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Article:

Are secondary disinfectants performing as intended?

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INTRODUCTION

Global access to safe drinking water and sanitation, the United Nation's sustainable development goal (SDG) 6, supports many related aspirations including good health and well-being (SDG3), sustainable cities and communities (SDG11), and life below water (SDG14). To make drinking water suitable for human use and consumption, water treatment must remove or inactivate pathogens and unhealthy pollutants. Primary disinfectants, broadly defined as chemicals or UV-light specifically added for biological inactivation in a treatment plant or water works, began to be used for drinking water treatment during the late 1800s. In these cases, chlorine was initially added to the raw water (i.e., treatment plant influent) or upstream of filtration, and the extent and performance of any secondary residual (broadly defined as the disinfectants present after treatment as measured throughout the distribution system) is not well documented. The town of Maidstone, England, was likely the first water system to deliberately introduce secondary disinfectants in its distribution system when it applied bleaching powder to clean water mains after a serious typhoid epidemic in 1897 (Baker, 1981). Nowadays, the use of chlorine and other secondary disinfectants is commonplace and, in fact, required in many countries.

Multiple physical and chemical disinfectants are used for primary disinfection during treatment. However, because the secondary residual must persist over long periods throughout the distribution system, the choice of secondary disinfectant is typically limited to chlorine-based disinfectants in the form of free chlorine and chloramines. Free chlorine provides good disinfection capability, but it also reacts with an extensive range of organic compounds forming harmful disinfection by-products (DBPs), many of which are carcinogens and potentially harmful to human health (Plewa, Wagner, & Richardson, 2017). Chloramines, which are formed by reacting free chlorine with ammonia, are weaker oxidants than chlorine itself and therefore can persist longer

in the distribution system and prevent further formation of regulated chlorinated DBPs, although they can contribute to non-regulated nitrogenous DBPs. These desirable characteristics have led many water systems to use chloramines for secondary disinfection, although the challenges of managing nitrification along with the potential toxicity of currently unregulated nitrogenous DBPs may deter their use in the future. Drinking water in many European countries is commonly supplied without any secondary disinfectant; in these areas, many cite the negative aspects of secondary disinfectants in the distribution system such as their undesirable taste and smell and toxicity associated with DBPs (Smeets, Medema, & Van Dijk, 2009) (Rosario-Ortiz, Rose, Speight, von Gunten, & Schnoor, 2016).

The US Environmental Protection Agency (USEPA) Surface Water Treatment Rule requires that water systems treating surface water sources maintain a detectable disinfectant residual in at least 95% of distribution system samples to "ensure that the distribution system is properly maintained and identify and limit contamination from outside the distribution system when it might occur; limit growth of heterotrophic bacteria and Legionella within the distribution system; and provide a quantitative limit which, if exceeded, would trigger remedial action" (US Environmental Protection Agency, 1989). Acknowledging that a lack of disinfectant residual does not necessarily indicate microbial contamination, heterotrophic plate count measurements below 500 organisms per mL can be used for the purposes of determining compliance in lieu of disinfectant residual. The USEPA Ground Water Rule requires primary disinfection for ground water sources with evidence of microbial contamination, but it does not specify the maintenance of secondary disinfectant residual throughout the distribution system (US Environmental Protection Agency, 2006). The European Union Drinking Water Directive does not include secondary disinfectants as a regulated parameter, either as a chemical parameter or as an indicator,

although several microbiological parameters are regulated and the point of compliance for all drinking water is "at the point, within premises or an establishment, at which it emerges from the taps that are normally used for human consumption" (European Commission, 1998).

Considering the factors enumerated in the USEPA SWTR and given the research that has taken place on distribution system water quality since its promulgation, this article is intended to ask the question: are secondary disinfectants are performing as intended?

Indicator or Trigger: Distribution System Integrity and Remedial Action

Understanding the performance and maintenance status of buried infrastructure like water distribution pipelines is an ongoing challenge. A variety of factors both internal and external to pipe networks affect the performance of the distribution system, including water quality changes, corrosion, pressure transients, accumulation and release of biofilm and associated contaminants, surface loading, improper installation, and third-party construction activities. The water industry is still testing and improving how it monitors systems in real-time, although advances have been made in measuring flows, pressures, physical water quality parameters like turbidity, and chemical water quality parameters such as disinfectant residual. Reliable methods for direct measurement of individual microbiological contaminants, particularly pathogens, do not yet exist on near real-time scales, although surrogate measures like adenosine triphosphate (ATP) have successfully shown changes in overall microbial activity.

Disinfectant residual measurements are available in real-time or near real-time, and as such, secondary disinfectants can act as a surrogate for other more complex water quality parameters. Given that the causes of variability in secondary disinfectant concentrations are numerous and often difficult to determine, disinfectant residual is best suited to serving as a trigger

for further investigation rather than as an absolute indicator of external contamination. Grab sampling as part of regulatory microbial monitoring (e.g. total coliform sampling) does not provide statistically-significant characterization of the full profile of secondary disinfectant residual levels in a distribution systems (Speight, Kalsbeek, & DiGiano, 2004). In cases of major contamination events such as a large-volume sewage intrusion, any new and significant disinfectant demand should be visible during real-time monitoring, but other parameters like oxidation-reduction potential (ORP) and conductivity may also be effective for this purpose (Hall, et al., 2007). However, events smaller in both duration and intensity as well as those at unmonitored locations would likely escape detection. As sensor technology improves and more hydraulic, chemical and biological sensors are deployed, the future of distribution system monitoring is one in which maintenance problems and contamination events are detected without the use of secondary disinfectant measurement.

Contaminant Limitation, including Heterotrophic Bacteria and Legionella Control

In terms of distribution system contamination, microbial contaminants pose the greatest acute threat to drinking water safety. Thus, the provision of disinfectant residual throughout the distribution system has been traditionally considered an important part of the multiple barrier approach. But given the typical disinfectant concentrations in use and the highly variable operating conditions of distribution systems, perhaps secondary disinfectants are not delivering the protection that we believe they are. Using the concentration multiplied by time (CT) concept for disinfection, the required contact time for inactivation of different microbial contaminants in the distribution system can be estimated, as shown in Table 1 for a typical distribution system with 0.5 mg/L of free chlorine at pH 7 and 5 degrees Celsius (41 degrees Fahrenheit).

Table 1. Summary of required contact time in distribution system for inactivation of selected microorganisms (at 0.5 mg/L free chlorine, pH 7, 5 °C)

Target organism and target level of inactivation	CT (mg/L-min)	Required contact time in distribution system (minutes)	Source
1-log Giardia lamblia	50	100	(US Environmental Protection Agency, 1989)
2-log viruses	4	8	(US Environmental Protection Agency, 1989)
1-log E. Coli	<0.05	<0.1	(World Health Organization, 2004)
1-log Cryptosporidium	Not effective	-	(World Health Organization, 2004)
1-log heterotrophic bacteria	<0.01	<0.02	(World Health Organization, 2004)
2-log Legionella[1]	50 - 250	100 - 500	(US Environmental Protection Agency, 2015)

1. A wide range of CT values for Legionella have been reported in different studies. Generally, Legionella inactivation requires higher chlorine concentrations than are typically found in water distribution systems. Biofilm associated Legionella may be as much as 100 times more resistant to inactivation than planktonic Legionella.

By the CT measure, secondary disinfectants at typical distribution system concentrations are able to provide protection against heterotrophic bacteria, which broadly includes E. Coli, and viruses within a reasonably short contact time. However, distribution system conditions are usually less ideal than in treatment, with potentially higher levels of turbidity and particulates affecting the disinfectant's efficacy. In fact, several studies have demonstrated the inability of secondary disinfectants to fully inactivate microbiological contaminants under a variety of scenarios (Propato & Uber, 2004) (Besner, Servais, & Prevost, 2008).

Compliance sampling for total coliforms reveals that the US has 10 times more positive sample results than the Netherlands, which does not use secondary disinfectants, when adjusted for population (Linden, Hull, & Speight, 2019). Factors such as distribution system pipe age and integrity, along with differences in treatment, compliance sampling and maintenance strategies, affect the compliance rates between the US and the Netherlands, so it is difficult to make a direct comparison based on the effects of secondary disinfection. Nonetheless, despite the widespread use of secondary disinfectants, microbial compliance rates for the US are not the best in the world.

Furthermore, Table 1 illustrates that chemical disinfectants are minimally effective against protozoan pathogens like Giardia and Cryptosporidium. These microorganisms are not regulated in the distribution system and are rarely analyzed at customer taps outside of special investigations, with the de facto control measure being maintenance of distribution system integrity. In the future, implementation of UV disinfection in the distribution system in areas of potential concern might serve as a replacement to secondary chemical disinfectants, particularly given the increasing affordability and accessibility of UV LED technology (Linden, Hull, & Speight, 2019).

For Legionella and other biofilm-associated microorganisms of concern, their inactivation is further hampered by the protection offered by biofilms. Monochloramine has been shown to penetrate deeper into biofilms than free chlorine, but it is a less powerful oxidant, so monochloramine requires longer contact times for inactivation of pathogens (US Environmental Protection Agency, 2015). Legionella control requires higher disinfectant concentrations than are typically used in distribution systems and therefore very little protection against this microorganism is provided under current conditions. There is a clear need to provide Legionella control, as evidenced by the growing waterborne disease burden associated with this

microorganism (Benedict, et al., 2017), but distribution system secondary residuals as used in practice don't deliver this protection.

Legionella control is currently focused on building water systems, and the role of the water distribution system should be to deliver the best possible water quality to support building water system treatment efforts. The most important water quality components for buildings are likely very low nutrients like organic carbon (much lower than typical finished water concentrations in the US) and absence of seeding organisms from the water distribution system and its biofilms.

The role of distribution system biofilms cannot be neglected in this discussion. Only a small fraction of the microbiological mass in distribution systems is in the form of free-floating bacteria (Flemming, Percival, & Walker, 2002). As an extension to the situation for Legionella, secondary disinfectants alone cannot fully control biofilm formation and the associated accumulation of organic, inorganic, and biological material. A higher secondary disinfectant residual has been shown to influence the composition of the biofilm microbiome and cause greater biofilm mobilization, along with its associated resuspension of contaminants, than lower or absent disinfectant residuals (Fish & Boxall, 2018). Certainly, the presence of secondary disinfectants is affecting the biofilm, but whether this impact is detrimental to water quality or not remains an open research question.

Secondary disinfectants also play a role in reactions related to chemical contaminants, including corrosion and the aforementioned DBPs. Highly complex corrosion chemistry is affected by the water's ORP, which is partly dictated by the secondary disinfectant residual, and changes to secondary disinfectants can have serious consequences for chemical contaminants such as lead (Edwards, Triantafyllidou, & Best, 2009). The use of chloramines as a secondary disinfectant for

compliance with chlorinated DBP regulations can result in undesirable nitrite and nitrate contamination due to nitrification and nitrogenous DBPs which, while currently unregulated, are increasingly a concern due to their toxicity (Plewa, Wagner, & Richardson, 2017). Taken altogether, today's water professionals should question whether secondary disinfection is an integral part of drinking water treatment, especially when looking beyond the microbiological inactivation aspects of maintaining a chlorine or chloramine residual.

CONCLUSIONS

It seems that the current use of secondary disinfectants in the US is partially meeting the goals of the SWTR as defined in 1989, although in several aspects they are not providing protection to the degree that was perhaps intended. Professor Thomas Drown of MIT, reviewing the use of sodium hypochlorite to disinfect water in 1894, posed a question that might be as pertinent now as it was then (Baker, 1981):

'Is it desirable in any case to treat a city's water supply with a powerful disinfectant like the hypochlorites? When the question is put in this bald way I cannot think it will receive the *approval of engineers and sanitarians… in cases where a water supply has got into such a* hopelessly bad condition that nothing will render it safe but disinfection by chloride of soda or chloride of lime, it is high time, I think, to abandon the supply, and in this opinion I feel sure most *water works engineers will coincide.'*

Climate change and population growth have eliminated the option of abandoning current water supplies in most cases around the world. But is the water industry overly reliant on secondary disinfectants to compensate for less-than-ideal treatment and distribution system management? And in the process, are we increasing the toxicity through the creation of disinfection by-products

that will never be fully understood, regardless of the investment in research? Advances in technology and scientific understanding mean that the future could look quite different and the water industry should be considering whether radical changes to managing distribution system water quality could better protect public health.

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