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ANALYSIS OF APPLYING SURFACE WATER TREATMENT TECHNOLOGY WHEN USING CHLORINE DIOXIDE AT WATER SUPPLY PLANTS

E.O. Mavrykin, Ph.D. student
Institute of Water Problems and Land Reclamation of the National Academy of Agricultural Sciences of Ukraine, Kyiv, Ukraine; https://orcid.org/0000-0002-6193-8890; e-mail: evgeniy_mavrikin@ukr.net

Abstract. *The results of the conducted research made it possible to establish that in EU countries chlorine dioxide (CD) is more often used for secondary or final disinfection of drinking water. By-products of this process are chlorites and chlorates, which are subject to control in the drinking water of all EU countries. Aldehydes and carboxylic acids can also be formed in drinking water, which leads to a decrease in the microbiological stability of tap water. Ozonation and filtration using a carbon filter are used in the final stage of drinking water purification, which contributes to a significant reduction in the dose of CD and water contamination with toxic chlorites. In the case of pre-oxidation of water with sodium hypochlorite, the largest amount of chlorites and chlorates is formed, while in the case of using potassium permanganate for the same purpose, the need for CD and the amount of chlorites and chlorates in drinking water reduced. Chlorination of natural water that has undergone CD pre-oxidation leads to complete oxidation of the chlorites that have formed, increases the effectiveness of disinfection, and provides a bacteriostatic effect in the distribution network. During 2021–2022, when using CD for the treatment of drinking water at the Dniprovska WTP in Kyiv it was established that the process of treating natural water with CD is accompanied by the formation of its by-products, mainly toxic chlorites, the levels of which depend on the applied doses of CD and are the lowest in winter, while the largest ones are observed in summer and do not always reach regulatory values (0.2 mg/l) and range up to 0.7 mg/l, which corresponds to the WHO recommended standard for this substance in drinking water. Italian scientists focus their attention on the fact that during the first years of using CD at each water supply station, optimal conditions must be ensured for the safe and effective use of this reagent. Therefore, CD is becoming widespread in the EU countries and Ukraine for the treatment of tap drinking water; it is an alternative method of water effective disinfection at water supply stations with traditional surface water purification technology. Using such a method for treating surface water requires a preliminary pilot experiment and should be carried out along with an analysis of the feasibility of using the methods for preliminary and/or final purification of drinking water from organic substances and additional disinfection. Today, based on experimental and natural studies, it is relevant to expand knowledge about the properties of CD in the case of its use in drinking water supply for the treatment of surface water with a high content of organic substances.*

Key words: *chlorine dioxide, chlorites, oxidation by-products, water supply stations*

Relevance of research. For the population of Ukraine, as in many European countries, the problem of providing drinking water of guaranteed quality is particularly relevant [1]. Data from scientific sources indicate the unsatisfactory quality of surface water in Ukraine in general and their critical condition in certain regions. Its condition is directly related to the composition and volume of return water, in particular, the efficiency of wastewater treatment and disinfection at sewage treatment plants in settlements and industrial enterprises. Climate change also has a significant negative impact on surface water quality [2–4]. Screening monitoring of the Dnipro River basin showed extremely high levels of predicted safe concentrations in water of herbicides, insecticides, fungicides, as well as pharmaceutical substances such as carbamazepine, lopinavir, diclofenac, efavirenz, etc.

As a result of the intensive inflow of biogenic compounds into rivers and reservoirs, the latter have a massive development of phytoplankton, which complicates the processes of treating high-quality drinking water at water supply stations (WSS) [5]. In the zone of active hostilities, providing a drinking water supply to the population is a particularly difficult task. Enterprises of centralized drinking water supply bear the burden of problems due to the non-guaranteed quality of the source water, the lack of stable conditions for the technological process of water treatment as well as for the operation of water treatment facilities and networks, etc. [6, 7].

Thus, in general, in the current conditions of anthropogenic load and climate change, which negatively affect the quality of surface water, namely, lead to an increase in the concentration of the organic component of water pollution, deterioration of microbiological indicators, etc., traditional coagulants and disinfectants are not able to ensure the normative quality of drinking water [8]. Disinfection is the priority process of its treatment. Due to the low cost, ease of operation, high efficiency, and stability in the distribution network, drinking water is chlorinated (with chlorine or sodium hypochlorite), but the identified shortcomings of the chlorination process became the basis for the application of chlorine dioxide (CD) in disinfecting tap drinking water [9–11].

Scientists have carried out a wide range of research on the properties of CD, in particular, when applying it at water supply stations, the advantages of using CD for disinfection of drinking water compared to chlorine, its disadvantages, features of combined action with other disinfectants, etc. However, today disinfection of drinking water with CD is becoming widespread in the EU countries and Ukraine [12]. Scientists emphasize the relevance of researching and analyzing an appropriate applied technology for each drinking water supply station, primarily due to the large difference in the composition of the source water and the applied technological approaches at WSS [13]. Because of the above, we analyzed data from the scientific sources on the experience of using CD at WSS.

Analysis of recent research and publications. For the first time in the world, CD was used to disinfect drinking water in Germany in 1894. The use of this reagent in water supply was restrained for a long time, but after solving the problem of industrial production of sodium chlorite, which is the main component in obtaining CD, as well as after establishing the fact that in the process of chlorination carcinogenic organochlorine is formed in water, CD started to be used for decontamination of tap drinking water [14, 15]. At the end of the 20th century, this reagent was already widely used, primarily for the secondary disinfection of drinking water at water treatment plants in the USA and Western Europe, and it also began to be used in Ukraine [16–18]. Today it is used for treating municipal water supply in 13 EU countries – Austria, Finland, France, Germany, Greece, Hungary, Italy, Luxembourg, Poland, Portugal, Romania, Slovenia, and Spain [19].

The purpose of the scientific research is to analyze the features of using CD for surface water treatment based on literary sources regarding the relevant field studies conducted.

Research materials and methods. Theoretical methods of scientific research such as analysis and synthesis, comparison, classification, and generalization were used in the study.

Research results and their discussion. Based on the analytical studies, it was established that in the EU countries, CD is more often used for secondary or final disinfection of drinking water, in particular for the treatment of surface water at the stage of disinfection after preliminary ozonation and before UV disinfection. For example, in the south-east of Poland in the Subcarpathian Voivodeship, there are the following water intakes and their water treatment technologies: A – the Wisłok River (capacity – 84,000 m^3/day, drinking water treatment technology: preliminary ozonation, coagulation, filtration (anthracite-sand layer), secondary ozonation, filtration (carbon layer), Cl2 disinfection, CD, UV disinfection); B – the Yasiolka River (capacity – 7000 m^3/day, drinking water treatment technology: preliminary oxidation, coagulation, filtration (sand-gravel layer), CD disinfection, UV disinfection); C – the Besko reservoir (17,000 m^3/day, drinking water treatment technology: preliminary oxidation, coagulation, filtration (anthracite-sand layer), CD disinfection, UV disinfection) [20].

Following the literature data, by-products are formed in drinking water after CD disinfection, the main of which are chlorites (standard by Directive 2020/2184/EU – ≤ 0,7 mg/l) and chlorates (standard by Directive 2020/2184/ EU – ≤ 0,7 mg/l), which are subject to control in drinking water in all EU countries. In particular, 50–70 % and 0–10 % of applied CD are transformed into chlorite and chlorate, respectively [21]. Carboxylic acids and aldehydes – low molecular weight organic compounds with high biodegradability can also be formed in drinking water treated with CD. CD, like ozone, reacts with organic substances in surface water as a result of which aldehydes are formed. Their presence in the water supply network is extremely undesirable due to the possibility of secondary growth of microorganisms present in running water and forming biofilms on the inner walls of distribution pipes, especially when the disinfectant remains disappear at the same time [22–27].

The other researchers [13], in the city of Poznań (Poland), conducted a research for several years on the method of using CD in the centralized drinking water supply for the treatment of surface water with high reactivity of natural organic matter, which led to a decrease in the microbiological stability of water. A particular increase in this phenomenon was observed after using CD for secondary disinfection. The sourse water of the Mosina WTP, located approximately 20 km from Poznań, is a mixture of groundwater and infiltration water (40 %) of the wells drilled on the shoreline in the Warta River dam to reduce organic matter in source water.

Despite numerous attempts, it was impossible starting use only CD due to the growth of psychrophilic bacteria in the water supply system. Therefore, the water was constantly disinfected with a mixture of two agents: gaseous chlorine and CD in a ratio of 60–40 %. However, mixing disinfectants with a seasonal change in their rate was not completely effective and worsened the organoleptic parameters of the water.

Cast iron and PVC/PE are the basic materials of the pipes; there are some sections of the water supply pipeline, which are made of asbestos-cement pipes, which can be successively replaced. The age of almost half of the pipes is 10–40 years; 15 % of the network was constructed more than 40 years ago [28]. Modernization of the WTP was carried out during 2010–2015, which primarily included the construction of the facilities for the final stage of purification, which involved ozonation and sorption with activated carbon, as well as an increase in the capacity of the WWTP from 100.000 to 150.000 m^3/day, while liquid chlorine was replaced with sodium hypochlorite. Figure 1 shows which technologies are correlated with the CD rate used at the Mosina WTP.

After the two-stage treatment, the water at the Mosina WTP was characterized by a reduced content of disinfection by-products and organic substances, which led to a decrease in the need for disinfectants. In 2015, the leaching of organic compounds, which were in the water supply network before the reconstruction was observed (in 2019, it was not observed) [13].

Another article presents the results of experiments conducted at the Fortore (Foggia, Southern Italy) and Mosina (Poznan, Poland) WTPs. Chlorine was added to the CD solution used for preliminary water disinfection.

The results showed high chlorite removal efficiency on GAC filters up to 5.500 and 10.000 bed volumes for mineral and vegetable GAC at Fortore and up to 11.000 bed volumes at Mosina. Natural organic matter (NOM) dissolved in raw Fortore water was also characterized. The presence of small molecules (< 500 Da) in water that was previously disinfected led to the formation of trihalomethanes (THM) and carboxylic acids due to a rapid reaction with Cl_2-ClO_2. Low molecular weight carboxylic acids are effectively removed from water during filtration through biologically active carbon layers. GAC filters showed removal percentages from 60 to 72 % for THM and from 14.6 to 43 % for total organic carbon (TOC) [29].

CD is usually used in medium and large drinking water treatment plants due to high production and management costs. Practical experience has shown that the service life of polyethylene pipes is significantly reduced due to

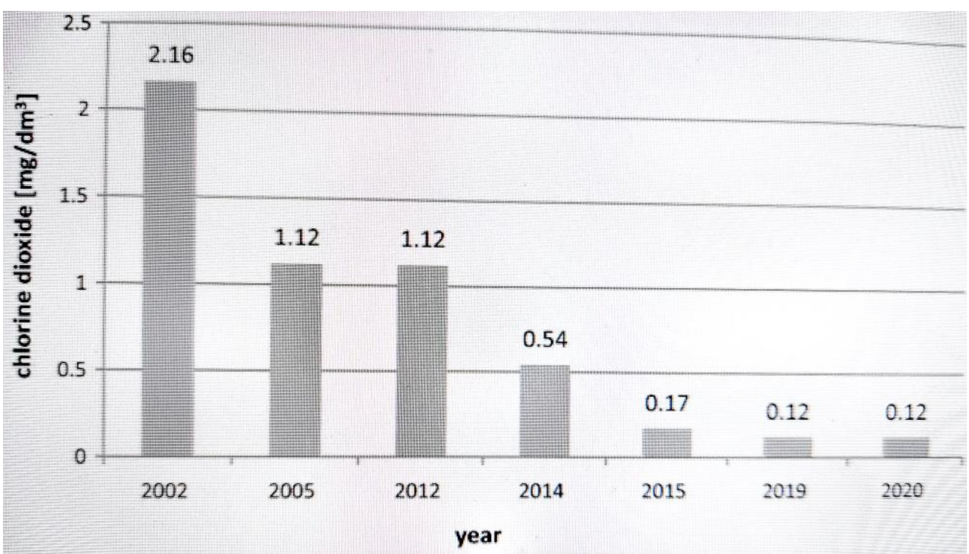

Fig. 1. Dynamics of CD rates depending on the drinking water treatment technology at the second stage: 2005 – modernization of post-filters for cleaning from iron and manganese compounds; 2012 – a new aeration plant was put into operation; 2014 – powdered coal was dosed; 2015 – the entire technological line was put into operation, including ozonation and filtration through a carbon filter (without powdered coal)

their being affected by the water with CD. The CD is more aggressive to plastic pipes, in particular polyethylene (PE) ones than other chlorine-based disinfectants. That can be explained by the fact that CD is a dissolved gas that diffuses faster into the polymer compared to other disinfectants that accelerate degradation reactions. Recently, many companies have offered new polyethylene pipes with a modified formulation, more resistant to CD. However, there is still no standardized test method for evaluating the long-term performance of polyethylene pipes. Further experiments are necessary to correlate the parameters of the chemical and mechanical characteristics of polyethylene pipes with their service life [30].

According to the research results of Italian scientists, it has been established that in the case of preliminary oxidation of water with sodium hypochlorite, the need for CD is high, and the largest amount of chlorites and chlorates is formed. At the same time, in the case of preliminary oxidation with potassium permanganate, coagulation with both iron chloride and aluminum sulfate reduces the need for CD, as well as the number of chlorites and chlorates in drinking water. Activated carbon reduces the content of CD by about 50 % and leads to a decrease in the formation of chlorite and chlorate [31].

Data from field studies on the combined use of CD and chlorine for the treatment of surface water in five cities of Ukraine (Zaporizhia, Dnipro, Sevastopol, Kremenchuk, Zhovti vody) showed that the most optimal is the use of CD for primary disinfection, while chlorine (liquid or sodium hypochlorite) is effective to be used at the stage of post-disinfection. Depending on the quality of the source water, the effective rate of CD at the pre-oxidation stage is 1.0–1.5 mg/l, which is 3–4 times less than the CD rate sufficient to achieve a similar effect. Chlorination of natural water that underwent peroxidation with CD leads to complete oxidation of formed chlorites, increases the effectiveness of disinfection, and provides a bacteriostatic effect (prolonged effect) in the distribution network [32].

By the literature sources, at the stage of industrial research (2017) on the use of CD for the treatment of drinking water at the Dniprovska WTP in Kyiv city, for primary disinfection a rate of 1.2–1.5 mg/l was used [15], for the secondary disinfection – 0.3–0.45 mg/l, and after its introduction into the technological process (2021-2022) [33] – 0.8–2.5 mg/l, and 0.2–0.6 mg/l respectively. Before entering clean water tank (CWT), the water undergoes the stages of treatment with CD, coagulation (with aluminum sulfate and iron chloride), settling, filtering, and disinfection with CD. The process of treating natural water with CD is accompanied by the formation of its by-products, mainly toxic chlorites, the concentrations of which depend on the applied rates of CD and are the lowest in winter and the largest in summer. In the summer period, the maximum concentration of chlorites in drinking water from CWT can be higher than the national hygienic standard (0.2 mg/l) and range up to 0.7 mg/l, which corresponds to the WHO recommended standard for this substance in drinking water. Based on the hygienic assessment of individual options for using CD in technology of drinking water preparation from surface sources compared to traditional chlorine technology, the advantages of using CD in water treatment instead of ordinary chlorine at the initial and final stages of surface water treatment are shown.

Italian scientists emphasize that in the case of applying CD, the staff of each WTP accumulate their own experience for the implementation of the optimal technological process. During the first years of applying CD, the personel at WTP should elaborate on their optimal conditions for the safe and effective use of this reagent. Correctly conducted pilot studies are a mandatory element of planning modernization changes in water supply facilities [13].

Conclusions. The CD is becoming widespread both in the EU countries and in Ukraine for the treatment of tap drinking water. It is an alternative method of an effective disinfection for water treatment plants with traditional surface water purification technology. It was established that the use of CD for surface water treatment requires a preliminary pilot experiment and should be carried out together with an analysis of the feasibility of using methods for preliminary and/or final purification of drinking water from organic substances and additional its disinfection. Thus, today it is relevant, based on experimental and natural studies, to expand knowledge about the properties of CD in the case of its use for the treatment of surface water with a high content of organic substances in the field of drinking water supply.

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АНАЛІЗ ЗАСТОСУВАННЯ ТЕХНОЛОГІЇ ОБРОБКИ ПОВЕРХНЕВОЇ ВОДИ ДІОКСИДОМ ХЛОРУ НА ВОДОПРОВІДНИХ СТАНЦІЯХ

Є. О. Маврикін, аспірант

Інститут водних проблем і меліорації НААНУ, Київ, Україна
https://orcid.org/0000-0002-6193-8890; e-mail: evgeniy_mavrikin@ukr.net

Анотація. *Результати проведених досліджень дозволили встановити, що в країнах ЄС діоксид хлору (ДХ) частіше використовують для вторинного або заключного знезараження питної води. Побічними продуктами такого процесу є хлорити та хлорати, що підлягають контролю у питній воді всіх країн ЄС, а також у питній воді можуть утворюватися альдегіди і карбонові кислоти, що призводить до зниження мікробіологічної стабільності водопровідної води. Через зазначене на заключному етапі очищення питної води використовують озонування та фільтрацію через вугільний фільтр, що сприяє суттєвому зменшенню дози ДХ та забрудненню води токсичними хлоритами. У разі попереднього окиснення води гіпохлоритом натрію утворюється найбільша кількість хлоритів та хлоратів, а у разі використання з цією ж метою перманганату калію знижується потреба у ДХ та кількість хлоритів та хлоратів у питній воді. Хлорування природної води, що пройшла преокиснення ДХ, призводить до повного окиснення хлоритів, які утворилися, підвищує ефективність знезараження та забезпечує бактеріостатичний ефект у розподільній мережі. Протягом 2021–2022 рр. використання ДХ для обробки питної води на Дніпровській ВС м. Києва встановлено, що процес обробки природної води ДХ супроводжується утворенням у ній його побічних продуктів, переважно токсичних хлоритів, рівні яких залежать від застосовуваних доз ДХ і є найменшими взимку, а найбільшими – влітку та не завжди досягають нормативних значень (0,2 мг/л) і коливаються у межах до 0,7 мг/л, що відповідає рекомендованому ВООЗ нормативу для цієї речовини у питній воді. Італійські науковці акцентують свою увагу на тому, що протягом перших років використання ДХ на кожній водопровідній станції повинні забезпечуватись свої оптимальні умови для безпечного та ефективного використання цього реагенту. Отже, ДХ набуває поширення в країнах ЄС та Україні для обробки водопровідної питної води, є альтернативним методом її ефективного знезараження на водопровідних станціях із традиційною технологією очищення поверхневої води. Застосування такого методу для обробки поверхневої води потребує попереднього пілотного експерименту та повинно здійснюватися разом з аналізом доцільності застосування методів для попереднього та/або заключного очищення питної води від органічних речовин та додаткового її знезараження. На сьогодні є актуальним на підставі експериментальних та натурних досліджень розширити знання щодо властивостей ДХ у разі його використання в практиці питного водопостачання для обробки поверхневої води з високим вмістом органічних речовин.*

Ключові слова: *діоксид хлору, хлорити, побічні продукти окиснення, водопровідні станції*