Treatment Features of High-Color Natural Waters

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Abstract. Based on studies conducted at the water treatment complexes in the Primorsky Territory, as well as by taking into account the global practice of water treatment, an analysis is undertaken on the dangerous consequences of preliminary chlorination during the bleaching of natural waters, as well as on the effect of chlorine-containing components on organic impurities contained in untreated water. The development features of the microbiome in wastewater treatment plants of unpredictable - including hazardous - species are also considered.

1. Introduction
The issue of clean water is the most important problem facing mankind and this is primarily due to the fact that the WHO defines water as the most important food product - largely on account of it being essential for our health. In this regard, it is important to consider all risk factors that, to one degree or another, can affect the quality of this product. It is quite commonplace to consider that the most dangerous pollutant of purified water is toxic substances. When such dangerous impurities are found in natural water - in the source of water supply - it is impossible to ignore this and, in accordance with existing requirements, the water treatment technology must deal with the need for their complete elimination from water. However, when substantiating technological solutions, issues associated with the transformation of the various chemicals used in water treatment are fully considered. The toxic substances and bacteriological products formed during water purification can often turn out to be no less dangerous than the well-known natural and anthropogenic pollutants of water sources. In addition, in recent years, new, not yet well-studied pollutants have been found that relate to organic substances (dispersed organic matter - DOM), including those of anthropogenic origin. Moreover, it is currently not possible to determine the degree of danger that they pose. In the modern practice of water treatment, when considering issues of transformation and the formation of carcinogenic substances, the possibility of dangerous microbiological contaminating purified water during the development of various types of microorganisms is not taken into account. The development of a microbiome of diverse quantitative and qualitative composition always occurs in an aqueous medium that has any nutritional value, in particular, the presence of DOM. The suppression of metabolic activity even with the preliminary chlorination of still untreated water is often not occur. Under such conditions, as the studies have proved, it is increasingly necessary to state that drinking water does not always meet the requirements that should define it as a food product.
The aim of this experimental work is to study the main technological factors that determine the negative changes in the quality of the treated water containing DOM in relation to toxic substances and bacteriological products, which will justify the strategy for improving pre-treatment technologies.

2. Analysis of the negative effects of prechlorination

In the practice of water treatment, as shown by the analysis of publications [1, 3, 4, 5], prechlorination (prechlorination) - which is considered by many authors to be especially effective in treating colored water - is in widespread use. In particular, the effect of chlorine-containing substances as the strongest oxidizing agents is noted insofar as they should contribute to the oxidation of dissolved organic substances, and therefore the improvement of coagulation conditions. It is also believed that by treating water with chlorine, some of the substances responsible for the color of the water are oxidized, and colloids of DOM are destroyed, which impedes the coagulation process. Along with a decrease in color, water disinfection occurs, which significantly improves the sanitary condition of the treatment plants. It was found that depending on the prechlorination of the source water, which leads to the composition of humic, it is possible to reduce its color by 25-60%, and this significantly affects the consumption of the coagulant [2, 3, 10].

In accordance with the guidelines for the implementation of SanPiN requirements 2.1.4.10741-01, the regulatory conditions for prechlorination must be observed at water treatment plants. The residual chlorine content should be at least 0.5 mg / l, with a contact time of water with chlorine of 30 minutes. It is known that microorganisms and viruses that are sufficiently resistant to the effects of chlorine reagents, in particular, hepatitis virus, Giardia cysts, etc. [1, 2, 17].

The main processes involved in the reagent treatment of natural waters for the purposes of domestic, drinking, and industrial water supply are always preceded by a stage of preliminary water treatment. At this stage, in accordance with the adopted technology, the initial water is mixed with various reagents - chemicals which should ensure the coagulation of water impurities and optimize the conditions for their removal from water. For this purpose, preliminary chlorination (ozonation or ultraviolet irradiation) and alkalization are traditionally carried out, while coagulants and flocculants, as well as some other chemical agents, are introduced. It is initially difficult to predict the result of the intensive mixing of contaminated water with oxidizing agents (chlorine, ozone). This is due to our insufficient knowledge regarding the complex and diverse mechanisms involved in the interaction of oxidizing agents with the impurities contained in the treated water and with the reagents introduced. The processes of transformation undergone by pollutants and chemicals in water lead not only to a change in their concentration, but also to a change in their chemical nature. At the same time, the hygienic and toxicological properties of water change [1, 5, 8, 12].

This is especially true when using natural waters which contain dissolved organic substances (DOM) that determine, in particular, color. In recent years, it is precisely this type of water that has come to pose one of the most important problems facing water supply around the world. In particular, in Russia there is virtually no river used as a source of water supply with which there would not be a problem connected with reducing its color to the normative level [2, 9, 10,]. In the Primorsky Territory, as in most regions, river and reservoir water can be characterized as low turbid with a relatively low content of suspended solids - coarse dispersed impurities and highly colored.

Thus, it becomes obvious that, at present, the vast majority of water supply systems using traditional technologies - in which the basis is the use of chemical components (chlorine during primary chlorination (prechlorination), coagulants, flocculants and other reagents) - do not fully take into account the nature of water impurities [4, 9, 12, 13]. The result of the interaction can significantly change the properties of water until toxic substances appear in it.

In particular, during the stages of prechlorination and the final disinfection of surface water sources which contain DOM, as well as the use of chlorine reagents, toxic volatile organochlorine compounds are formed that are not removed by traditional methods of water purification. The concentration of these compounds in drinking water is 2-3 times higher than the MPC [1, 4, 5]. Nevertheless, prechlorination in the practice of water treatment is still the most common method of pretreating
natural waters before removing their various impurities at the main facilities. First of all, it seeks to prevent the biological fouling of structural elements in the water treatment devices by suppressing the activity of microflora development. Such microflora, including pathogenic types, always accompany organic pollutants such as DOM. As you know, it is precisely such substances - constituting various derivatives of organic acids (humate-fulvate compounds) - that cause the coloring of water. Until recently, prechlorination was considered an indispensable technological method for the purification of colored water, since it was believed that chlorine, as an active chemical agent with strong oxidizing properties, not only inhibits microflora, but also destructively affects colloidal systems formed on the basis of such aqueous impurities as humate-fulvate substances.

In our opinion, such a mechanism of oxidizing – that is, the destructive effect of chlorine-containing components on colloidal systems which determines, in particular, the color of natural waters – is permissible only to a certain extent. In this embodiment, chlorine appears to be a destructor that destroys DOM colloids and “oxidizes” these substances. In fact, colloids are represented in a rather wide range of dispersion (10-7-10-8m) and when embedding chlorine molecules in the structure of complex organic compounds, it is difficult to determine the form of the substances and their degree of dispersion. Since organochlorine compounds are believed to be more resistant to degradation compared to conventional organic acid salts (humates and fulvates), it can be assumed that they have a higher degree of dispersion, close to true solutions. To date, many studies have confirmed that chlorine, introduced at the first stage of the treatment of waters containing impurities in the form of DOM, is embedded in complex molecules of organic compounds that are recognized as toxic substances. To date, more than 500 different organochlorine compounds (COS) have been identified that are formed after the preliminary chlorination of water containing DOM [1, 4, 5]. In addition, in the molecule structure of complex organics, chlorine ceases to be an active agent that destroys the gene of microorganisms. Therefore, prechlorination ceases to be a barrier to the penetration of various bacteria and viruses into the treated water.

Among the regulatory requirements for the quality of drinking water recommended by the WHO, a number of controlled indicators for toxic organic compounds, including organochlorine, are additionally included. The negative impact of these substances on human health is considered in depth in many works [1, 3, 4, 5]. The removal of organochlorine compounds from drinking water during its treatment is one of the most urgent problems in the water sector of Russia [1, 4, 5]. The main reason for the formation of toxic organochlorine compounds is primary chlorination. As the practice of operating wastewater treatment plants of drinking water supply systems shows, the total content of organochlorine compounds after introducing primary chlorine at a dose of 3-5 mg / l increases tenfold with respect to the source water [2, 4]. This proves that water treatment facilities operating according to the traditional scheme (prechlorination, coagulation, sedimentation, filtration) do not create a reliable barrier and toxic organochlorine substances can remain in drinking water.

Studies have shown that the presence of DOM in natural waters is always accompanied by specific types of microorganisms [6, 10, 12, 15]. At the same time, microorganisms contained in still untreated water should also be classified as aqueous organics. Their development and dying off, as well as the isolation of metabolic products, and the added amount of organic matter can be regarded as a special type of impurity. It should be noted here that such components of organic nature are generally not considered as pollutants that require removal from treated water, including in drinking water supply systems.

The current situation leads a lack of clarity regarding what types of microorganisms face after carrying out prechlorination and the introduction of other reagents, as well as how these microorganisms mutate and adapt (mutate) under new conditions. It is only certain that microflora is not completely suppressed and in the presence of DOM it “participates” in this process throughout the entire process of water purification. It is almost entirely proven that water chlorination, carried out on a large scale, caused the widespread appearance and spread of chlorine-resistant microorganisms [5, 7, 8]. For this reason, it is acceptable to assume that the ability of pathogenic and conditionally pathogenic bacteria to restore their vital activity after disinfection is responsible for the complexity of
ensuring the epidemiological safety of water supply systems. It turns out that over the past 15 years, the stability of pathogenic microflora has increased not only in relation to chlorine (5–6 times), but also to the action of ultraviolet rays (4 times) and ozone (2–3 times) [7]. To a large extent, this is due to the fact that, as it is believed, a significant part of chlorine during prechlorination is used either to oxidize DOM contained in unclear water [7] or to incorporate complex organic molecules into the reservoirs with the formations of toxic organochlorine compounds. In this case, there is a requirement to increase the consumption of chlorine-containing reagents and to increase the contact time of untreated water with chlorine, yet this does not guarantee the destructive effect of chlorine on DOM and sufficiently complete suppression of the activity of microorganisms. In a number of works, it is noted that chloragents are able to affect only the vegetative forms of microorganisms, while viruses, cysts, protozoa spores and helminth eggs are resistant to its effects. This, in particular, is evidenced by the results of a survey on existing water treatment plants.

3. Experimental studies on prechlorination in existing water treatment plants

Thus, the analysis of published works shows that it is fairly acceptable to assume that toxic chlororganic compounds are formed during prechlorination, and that microflora in the presence of DOM is not completely suppressed but rather “participates” in this process throughout the process of water purification. Microorganisms mutate and adapt to changing conditions and only those species which find these conditions acceptable and not suppressive to their viability remain. In this case, the toxic organochlorine substances, which are formed and remain in the colloidal state, turn out to be even more resistant to degradation caused only by their higher degree of dispersion and lower molecular weight. In particular, it is known that organic substances with a high molecular weight (of 30-40 kDa) interact relatively easily with destructors and are removed from the colloidal solution. Substances with a lower molecular weight remain in the solution [9].

The rate of the formation of organochlorine compounds is quite high and this, for example, is confirmed by the experience of operating clarifiers with granular contact loading of a water treatment plant in Fokino (Primorsky Territory). Clarifiers at this station are used as the main treatment facilities which should provide a single-stage treatment of water from the Volchanetsky reservoir. The color of water in it (Fig. 1), which determines the presence of DOM of varying degrees of dispersion and molecular weight, reaches 170-240 degrees Pt/Co in different seasons of the year.

![Figure 1. Changes in water colour in the Volchanetsky reservoir (Fokino, Primorsky Territory) – 2017; – 2018; – 2019.](image)
Moreover, as already noted, chlorine is embedded in complex compounds, reduces its disinfecting potential, and ceases to suppress microorganisms. Evidence of this effect is found in clarifiers with granular contact loading. Structurally, the distribution system of clarifiers of the type KO-2 is a parallel perforated pipe - side bends with shutters. After 7 years of operation, the distribution system was dismantled, during this reconstruction which the main reasons for the decrease in the efficiency of clarifiers were identified. Corrosion causes the destruction of lateral branch elements, leading to the formation of through cavities sized 50-120 mm (Fig. 2). The corrosion process of any metal structure, as you know, starts from the surface and deepens. In this case, recesses (ulcers, spots) are formed on the surface, which are filled with products of the corrosion.

According to the operating experience of distribution systems (tubular lateral branches) of clarifiers with contact overloading (corrosion), there is already observable rusting of the pipelines’ walls in the third year of their operation. However, such defects can be detected only by indirect signs (reduced cleaning effect) and when the sand load is completely removed from the clarifier body or at the time of the accident. The greatest corrosion damage to the lateral bleach outlets occurs with ulcerative and, to a lesser extent, pitting types of corrosion, which at the final stage usually turn into thorough corrosion with the formation of through cavities. The cause of corrosion can only be the increased aggressiveness of the environment, which, under the conditions of the work of clarifiers, is determined either by the presence of chlorine reagents or microorganisms that release aggressive enzymes.

![Figure 2. Removed lateral bleach outlets with grain-like contact loading and fragment (right) with the formation of through cavities.](image)

Solutions of chlorine-containing reagents are known to be corrosive and therefore can cause corrosion damage to elements of the distribution system of clarifiers. During the preliminary chlorination at waterworks when treating natural waters to improve the coagulation process, bleach and disinfect water, and also to improve the sanitary condition of structures, the normalized dose of chlorine (based on active chlorine) is 3-6 mg / l [1, 5, 9] while the content of residual chlorine usually does not exceed 0.5 mg / L.

However, a significant proportion of chlorine is neutralized during the formation of complex complexes of organochlorine compounds since some of it interacts with DOM, and its corrosion activity decreases. At the same time, it turns out that during prechlorination, microorganisms are only partially suppressed. The tuberous forms that appear on the surface of the metal elements of the lateral branches indicate an immobilization of microorganisms in such areas. Colonies of bacteria tend to gain a foothold on immovable objects and this reduces the energy they consume from moving in the water stream. In addition, the accumulation of glandular mucus and the formation of a continuous layer on the surface of the side discharge curtain (see Fig. 2) also indicate the presence of microorganisms. Therefore, the destruction process of the metal elements contained in the clarifier distribution system is determined mainly by biocorrosion. Continuous mucous formations were recorded when structures were examined during the dismantling of distribution systems of clarifiers.
Once dried, such formations are transformed into a dense crust consisting mainly of iron oxides. Thus, microorganisms are definitely not suppressed in the presence of DOM in treated water during the process / stage of pre-chlorination and are the main cause behind the biocorrosion of metal elements in wastewater treatment plants.

Based on a previous analysis of publications, it was determined that microorganisms secrete highly active and quite aggressive enzymes that play an important role in their metabolism, in particular [6; 11, 15, 16]. Enzymes (enzymes) are biological catalysts of a protein nature that are freely released into the environment, catalyze the breakdown of complex organic substances and turn them into simpler ones.

This is what ensures the extracellular “digestion” of organics - its transformation into an assimilable form. For example, an enzyme such as peroxidase catalyzes the oxidation of organic substances by hydrogen peroxide, which can simultaneously provoke corrosive processes. Cytochromes contain iron atoms and ensure the occurrence of redox reactions as a result of the transition of iron from a divalent state to a trivalent one.

A similar mechanism of corrosion destruction experienced by the lateral branches of clarifiers KO-2 is also found in other water treatment plants, in particular, in Shtykovsky (Vladivostok). Here, the period of contact between water and chlorine also does not exceed 10-15 minutes, while the biocorrosion of the steel elements of the distribution system - lateral outlets (Fig. 3. Photos of the authors) forces them to be replaced after 5-7 years.

**Figure 3.** Corrosion of the lateral outlets in Shtykovsky water treatment plants (Vladivostok).

Indirect signs of microflora activating after prechlorination - manifested in the development of biological corrosion processes - are found in almost all water treatment facilities examined in the Primorsky Territory. In addition, biocorrosion is also observed in the structures of cities such as Ussuriysk (Rakovskiy reservoir), Dalnegorsk (Nezhdankinsky reservoir), Dalnerechensk (Bolshaya Ussurka river), Nakhodka (alluvial aquifer of the Partizanskaya river), Bolshoi Kamen (Petrovsky reservoir).

As already noted, in the worldwide practice of water treatment, chlorination is still the most common method of disinfecting aqueous media. This also applies to prechlorination actively used in the pretreatment systems of natural waters. In Russia, according to the regulatory requirements for chlorination, the content of residual chlorine should be at least 0.5 mg / l when it comes in contact with treated water for 30 minutes. It is believed that under these conditions it is possible to reduce the content of E. Coli and other microorganisms, as well as some viruses, by more than 99% [8, 10]. To purify water from microbiological contaminants resistant to the effects of chlorine reagents (hepatitis A virus or Giardia cysts), sufficient efficiency is achieved only with an increase in the contact time of water with chlorine from 0.5 to 3.0 hours while, at the same time, the residual chlorine content must be at least 0.5-0.6 mg / l. In most foreign countries, the same approach is often used, including in the presence of DOM in the source water, causing water staining (i.e. increased intensity of colour).

In particular, a survey (August 28, 2004 - September 4, 2004) of a number of water treatment plants in California and Nevada (USA), where the sources of water supply are rivers in which the
concentration of DOM increases significantly during certain seasons of the year and, accordingly, the color of the treated water increases, showed that prechlorination is also used [17, 18]. Sand traps of various designs, hydrocyclone plants, as well as special sedimentation separation facilities (water ladles, pools) are used as devices for reagent-free water treatment and preliminary chlorination.

At the Pine Lake water treatment plant (California, USA), the primary chlorination of water is carried out in an artificial soil tank (Fig. 4 - photo of the authors), in which the contact of water with chlorine is ensured for at least 40-60 minutes. During periods of precipitation, the color of water can intensify by 200 degrees Pt/Co. At the same time, the content of DOM increases accordingly. At the time of the survey (September 4, 2004), the color of water did not exceed 40 degrees (standard units of Hazen) [17, 18].

**Figure 4.** Ground reservoir for primary chlorination and water settling with a distribution chamber (in front of the lake) (Pine Lake water treatment plant, California, USA).

After stabilizing water treatment through liming and the introduction of reagents (coagulants and flocculants), cleaning is carried out on clarifiers with a suspended sediment layer of the “Pulsator” type, followed by a filtration by quick filters with sand loading. Clarifiers have a fairly simple design: the source water distribution system is made from parallel, perforated asbestos-cement pipes in its lower section, and the clarified water is discharged by a similar system located in the upper section of the reinforced concrete tank. It is important to note that clarifiers do not have metal elements but, during the treatment of water, the precipitate formed consists of ferrous compounds which indicate the presence of DOM (in the form of iron humates) in the source water.

Moreover, towards the end of the clarifier’s working cycle, at the stage prior to the discharge of sediment from its lower zone, glandular formations in the form of mucous sediment accumulate between water inlets on the surface of the asbestos-cement pipes that discharge clarified water (Fig. 5. photo of the authors). Therefore, even with a relatively low content of DOM, color ranges by 40 degrees. After prechlorination and treatment in a contact medium of clarifiers in water, Pt/Co retains a high content of ferrous compounds. These compounds are formed during the relatively delayed destruction of colloidal humate organometallic compounds, which manage to transition from the colloidal state to the state of hydraulic dispersion in the zone from the surface of the suspended sediment layer to the level of the discharge system’s pipes.

Moreover, the characteristic consistency of the sediment located on the surface of the pipes as well as the presence of mucus in its composition indicates that microorganisms are present. The same microorganisms turned out to be resistant to prolonged exposure to chlorine in a contact ground tank and could not help but participate in the process of clarifying water as it passes through a layer of suspended sediment. The accumulation of sediment on the surface of the discharge pipes occurs throughout the entire cycle of the clarifier. The nature of where areas with immobilized microorganisms are located, and the accumulation of mucous sediment between the holes of the
perforation tubes indicates that a significant part of the sediment remaining in the colloidal solution of DOM and microorganisms arrived at the filters together with the purified water.

![Image of perforated tubes with sediment](image)

**Figure 5.** A clarifier with a suspended sediment layer - “Pulsator” and a fragment (on the right) of a perforated asbestos-cement pipes that discharge clarified water; pipes have glandular mucous sedimentation on its surface (Pine Lake Water Treatment Station, California, USA).

Thus, even when there is prolonged contact between treated water and chlorine, and when the source water exhibits a low color index, organic substances and microorganisms remain in the water during treatment in the type of clarifier which has a suspended sediment layer, and a significant part of them accumulates on the filters without the guarantee of their complete extraction after filtering and the completion of disinfection. It remains only to rely on the fact that the toxicity of DOM and the danger of microorganisms remaining after such treatment of water cannot significantly affect human health.

Moreover, while the efficiency of DOM removal due to prechlorination is not determined here, a decrease in color and turbidity can be judged only by the readings of photocalorimeters. According to the transmittance of the treated water, the data is automatically processed and, depending on even minor changes, the flow of chlorine in front of the contact tank, coagulants and lime suspension in front of the settlers is regulated. The efficiency of water treatment at this complex is considered quite sufficient except for periods during which there is a significant increase in the intensity color, and, consequently, an increase in the concentration of DOM.

It should be noted that one of the advantages of horizontal settling tanks during the purification of high-color natural waters is the long residence time of water in them. In the presence of impurities such as DOM in water, the duration of the treatment is of some importance since in horizontal sedimentation tanks, although slow, a decomposition can occur of complex organometallic compounds which have a relatively large molecular weight and are less resistant to degradation.

At a water treatment plant in Reno (pc. Nevada, USA), when examining the treatment facilities, low color of water was also observed (up to 40 deg. PSC). Here, prechlorination is used before sedimentation clarification, but horizontal sedimentation tanks have been reconstructed and equipped with thin-layer shelf modules to increase productivity. Mucous formations accumulate in sedimentation tanks (Fig. 6. Photos of the authors), which determine the presence of microorganisms and, therefore, indicate the presence of DOM in the treated water after prechlorination.
Figure 6. Shelf thin-layer modules embedded in a horizontal settling tank with accumulated mucous sediment and a fragment of the shelf module (on the right) (water treatment complex in Reno, pc. Nevada, USA).

At the last stage of cleaning in Reno, two-layer fast filters are used with a load of crushed anthracite which have a thickness of 150 cm, and with quartz sand (1.2 m) which have a total filter height of about 6.0 m. Such filters can significantly resolve the problem of cleaning high-color water, but with an extreme concentration of DOM with color up to 200 degrees. In this case, on the Pt/Co scale the required degree of purification is not achieved.

4. Findings
The main objective of the technologies used for water purification is its deep clarification and discoloration while ensuring sanitary safety. When treating natural waters containing DOM, traditionally-used coagulation technologies with mandatory pre-chlorination do not elicit such a result. On the contrary, during prechlorination, toxic organochlorine compounds are formed, and microflora in the presence of DOM are not completely suppressed but instead “participates” in this process of water purification throughout this whole process. Microorganisms mutate and adapt to changing conditions and only those species for which these conditions are acceptable remain. Since the viability of certain types of microorganisms is not suppressed, they, detecting their presence and active vital activity, cause the all the metal elements of the treatment devices to undergo biocorrosion, which can only be caused by the sufficiently high nutritional value of the treated water - the presence of DOM in it. Indirect signs of microflora activating after prechlorination, manifested in the development of biological corrosion processes, are found in almost all water treatment facilities examined in the Primorsky Territory.

Thus, an analysis of the results of studies of existing water purification systems both in Russia and abroad confirms that chlorine during pretreatment of natural waters containing DOM is not an effective destructor of complex compounds, but, on the contrary, increases the risk of the formation and increase in the concentration of toxic substances. Weak activity of chlorine in relation to microflora does not exclude the biocorrosion of metal elements in the water treatment devices. It is also confirmed that DOM is present at each stage of purification after prechlorination, and microorganisms of specific species accompany this process. In this case, even after the final disinfection of purified water, the presence of DOM and microorganisms in the supply and distribution systems - in pipelines and directly at the draw-off points - is not ruled out. It should also be recognized that both the diverse composition of microflora, and the degree of danger of microorganisms and organic substances, including toxic ones, have still not been studied adequately. Consequently, modern technologies involved in the purification of natural waters do not always make it possible to guarantee the safety of the use of such a food product as drinking water, and need improvement.
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