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## DRINKING WATER DISINFECTION AND FORMATION OF BY-PRODUCTS

### DEZINFEKCE PITNÉ VODY A VZNIK VEDLEJŠÍCH PRODUKTŮ

#### Abstract

In the paper an account of the contemporary ways of drinking water disinfection and of formation of disinfection by-products is presented. Various types of disinfection by-products with respect to applied water treatment technologies and connected to health risk are discussed. Many of these by-products are toxic and some of them are considered as carcinogenic and mutagenic for human organism. Elimination and regular monitoring of their content are necessary. Therefore technologies of removal of disinfection by-products and of their precursors are presented by the paper as well. The account is complemented equally by basic information about analytical methods applied.

#### Abstrakt

V článku se pojednává o současných způsobech dezinfekce pitných vod a o vzniku vedlejších produktů dezinfekce. Pro různá dezinfekční činidla jsou uvedeny dosud známé vznikající sloučeniny, které mají mnohdy toxický vliv pro lidských organismus. V některých případech byla prokázána i jejich karcinogenita nebo mutagenita.

Odstraňování a pravidelný monitoring těchto látek je nezbytný. Proto jsou v příspěvku uvedeny technologie k odstraňování vedlejších produktů dezinfekce nebo jejich prekurzorů. Přehled je doplněn i základními informacemi o používaných analytických metodách.

**Key words:** Drinking water, disinfection method, disinfection by-products, determination of disinfection by-products.

#### Introduction

Disinfection of drinking water prior to its distribution into water supply systems is necessary and has been used since the early 1900s (White, 1992). Besides chlorine, the first and the most widespread disinfection reagent, there are other disinfectants, such as chloramines, chlorine dioxide, ozone and the UV radiation. In addition, for small sources of drinking waters (e.g. private wells), oligodynamic compounds of copper and silver are used.

Disinfection is generally accomplished by chemical oxidation and it is used within and/or as a final step in water treatment plants following clarification/filtration of water. Characteristics of an ideal disinfectant include an effective removal of pathogens, long-term stability in water, an easy handling and measurement by the analytical methods, no undesirable by-products formation, and low cost.

However, none of the used disinfectants meets all these requirements. Choosing an appropriate reagent is a compromise which depends on the quality of the source water including type of micro-organisms, the size of the supplied population, complexity and reliability of technology, and disinfection efficiency (Haas, 1990).

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The effectiveness of disinfection can be expressed mathematically by the *C.t* concept for a specific micro-organism:

$$\text{Constant} = C^n t, \quad (1)$$

where *C* is the disinfectant concentration in mg/l, *t* is time in minutes for inactivation of fixed percent (mostly 99%), *n* is an exponent in the range of 0.5 to 1.8 for most disinfectants. The *C.t* values are summarised for various micro-organisms and disinfection reagents (Hoff, 1986).

Formation of the disinfection by-products (DBPs) has been known since mid-1970s when gas chromatography has started its rapid development (Bellar et al. 1974; Rook 1974; Symons et al. 1975). The first identified compounds were trihalomethanes (THMs). Since this time, new analytical techniques have been elaborated and new by-products have been found. Most of DBPs are detrimental to human health and must be regularly monitored in drinking water networks. Some of them are potential carcinogens, and a short-term exposure can lead to dizziness, headaches, as well as to problems associated with the central nervous system. Their limit concentrations are usually given by government regulations. There is a toxicity database that reflects a wide range of DBPs resulting from different disinfection practices (Boorman et al., 1999). The aim of this paper is to give a brief account of DBPs produced by the contemporary disinfectants.

## **Disinfectants and their by-products in water treatment**

### ***Chlorine***

Chlorine has been used for drinking water disinfection since the beginning of the last century and it still remains as the major chemical for this purpose round the world. It is introduced in water as a gas or as the sodium or calcium hypochlorite. There is a lot of DBPs, mainly halogenated compounds that originate by the reaction of free chlorine and/or bromine with natural organic matter (NOM), such as humic and fulvic substances, in water (Janda and Švecová, 2000). Free bromine results from the oxidation of the bromide ions in source water. It was also found that not only humic matters serve as haloform precursors, but also algae provides chloroform after chlorination (Hoehn et al., 1980).

The main organic DPBs are trihalomethanes (chloroform, bromoform, bromodichloromethane, dibromochloromethane), chlorinated phenols (2-chloro-, 2,4-dichloro- and 2,4,6-trichlorophenol), haloacetones (1,1-dichloropropanone, 1,1,1-trichloropropanone), haloacetic acids (trichloro-, dichloro, monochloroacetic acid, dibromo- and monobromoacetic acid), haloacetonitriles (dichloro-, trichloro-, dibromo- and bromochloroacetonitrile), chloropicrin, chloral hydrate, 3-chloro-4-(dichloromethyl)-5-hydroxy-2(5H)-furanone, etc. The inorganic DPBs include chloramines and cyanogen chloride (Krasner et al., 1989).

Halogenated DBPs occurring in drinking water in the highest concentrations include THMs and haloacetic acids (HAAs). They are considered to be toxic, THMs were even found to be mutagenic and carcinogenic chemicals for human beings. The limit concentration of THMs in drinking water in the Czech Republic (the governmental regulation No. 376/2000) is 150 µg/l. Unfortunately, the limits of HAAs are missing as yet. 3-Chloro-4-(dichloromethyl)-5-hydroxy-2(5H)-furanone is the member of the hydroxyfuranone class that has been studied most extensively. It is considered to be mutagenic (Kronberg, 1999).

Chlorophenols are featured by their strong odour that indicates their presence in water even at  $10^0$  to  $10^{-1}$  µg/l. They are not toxic at such concentrations, but they cause undesirable taste and odour. The hygienic limits of drinking water are derived from such organoleptic characteristics.

By the so-called sum parameters quick and quite sufficient information about presence of halogenated DBPs in water are often provided. The most important sum parameters are total organic halides (TOX), total halides (TX), adsorbable organic halides (AOX), purgeable organic halides (POX), and extractable organic halides (EOX). The NOM concentrations have been often characterised by the sum parameters such as total organic carbon (TOC) and dissolved organic carbon (DOC).

### ***Chloramines***

Chloramines are formed when chlorine and ammonia are added to water. They persist in water supply systems and continue to disinfect in the long systems. Chloramines are weaker oxidising agents than chlorine. They are therefore less likely to cause undesirable tastes and odours by oxidation of NOM.

By chloramination considerably lower concentration of organic halides than chlorination is produced. On the other hand, an increased production of cyanogen chloride is observed when monochloramine is used as a disinfectant (Jacangelo, 1989; Krasner et al., 1989). Halogenation reactions occur when chloramines are formed before free chlorine reacts with ammonia. In addition, chloramines slowly hydrolyse to free chlorine in the aqueous solution.

### ***Chlorine dioxide***

Chlorine dioxide is a more effective disinfectant and oxidant than chlorine. Unlike chlorine, it does not cause the disagreeable odour of drinking water. Its major benefit is avoiding of formation of chlorinated DBP such as THMs, HAAs, and chlorophenols. However, chlorine dioxide is reduced into chlorite by iron, manganese, and organic matter in water (Werdehoff and Singer, 1987). Under alkaline condition (pH>10) it rapidly hydrolyses producing chlorite and chlorate ions (Aieta and Berg, 1986):



Chlorite is relatively stable in the presence of NOM but can be oxidized to chlorate by free chlorine if added as secondary disinfectant:



Chlorite and chlorate may cause hemolytic anemia at low level of exposure while higher levels of exposure can result in an increase of methemoglobin. In the Czech Republic, the governmental regulation No. 376/2000 gives the concentration limit only for chlorite at 0,20 mg/l. For example in Switzerland, the sum of chlorite and chlorate was limited at 300 µg/l as early as in 1986.

Moreover, by chlorine dioxide some organics are produced (Richardson et al, 1994). Using spectral identification techniques, more than 40 different DBPs (at sub-ng/l levels) were identified including carboxylic acids and maleic anhydrides.

### ***Ozone***

Ozone has had a long history of use for disinfecting and for the control of taste, odour, and colour in Europe. It is the strongest biocide and oxidising agent. When bromide ions occur in raw water, they are oxidised to bromine and this leads to forming of brominated THMs and other brominated DBPs similarly as in the case of chlorination. Reacting with NOM, ozone also forms aldehydes, such as formaldehyde, acetaldehyde, and heptanal (Jacangelo et al., 1989; Song et al., 1997) and many other compounds at low levels, e.g., carboxylic acids, aliphatic and alicyclic ketones, glyoxal, methyl glyoxal etc. (Glaze et al., 1989). Formaldehyde is suspicious of carcinogenicity. According to the regulation No. 376/2000, its limit concentration is 0.9 mg/l. Organic acids are also formed. There are predominantly acetic, formic, oxalic, and succinic acid in treated water.

Bromide ions are oxidated in several steps including intermediate oxy-bromine species HBrO and BrO<sup>-</sup> (pK<sub>a</sub>=8.7):



The other pathways of bromate formation are reviewed by, e.g., Song et al. (1997). A numerical kinetic model was described by Westerhoff et al. (1998). Hypochlorite ions from water chlorination also react with ozone forming chlorate:



The bromate limit in drinking water is 0.025 mg/l.

### ***Ultraviolet radiation***

Ultraviolet light is generated by low-pressure mercury lamps. By UV radiation the chemical bonds of many organic molecules are disrupted and hence it is a potent disinfectant means. It causes specific deleterious changes in the nucleic acids of cells, resulting in their death or mutation. The major disadvantage of UV is the

fact that it leaves no residual disinfectant in treated water. On the other hand, no evidence, that UV treatment increases the concentration of biodegradable dissolved organic carbon and assimilable organic carbon, was found (Shaw et al., 2000).

### **By-product control strategy**

The by-product control strategy includes removing the DBP precursors, changing disinfectants, and/or removing DBPs themselves. The principal DBP precursors are NOM. The content of bromide in raw water is also important. In many cases, the DBP removal is difficult, and that is why the control strategies focus on the first two methods. Regarding the DBPs formation, it is important to determine the risk/benefit ratio of the disinfecting agents and to choose a suitable one.

#### ***Removal of precursors***

Removal of a majority of the DBP precursors from raw water is mostly performed by traditional coagulation and flocculation followed by sedimentation and/or filtration (Fišar et al., 2001). Chlorine is added at the end of a treatment process. By aluminium and iron salts variable amounts of NOM can be removed depending on pH of water. For aluminium salts, the optimum pH range is pH=5.5 to 6.0. Improvements in the removal of NOM can be achieved by the precipitative softening process so-called "enhanced softening" that involves raising the amount of lime added during treatment. However, it subsequently results in greater chemical costs and the increased production of sludge (USEPA, 1999).

Pre-oxidation of NOM prior to coagulation has destructive effects on their high molecular structures and it is supposed to suppress the DBP formation. However, the practice is not so unambiguous. For example, the using of pre-oxidation by potassium permanganate can even increase the chloroform concentrations. It could be explained by the setting of better reaction conditions due to creation of more reaction sites. Pre-oxidation by ozone also leads to similar results (Janda and Strnadová, 1982). There is a lot of the technological factors of ozonation (time, temperature, dosage, way of ozone addition, type of NOM, pH, etc) that can decrease or increase the DBP concentration as it was demonstrated in the case of chloroform by the last mentioned authors. The complexity of this problem has been demonstrated by contrary results of, e.g., Vahala et al. (1999) who have found the significant decrease of AOX (about 37 %) when pre-ozonation was used.

The using of membrane ultrafiltration and nanofiltration enhanced by pre-coagulation gives the similar results as a conventional coagulation/sedimentation process (Bian, 1999). Even in the membranes operated under the cross-flow mode, the humic substances accumulated on membrane surface, because their back transport velocity is always smaller than that of the permeate flux. By pre-coagulation the removal of humic substances is effectively enhanced.

The granular activated carbon (GAC) sorption can be used after filtration to remove additional NOM. Of course, the improved filtration can be reached by membrane technologies.

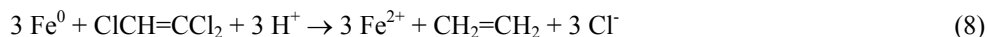
A new way of the NOM reduction is an ozone biofiltration. This method uses the biological mechanisms in the filter bed for the reduction of NOM along with a preceding ozonation. The NOM with a higher percentage of the high molecular compounds show the greatest enhancement in biodegradability (Hozalski et al., 1999).

#### ***Removal of by-products***

The traditional ways of the DBP removal from water are sorption on GAC and air stripping. Sorption on GAC is generally considered as a low effective procedure. The GAC is not effective for sorption of the polar and the high molecular weight compounds (Tuček et al., 1988). By the long contact time and frequent regeneration the GAC application is made expensive. Moreover, GAC is rapidly degraded by chlorine and the addition of a disinfectant to the GAC bed can result into a specific reaction in which previously sorbed compounds leach into treated water.

Stripping technology is suitable for removal of the volatile organic compounds, such as THMs. The design of the packed aeration towers has been described, e.g., by Linek et. al (1998). The main drawbacks of this approach are high costs and non-definite DBP removal because of the DBP transfer from water to air.

A promising possibility of the removal of halogenated hydrocarbons is application of zero valent iron. This in-situ process is based on the filtration of water through a permeable barrier formed by granular iron. The halogenated hydrocarbons are reduced to non-toxic hydrocarbons (Vašek and Janda, 2001; Prousek and Priesolová, 2002), for example:



The technologies for the removal of bromate, chlorite, and chlorate have been developed during the last years. The application of reverse osmosis and nanofiltration is possible but these technologies require the use of conventional treatment (coagulation, clarification, filtration) prior to their application. Our research is intended to find cheaper and easier methods. There are three promising strategies: their reduction by, e.g., ferrous salts, sorption on activated carbon (granular and/or powdered), and decomposition of bromate to bromide by UV radiation.

## Analytical techniques

The disinfection by-products have been determined by a variety of analytical methods. The organic DBPs are analysed mostly by gas chromatography (GC) and/or high performance liquid chromatography (HPLC). For halogenated DBPs, gas chromatography with the electron capture detector (GC-ECD) has been used for many years. Nowadays, modern laboratories are equipped with a powerful combination of gas chromatography and mass spectrometry (GC-MS) which is suitable for the determination of the sub- $\mu\text{g/l}$  concentrations and identification of unexpected and unknown compounds in waters. However, the effective isolation of DBPs is necessary. For this purpose, several extraction procedures have been routinely applied: liquid-liquid, liquid-gas (head space, purge and trap) and solid phase extraction methods. Their principles and applications in water analysis are described elsewhere in literature (Churáček et al., 1990; Popl and Fährndrich, 1999). Volatile compounds are preferably isolated by the head space or the purge and trap techniques on-line connected to a GC separation column. The using of solid-phase microextraction (SPME) for the determination of volatile halogenated hydrocarbons in water samples has been already successfully tested (Janda and Viden, 1998).

Inorganic anions, such as chlorite, chlorate, and bromate, are determined by ion chromatography with a conductivity detector (Hautman, 1992; Achminke and Seubert, 2000; Jackson, 2001). The method for determination of chlorite in tap water by capillary isotachopheresis on-line coupled with capillary zone electrophoresis has been developed (Praus, 2004). In the last mentioned paper the methods for determination of chlorite in drinking water are reviewed.

In the Czech republic, by the regulation No. 376/2000 also application of methods that comply with demands on their precision, accuracy and determination limits is required. Specific methods, which are strictly given but the standardised procedures, are preferred.

The sum parameters, that have been mentioned above, are determined by the single or multiple-purpose analysers in a short time and with a low cost. In case of TOC and DOC, the results are more reliable than the permanganate method that has been still used until nowadays.

## Conclusion

Disinfecting of drinking water is necessary for the prevention of water-borne diseases. The introduction of water chlorination resulted in a large drop in mortality from infectious diseases and it is a major public health advance. However, the disinfection is closely connected with the formation of the by-products that are often toxic for human beings.

The high concentration groups of DBPs that occur in chlorinated drinking water include trihalomethanes and haloacetic acids. By chloramination their amount is significantly reduced. Other types of disinfection, such as adding of ozone or chlorine dioxide or the application of UV radiation, result in the formation of the non-chlorinated DBPs. By the strong oxidative properties of  $\text{O}_3$  and  $\text{ClO}_2$  a rise of aldehydes, ketones, carboxylic acids, and brominated organics is given. The inorganic DBPs include chlorite, chlorate, and bromate ions.

The occurrence of DBPs in drinking water can be limited by a selecting of an appropriate disinfectant, removing of DBPs from treated water, and removing of their organic precursors. Where it is possible, the

precursor removal is more efficient and can be effectively achieved by traditional coagulation technologies. The methods of the removal of oxyhalides have not been completely solved as yet.

DBPs are mostly determined by chromatographic techniques. By the Czech regulation No. 376/2000 the limit values of some DBPs are stipulated and also requirements on the methods that can be used for their determination are prescribed. Unfortunately, some DBPs are not mentioned by their list in this important document, such as haloacetic acids, chlorate, chlorophenols, aldehydes except formaldehyde, ketones etc.

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## Resumé

Dezinfekce je pro zabezpečení mikrobiologické nezávadnosti pitné vody nezbytná. Historicky prvním a stále nejrozšířenějším dezinfekčním činidlem je chlor. Dezinfekční účinnost a oxidační schopnosti chloru jsou velmi dobré, avšak jeho působením vzniká široké spektrum vedlejších produktů. Tyto látky jsou pro vysokou toxicitu závažným zdravotním rizikem. Nejčtetnějšími látkami jsou trihalogenmetany a haloctové kyseliny.

Použijeme-li jiná dezinfekční činidla, můžeme tvorbu těchto vedlejších produktů omezit (chloraminace, ozonizace) nebo úplně vyloučit (oxid chloričitý, UV záření). Změnou dezinfekce však dochází k tvorbě nových produktů. Silné oxidační účinky ozonu a oxidu chloričitého vedou k tvorbě aldehydů (formaldehyd), ketonů a karboxylových kyselin. Současně s organickými látkami se v upravených pitných vodách nacházejí i anorganické produkty, jako jsou chloritany, chlorečnany a bromičnany.

Omezování výskytu těchto látek v pitných vodách je nezbytné. Principiálně toho lze dosáhnout těmito způsoby: Odstraňováním vedlejších produktů z upravené vody, odstraňováním jejich prekurzorů (nejčastěji huminových látek) a změnou dezinfekčního činidla, je-li to možné. První způsob je problematický z hlediska nízkých účinností a vysokých nákladů současných technologií - jedná se zejména o sorpci na aktivním uhlí, stripování vzduchem a membránové filtrace. Proto se dává přednost odstraňování prekurzorů. Ke slovu tak přicházejí technologie koagulace kombinované s dalšími procesy, jako jsou předoxidace, sorpce na aktivním uhlí, membránové filtrace, biofiltrace apod.

Do složitosti problematiky vedlejších produktů dezinfekce bezesporu patří i výběr a použití vhodných analytických metod pro jejich sledování. Stanovení organických látek se provádí pomocí chromatografických technik, z nichž nejučinnější je plynová chromatografie s hmotnostní detekcí. Anorganické látky se nejčastěji sledují pomocí iontové chromatografie. Současná legislativa ve formě vyhlášky 376/2000 Sb. určuje parametry a četnost jejich sledování a klade také požadavky na výběr analytických metod. Bohužel, ve vyhlášce stále chybí některé důležité polutanty, jako např. chlorečnany, haloacetylové kyseliny, chlorfenoly, další karboxylové sloučeniny kromě formaldehydu apod. Z praktického hlediska lze doporučit i doplnění skupinových parametrů např. AOX.

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