporator under different operating conditions

The developed prototype is a type of forced-air evaporator in which foaming of the working solution is used to increase the interfacial surface area. Such a device takes into account the specific properties of chlorine-active solutions. Thus, only a chemically and corrosion-resistant container, inlet and outlet pipes, bubblers and a heater (which is not mandatory) are in direct contact with the working solution. All auxiliary equipment (compressor, fan, if necessary, air filter and heater) is

CO₂ leads to the neutralization of free alkali in the solution with the formation of sodium bicarbonate NaHCO₃ and, accordingly, to a decrease in pH. The amount of absorbed CO₂ primarily depends on the amount of free alkali in the initial solution. At the same time, taking into account the electrochemical origin of the latter, this amount is small (up to 10⁻³ mol/l), and after saturation the solution has a weakly alkaline reaction. However, even at such a pH (about 8.50), the presence of dissolved CO₂ in it and, accordingly, its further absorption from the air is practically impossible [31]. The relaxation of pH to 8.50 after its short-term decrease during bubbling of air with a high concentration of CO₂ is also explained by its gradual desorption from weakly alkaline media. At the same time, in the pH range of 8.50–8.60 in a CO₂-saturated working solution, the ratio between the equilibrium concentrations of HOCl and NaOCl is close to 1:9 (Fig. 1). As a result, a hypochlorite buffer system arises, which gives the solution a certain buffer capacity. This prevents a change in acidity during further interaction with CO₂ of the air supplied through the inlet pipe, which is possible in thin films of unstable surface foam. A more pronounced gradual increase in the pH of the working solution with an initial pH of 7.50 during the operation of the unit is due to a more intensive removal of the acidic component HOCl. For solutions with an initial pH of 5.50, this trend is even more noticeable, again, due to a more intensive removal of HOCl. On the other hand, this is also due to the absence of buffer properties in such a solution, caused by the absence of a strong electrolyte with the ClO⁻ ion. In addition, at such an acidic pH, CO₂ absorption does not lead to NaHCO₃, but to sodium carbonate Na₂CO₃, the solutions of which are more alkaline. Interestingly, in the case of a working solution with a pH of about 8.60, an increase in C_{TC} is observed with an increase in the CO₂ concentration in the air even with a constant acidity of the solution itself. Probably, when its foamed surface comes into contact with CO₂-containing air supplied by a fan, carbon dioxide is rapidly absorbed by the thin wall of the foam bubbles. This leads to a decrease in pH in it, despite the buffer capacity of the solution. Accordingly, a local increase in the concentration of HOCl appears, which is immediately released into the air. For a solution with a pH of 7.50, this process is prevented by its higher buffer capacity.

Comparing the C_{TC} values for solutions with pH 8.55 and 7.52 at 20 °C (Table 1), it is possible to conclude that they are strictly proportional to the HOCl concentration in the solution, which can be determined from Fig. 1. Thus, at pH 8.55 the HOCl content is about 11 %, and at pH 7.52 – about 50 %, i.e. 4.5 times more. And, as follows from Table 1, it has been experimentally confirmed that C_{TC} at pH 7.52 exceeds that for pH 8.55 by 4.6 times, i.e. exactly the same number of times (taking into account the photometric error). Thus, by acidifying the working solution, it is possible to increase the C_{TC} quite accurately. However, a significant disadvantage of acidifying the working solution is a decrease in its stability. Thus, the concentration of free chlorine after 6 hours of operation at pH 7.55 decreases by 80 mg/l. Further storage of the solution in a container with the fan and compressor turned off results in a further drop of 130 mg/l over the next 10 hours. A more rapid decomposition of free chlorine in media close to neutral is a characteristic feature of NaOCl solutions [32]. In addition, it is necessary to take into account the tendency for the pH of acidic solutions to increase during operation of the installation. This fact does not allow maintaining a constant C_{TC} value for a long time, contributing to its gradual decrease.

Interesting results were obtained during continuous use of the device for 2 days. The first 36 hours the C_{TC} remained

practically unchanged, despite the gradual increase in the pH of the working solution as it evaporated. At the same time, according to the diagram (Fig. 1), already at pH 9.0 the HOCl concentration in the solution is almost half that at pH 8.55, which should have led to its corresponding decrease in the air. The absence of such differences is probably explained by a complex of processes occurring in the unstable foam on the surface of the solution, and not in its volume. First of all, this is the already mentioned absorption of CO₂ by thin films. Also, intensive evaporation of water from the foam wall leads to the concentration of electrolytes in it, which, according to Sechenov's law, leads to the salting out of volatile components, in this case HOCl. In addition, an increase in the free chlorine concentration in the solution due to the predominance of water evaporation over HOCl emission plays a certain role.

As follows from Table 1, the HOCl (g) emission from the evaporator practically does not depend on the power of the air flow processed in it, at least in the ranges described in the study. This is explained by the fact that the evaporation process occurs under non-equilibrium conditions, and with an increase in the air flow rate, the intensity of evaporation of volatile components (water and HOCl) from the foamed surface of the working solution increases proportionally. And since the ratio of equilibrium concentrations of these compounds in the solution at a constant pH does not change, the ratio of partial pressures of their saturated vapors does not change either. So, the C_{TC} remains constant regardless of the air flow rate. In practice, this will allow the system to be adjusted in accordance with the requirements for air exchange for a specific room and maintain the effective concentration of the antimicrobial agent in the air without significant changes in the design of the installation and the volume of the container, but only by adjusting the flow rate of the treated air, which is impossible for evaporators of a other design.

The use of bubbling, as expected, allows to significantly increase the HOCl emission into the air. It is obvious that an important characteristic of the bubbling process is the number and size of air bubbles, which is determined by the properties of the bubblers. In addition, the composition of the solution also affects the distribution and life time of air bubbles. Thus, it is even visually noticeable that the size of the bubbles in a solution with pH 7.50 is smaller than at pH 8.50, which can be explained by the higher content of the volatile component in their wall. This probably explains the differences in the influence of the column's height of working solutions of different acidity on achieving the maximum C_{TC} . In this work, the quantitative characteristics of the resulting gas emulsion were not determined. This technological aspect requires further study.

The effect of temperature on the HOCl (g) release process is predictable. It increases with increasing temperature of both the air stream (which is partially compensated by the endothermic effect of evaporation in the foam) and the working solution due to an increase in the evaporation rate and a decrease in the solubility of HOCl. In this case, the temperature of the working solution has a much greater effect, increasing the C_{TC} by approximately 30 % with an increase of 10 °C. However, at the same time, the stability of NaOCl solutions decreases upon heating [32].

In addition to adjusting the pH of the working solution, it is possible to increase the HOCl emission into the air by increasing the total concentration of free chlorine in the solution at a fixed pH. NaOCl solutions are available in a wide range of concentrations, which primarily depends on the

method of their production. Solutions obtained by the “chemical” method, i.e. by dissolving Cl_2 in a concentrated sodium hydroxide solution, have a high concentration, up to 25 %. However, such solutions also contain a large excess of free alkali. Solutions obtained by electrochemical methods, i.e. by electrolysis of NaCl solutions, can reach a concentration of 20 % (when using membrane electrolyzers) or up to 1 % (when using membraneless flow systems with rare-earth metal electrodes). The pH level of electrochemically generated solutions is usually lower than that of “chemical” ones, and it is they that are mainly used for medical purposes.

The data in Table 2 demonstrate that for diluted SEKO-BREN solutions with pH 8.55 and 7.50, the experimentally obtained C_{TC} was higher than that calculated by dilution, and for the solution with pH 8.55 this was much more pronounced. Thus, when the initial solution of 1020 mg/l NaOCl is diluted 2 times, one could expect C_{TC} values of 0.15 mg/m³ and 0.7 mg/m³ for solutions with pH 8.55 and 7.50, respectively. However, the found C_{TC} exceed the calculated ones by 0.07 mg/l (approximately 50 %) and 0.06 mg/l (approximately 10 %) for solutions with pH 8.55 and 7.50, respectively. This is probably due to a decrease in the buffer capacity of the working solutions upon dilution and a more intense absorption of CO_2 by the foam film. For further dilutions, especially in the case of a solution with pH 7.50, the decrease in C_{TC} is proportional to the decrease in the free chlorine concentration in the solution (taking into account the photometric error).

A similar situation is observed for highly concentrated NaOCl solutions (Table 3). Thus, when a 10280 mg/l NaOCl solution was acidified with HCl to pH 8.57, the C_{TC} was 2.65±0.22 mg/m³. This is approximately 12 % less than would be expected based on extrapolation of data obtained for SEKO-BREN with a concentration 10 times lower, for which approximately 0.3 mg/m³ of total chlorine in air was determined. Such differences are probably due to the different salt composition and different buffer capacity of these two solutions. The data in Table 3, as in the case of SEKO-BREN, clearly indicate that a decrease in the free chlorine concentration in concentrated working solutions causes a strictly proportional decrease in the total chlorine concentration in the air treated by them. As seen, for 2040 mg/l NaOCl solution, the obtained C_{TC} value is 2 times higher than for 1020 mg/l SEKO-BREN, and further extrapolation to a concentration of 1020 mg/l leads to C_{TC} values of about 0.3 mg/m³, which correlates with the indicators for the base SEKO-BREN. When acidifying a solution of 2570 mg/l NaOCl to pH 7.54, the obtained C_{TC} value was 3.77±0.34 mg/m³, which is 5 times higher than at pH 8.55 at the same free chlorine concentration. This is also in accordance with Fig. 1, according to which the concentration of volatile HOCl with such a change in pH also increases by about 5 times. It can be concluded that at the same pH and approximately the same salt content of working NaOCl solutions, the amount of HOCl emitted into the air is strictly proportional to the free chlorine concentration in them. The importance of our research in this context lies in establishing certain “reference” C_{TC} s when using a device of this design. This allows to predict and regulate the content of the antimicrobial component in the air quite accurately when changing the parameters of the working solution. A significant disadvantage of using concentrated solutions is their high decomposition rate and unstable, constantly increasing pH level, which does not allow achieving a constant

HOCl concentration in the air during long-term (even for 1.5–2 hours) operation of the installation.

The set of experiments fully describes the influence of the main chemical and technological factors on the emission of HOCl (g) into the air. This favorably distinguishes the study from previously conducted works [24, 25, 30], which focus mainly on measuring the active chlorine concentration in the air without reference to the properties of the working solution and air flows.

Thus, it is shown that the release of HOCl into the air during its interaction with foamed NaOCl solutions in a special type of evaporator is determined mainly by its concentration in the working solution. The latter, in turn, depends on the free chlorine concentration in it and its pH. To ensure gradual and constant HOCl emission, it is important to use solutions that are as stable as possible in organized conditions and are able to maintain the acidity level for a long time during the operation of the installation. Under the conditions studied, these qualities are best met by an electrochemically generated NaOCl solution with a concentration of approximately 1000 mg/l at a pH of 8.50–8.60. Its use allows for two days of continuous operation to practically constantly maintain the total chlorine concentration in the air leaving the installation at a level of 0.30 mg/m³ at a volumetric flow rate of treated air of about 50 m³/h at 20 °C. This concentration is satisfactorily extrapolated to both more and less concentrated solutions provided that the pH level is maintained at the same level. According to the data given in [24], C_{TC} already at the level of 0.03 mg/m³ provides a reduction in the microbial number in the air at a rate of 13 %/min, concentration of 0.3 mg/m³ and higher, the rate of pathogen inhibition is 65 %/min depending on the type of microorganism. In addition, this solution has a number of other advantages. In particular, our previous works show that it is technologically possible to synthesize it in a accumulative electrolyzer, which can be a container of a constructed evaporator. Also, the method of electrolysis in the polarity reverse mode developed for this purpose will allow avoiding the use of electrodes made of expensive materials and preventing the formation of undesirable sodium chlorate [33, 34]. In general, the results of the study indicate broad prospects for the use of evaporators of this design with chlorine-active working solutions for complex gas-phase disinfection of premises, including in the presence of people. At the same time, this work contains a number of limitations that must be taken into account when attempting to apply its results in practice. Thus, the air was studied, which does not contain chemical (dust particles, evaporation of construction materials, ammonia, etc.) and microbiological contaminants inherent in domestic, medical and industrial premises. Such impurities can significantly affect the properties and stability of the working solution. Also, the laboratory prototype of the evaporator obviously does not allow for experiments with air flows, the power of which is close to the centralized ventilation systems of large premises. In addition, the work did not study the characteristics of the gas emulsion and foam that arise when air is bubbled throughout the working solution. At the same time, even the experiments conducted in the first approximation show that this has a significant impact on the process of HOCl (g) emission. A significant drawback of the study is the use of only a NaOCl solution with a high NaCl content as the working solution compared to industrial preparations. The latter, in addition, may contain other impurities (for example, NaOH) in significant quantities, which may make it impossible to extrapolate the results obtained in

the study to them. The second drawback of the work is the lack of statistical data processing and derivation of equations that would allow quickly calculating the C_{TC} for given properties of the working solution. This drawback can be corrected in further work using a more accurate and convenient method for determining C_{TC} using a special electrochemical sensor. A promising continuation of this area of research will be the optimization of the design of the evaporation device and its equipment with an electrolysis unit. It is also necessary to conduct a detailed study of the effectiveness of disinfection treatment of premises using this technology in different operating modes of the installation.

7. Conclusion

1. A prototype of a forced air evaporator has been developed, in which the contact surface of the treated air with the working solution is increased by foaming the latter. It has been shown that a device of this design can provide uniform emission of HOCl (g) and at the same time takes into account the specific properties of chlorine-active solutions.

2. The concentration of HOCl (g) in the air treated in the designed evaporator mainly depends on the HOCl concentration in the working solution and varies in full accordance with the known diagrams of the molar distribution of active chlorine components. Using the electrochemically generated solution of 1000 mg/l NaOCl at a pH of approximately 8.5, it is possible to ensure the content of HOCl (g) in the air at the level of 0.3 mg/m³ for a long time, almost regardless of the airflow capacity.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Data availability

Data will be made available on reasonable request.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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