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Water Chlorination. Part 1. Water, Water Almost Everywhere, but Is It Fit to Drink?

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In a world of technological advances, we tend to take for granted some things that are essential to our well-being. Take water, for example. We use water constantly, yet rarely do we stop to consider where it comes from or how it has been treated. In Philadelphia the local water authority supplies water to about two million people every day. This water is drawn from a variety of sources and travels through hundreds of miles of pipes before it reaches our faucets. One of the most amazing things about water distribution is that, in spite of the seeming difficulties in supplying water to so many people in so many places, waterworks can routinely provide water that is safe to drink. That this is possible is due in large part to a process that has had a major impact on public health: chlorination. Let's take a look at this process to learn what it does and why we use it.

A search of the scientific literature turned up several hundred books and articles about drinking water, methods of water purification, and the possible consequences, both good and bad, of some of those methods. In this first part of a two-part essay, we will consider the history of water treatment, the chemistry and biologic activity of chlorine, current water practices, and the possible hazards related to the use of chlorine. In the second part, we will consider alternatives to chlorination of drinking water, organizations that are concerned with water treatment, the current state of water treatment and chlorination worldwide, and what ISI® research fronts reveal about chlorination.

History

The chlorination of drinking water is really a major part of the story of modern water treatment. Consequently, the history of drinking-water treatment is a good place to start a discussion of chlorination. The Safe Drinking Water Committee of the National Research Council, Washington, DC, has provided a good historical review in its 1977 volume *Drinking Water and Health*.[1] Humans have experimented with various ways of obtaining safe water since prehistoric times. The earliest references to water treatment appear in ancient Sanskrit and Egyptian hieroglyphic writings. According to M.N. Baker, associate editor, *Engineering News-Record*, as early as the fourth century B.C., the Greek physician Hippocrates recommended straining and boiling water before drinking it.[2]

In spite of such attempts at water purification, however, it was not until early in this century that the proper combination of knowledge and technology allowed effective protection of the general public from unsafe water. As a result, an enormous number of people throughout history have suffered and died from waterborne diseases.[2] To this day, waterborne diseases are a major public-health problem, especially in the less-developed world.

A London physician, John Snow, provided the first proof that public water supplies could spread diseases among humans. In 1854 Snow showed that outbreaks of cholera could be tied to the use of drinking water contaminated with sewage.[3] Snow's work contributed to efforts to promote water-purification measures such as sand filtration that could remove some of the disease-causing agents found in water.

Until the late nineteenth century, most attempts at water treatment simply aimed at clearing up turbid waters—those containing enough solid particles to make them appear cloudy. In the latter part of the century, however, the first bacteriologists, such as Germany's Robert Koch, presented convincing evidence for a germ theory of disease, which provided a more important reason for treating water. In 1884 Koch isolated *Vibrio cholerae*, the bacterium that causes cholera.[4] It was later found that sand filtration of water, already in use in some cities, removed the cholera bacteria from the water. Filtration was thus established as a method of preventing waterborne disease.[1] Incidentally, cholera will be the subject of a future essay in *Current Contents®*.

Filtration of drinking water was first used in the US around 1890. As in Europe, water supplies were run through large sand filters to remove particles, including organisms, that degraded the water quality. Here, the principal concern was not cholera, as in Europe, but typhoid fever, caused by another bacterium, *Salmonella typhi*.[1]

While filtration dramatically improved the quality and safety of drinking water in many places, it was its combination with chlorine that provided a practical, inexpensive means of controlling bacteria in water. Baker gives a detailed account of the history of chlorination. G.A. Johnson, of Hering and Fuller, a New York manufacturer of water-treatment equipment, introduced chlorination as a process for purifying the water supply of the Chicago stockyards in 1908.[5] Later that year, the water company supplying Jersey City, New Jersey, established the first facilities for chlorinating an urban water supply.

Interestingly, Jersey City's chlorination plant was a successful attempt by a private water company to avoid a large expense. Adding chlorine to the water was cheaper than paying to build sand filters or preventing contamination of the city's water source by sewage. Allen Hazen, author of a 1914 book on water treatment, noted that adding chlorine to water was an attractive solution because chlorine was plentiful and inexpensive.[6] Advocates of chlorination received a major boost when a New Jersey court ruled that the process met a contractual requirement that the East Jersey Water Company supply Jersey City with water that was pure and wholesome for drinking.[2] By 1914 most of the water supplied to US cities was being chlorinated.[6]

Chlorine

What is this substance that water suppliers put in our water? In Webster's, chlorine is defined as "a common nonmetallic univalent and polyvalent element belonging to the halogens that is best known as a heavy greenish yellow, irritating, toxic gas of disagreeable odor...and is used chiefly as a powerful bleaching, oxidizing, and disinfecting agent in water purification...."[7] For purposes of this discussion, the most significant points of this definition are that chlorine is a halogen and that it is a strong oxidizing agent and disinfectant. As an oxidizing agent, it has a strong tendency to withdraw electrons from other atoms or molecules. Chlorine is commonly produced by electrolysis, the splitting of sodium chloride in aqueous solution by applying an electric current.

Chlorine Chemistry

According to a review by J. Carrell Morris, now retired and former chairman of the American Water Works Association's Research Committee on Disinfection and Chlorination, about 73 percent of all municipal potable (drinkable) water supplies in the US, delivering 95 percent of all public potable water, use chlorine as a disinfectant.[8] The reasons for such widespread use are summarized by Gordon M. Fair and colleagues, Graduate School of Engineering, Harvard University.[9] Chlorine can be liquefied under pressure at room temperature, making it easy to store and transport. Chlorine is also highly soluble in water, making it easy to add to water supplies in carefully controlled amounts. Chlorine gas reacts rapidly with water to form hypochlorous acid and hydrogen and chloride ions. Hypochlorous acid, in turn, reacts instantaneously and reversibly with water to form hypochlorite and hydrogen ions. These reactions are shown in Figure 1. The hypochlorous acid and hypochlorite ions together are termed "free chlorine." The relative concentration of hypochlorous acid and hypochlorite ions depends on the pH, or acidity, of the water. The relative hypochlorite ion concentration increases sharply above a pH of 7.3. The percentage of free chlorine present as hypochlorous acid, however, declines sharply above a pH of 7.5. Since hypochlorous acid is the principal disinfectant in chlorine solutions, the disinfectant efficacy of chlorine also declines above pH 7.5.

Figure 1: Reactions of chlorine in water.

$$Cl_2 + H_2O \longrightarrow HOCl + H^+ + Cl^-$$

$$HOCl \xrightarrow{H_2O} H^+ + OCl^-$$

Cl_2 = chlorine gas
H_2O = water
$HOCl$ = hypochlorous acid
H^+ = hydrogen ion
Cl^- = chloride ion
OCl^- = hypochlorite ion

Hypochlorous acid can react with ammonia or organic amines that may be present in water to form compounds called chloramines. These compounds retain the oxidizing power of chlorine but have lower disinfecting powers. Free chlorine reacts with a wide variety of inorganic and organic compounds. The reactions may involve oxidation, in which chlorine is not added to the organic molecules, or it may involve addition of chlorine to form chlorinated organic compounds. Both types of reactions, by using up available chlorine, create a "chlorine demand" that can reduce the disinfecting power of chlorine when organic compounds are present in the water. Sufficient chlorine must therefore be added to overcome the effects of such "side-reactions."[9]

Biologic Actions of Chlorine

D.E. Green and P.K. Stumpf, Department of Medicine, College of Physicians & Surgeons, Columbia University, New York, first elucidated the biologic activity of chlorine in 1946.[10] They noted that a chlorine level of 0.2 to 2.0 parts per million (ppm) is sufficient to disinfect water that is not grossly contaminated with organic, nitrogenous material. They also showed that chlorine interferes with a bacterial enzyme, present only in minute amounts, that is necessary for the oxidation of glucose, a vital cell function. They postulated that chlorine is a more effective disinfectant than other oxidizing agents because its small molecular size allows it to penetrate the bacterial cell membrane, where it can react with the glucose-oxidizing enzyme. Thus, exposure of bacteria to chlorine for only half a minute reduces the bacterial oxidation of glucose by 95 percent, while a five-minute exposure is sufficient to kill the bacteria. Green and Stumpf noted that certain types of spores, which are nongrowing, resistant forms of bacteria that do not depend on glucose oxidation, are not very susceptible to chlorine treatment.[10]

Chlorine kills enteric (intestinal) protozoa (nucleated microorganisms that are generally unicellular, motile, and nonphotosynthetic) such as *Entamoeba histolytica*, flatworms such as schistosomes, and viruses such as those that cause polio and hepatitis,[9] but not as effectively as it kills bacteria. Incidentally, I will also discuss schistosomiasis in detail in a future essay. Richard S. Engelbrecht, Department of Civil Engineering, University of Illinois, Urbana-Champaign, and colleagues reported on the inactivation of eight different types of viruses, including six enteric viruses, by chlorine. They noted that the viruses differ in their susceptibility to chlorine and in the effect of pH on that susceptibility.[11]

Some organisms that cause disease are resistant to chlorine treatment. Gunther F. Craun, Health Effects Research Laboratory, Environmental Protection Agency (EPA), Cincinnati, Ohio, reviewed outbreaks of giardiasis in the US. This disease is caused by a protozoan, *Giardia lamblia*. Craun noted that disinfection of water alone at conventional contact time and concentration is generally not sufficient to prevent the spread of waterborne giardiasis, which has often been found to occur in unfiltered water supplies. According to Craun, such water supplies are usually found in rural, mountain areas and are often so cold that the reactions of chlorine are slowed significantly.[12] However, Shun Dar Lin, Water Quality Section, Illinois State Water Survey, Peoria, cites evidence that outbreaks of giardiasis are increasing and that the organisms can occur in almost any surface water supply. Outbreaks of waterborne giardiasis are usually attributable to failures in water-treatment systems.[13]

Chlorination and Water Treatment

George E. Symons, a consultant and technical editor to the journal *Water & Sewage Works*, discussed the role of chlorination in water treatment today. In the US, a number of methods are generally combined to treat water. A typical process might begin with flocculation or coagulation: chemicals such as alum (a double sulfate of aluminum and an alkaline earth element or ammonium) are added to the water, causing solid particles to clump together. Large amounts of particles can be removed from the water by allowing these coagulated solids to settle out. The water can then be filtered through a sand filter. Filtration may be rapid or slow, depending on the system. If necessary, the water can be aerated by spraying it to remove objectionable odors. Chemical disinfection can occur before or after water is filtered; "booster" chlorination is sometimes practiced after the water has entered the distribution system. Since residual levels of chlorine remain in the water for some time after chlorination, the chemical can continue to disinfect the water after it leaves the treatment plant. The persistence of chlorine in the water is considered important because of the opportunities for contamination in the distribution system.[14] Methods of chlorination in the US have been essentially unchanged for over 50 years.

Incidentally, some water-treatment plants also add fluorides to water as a means of preventing tooth decay. Fluoridation is a controversial subject that we will examine in the future.

Chlorination is not limited to drinking-water supplies. Industries use chlorine to prevent the fouling of cooling-water systems by microorganisms. Food-processing plants use chlorinated water to preserve the freshness of foods by killing bacteria that cause spoilage. Sewage-treatment plants add chlorine to raw or treated sewage to reduce the bacterial count before the sewage is released into rivers or other bodies of water.

Just about anyone who has ever used a swimming pool is aware of chlorine. The chlorine level in a pool has to be higher than the level in drinking water in order to protect swimmers. According to Steven Clark, Office of Drinking Water, EPA, Washington, DC, the chlorine level in a swimming pool might be about 0.5 to 1.0 ppm, compared with perhaps 0.1 to 0.3 ppm in the drinking water that reaches consumers.[15] A series of volumes edited by Robert L. Jolley, Chemical Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, and colleagues discusses the uses of water chlorination in detail, along with many other aspects of the process.[16-20] These volumes, entitled *Water Chlorination: Environmental Impact and Health Effects*, are the proceedings of a biennial conference of the same name, sponsored by such national agencies as EPA and Oak Ridge National Laboratory.

The benefits of water chlorination have been dramatic. The disinfectant action of chlorine with filtration can completely eliminate a wide variety of disease-causing organisms. Nevertheless, a growing number of investigators have been finding evidence that chlorine may have adverse effects as well. Some of these effects have been well documented, while others are more speculative.

Adverse Effects

The adverse effects of chlorination result both from chlorine and from compounds formed by the reactions of chlorine with other chemicals present in water. First, chlorine itself affects living organisms and ecosystems. William A. Brungs, National Water Quality Laboratory, EPA, Duluth, Minnesota, discusses the effects of chlorine on aquatic life. As I noted above, wastewaters are frequently chlorinated to reduce the numbers of pathogenic organisms released into the environment. Brungs states, however, that chlorinated wastewaters, when released into rivers, have toxic effects related to the levels of residual chlorine. Chlorine can kill aquatic life. The lethal level varies from one species to another. A variety of tests indicate that trout, salmon, and plankton (surface-dwelling microscopic organisms that serve as food for fish and other larger organisms) are particularly susceptible to chlorine toxicity. For example, 50 percent of rainbow trout are killed within 96 hours by a residual chlorine level of 0.014 to 0.029 mg per liter. Further, long-term exposure to chlorine can be toxic at levels much lower than those that will quickly kill affected organisms. Warm-water fish, snails, and crayfish are somewhat less susceptible than cold-water fish.[21] Ekrem V. Kalmaz, Department of Engineering Science and Mechanics, University of Tennessee, Knoxville, and Oak Ridge National Laboratory, notes that chlorine affects estuaries and seawater as well as rivers. These toxic effects, however, are more complex than those in fresh water, in part because seawater contains more chemicals that can react with chlorine.[22]

Jack Coughlan, Marine Biology Laboratory, Fawley Power Station, Hampshire, England, and John Whitehouse, Central Electricity Research Laboratories, Leatherhead, England, have reported that chlorinated water released by electric power plants has affected all classes of marine and freshwater plankton. Smaller plankton organisms tend to be less tolerant of chlorine than larger ones. According to the study, the wider ecological implications are difficult to assess.[23]

Chlorine in its gaseous form can be harmful to humans. Frederick W. Koerker, manager, Quality Standards, Dow Chemical Company, Midland, Michigan, in the *McGraw-Hill Encyclopedia of Science and Technology* notes that this strong oxidizing agent attacks the tissues of the nose, throat, and lungs. A concentration of 15 ppm in the air can cause irritation. Chlorine's strong odor, however, allows easy detection of any hazard. Gaseous chlorine is used in some water-treatment plants, but where handling the gas presents problems, liquid or powdered disinfectants containing sodium hydrochlorite or calcium hydrochlorite can be used.[24]

Toxic Interactions of Chlorine

Of growing concern is a wide range of chemical compounds formed by the reaction of chlorine with organic matter in water. Organic contaminants are increasingly found in water supplies because of rising pollution of our streams and rivers, but some are produced by natural processes as well. The scientific evidence relating to these organochloride reaction products is detailed in the volumes edited by Jolley and colleagues.[16-20] The evidence presented indicates that some of these compounds can cause cancer and other diseases in test animals and thus may be a risk to humans. Of particular concern are compounds of a class called trihalomethanes, commonly known as THMs, which are forms of methane—a single carbon atom bonded to four hydrogen atoms—in which three of the four hydrogen atoms have been replaced by chlorine, bromine, iodine, or fluorine. THMs include chloroform, bromoform, and iodoform molecules.

The toxic properties of THMs and other chlorinated organic compounds have been demonstrated by a variety of experiments. Examples of these will indicate the kinds of evidence available.

Albert M. Cheh and colleagues, Gray Fresh Water Biological Institute, University of Minnesota, Navarre, studied products of chlorination for possible mutagenicity, the capacity to induce changes in genetic material. They exposed *Salmonella* bacteria to untreated water and to water that had been treated with either chlorine or chloramines. Chloramines are compounds formed by the reaction of dilute hydrochlorous acid with ammonia.[7] Based on the Ames test (a standard assay of mutagenicity), the chlorinated water had significant mutagenic activity while the untreated water had none. The researchers concluded that the mutagenic contaminants in the chlorinated water were produced by the process of chlorination. Since chloramines are less reactive than chlorine, they resulted in less mutagenicity.[25] You will hear more about chloramines in Part 2.

L.W. Condie and coworkers, Toxicology and Microbiology Division, Health Effects Research Laboratory, EPA, examined the toxicity of halomethanes, including THMs, in a mammalian system. They fed laboratory mice various high doses of the chemicals for two weeks. Exposure to the test compounds produced liver and kidney damage in the mice.[26]

Much of the concern about THMs stems from evidence that they can cause cancer. Melvin D. Reuber, Frederick Cancer Research Center, National Cancer Institute (NCI), Bethesda, Maryland, summarizes much of the evidence as it relates to chloroform—the most common THM in drinking water. Animal studies have shown that chloroform, given orally in high doses, can induce malignant tumors in mice, rats, and probably dogs. The organs affected include the liver and kidneys. In addition to being carcinogenic, chloroform also produces toxic changes in the tissues of various organ systems.[27]

How great is the hazard presented to humans by chlorinated compounds in drinking water? Some studies have claimed links between chlorination of water and the incidence of certain cancers in human populations, but others have not. Three of these studies will serve as examples.

Kenneth P. Cantor, Environmental Epidemiology Branch, NCI, and colleagues studied associations between sex- and site-specific cancer mortality rates and the levels of THMs in the water supplies of US counties. Their analysis took into account socioeconomic, industrial, and demographic factors. Data from the study showed correlations between the levels of THMs and the mortality rates for several types of cancer, including those of the bladder and brain in men and women, as well as non-Hodgkin's lymphoma and renal cancer in men. Cantor and his coworkers believe that the evidence that THMs cause cancer in humans is sufficient to warrant further study.[28]

Marise S. Gottlieb and Jean K. Carr, Tulane University School of Medicine, New Orleans, examined populations in 13 parishes (counties) of Louisiana, a region marked by very high rates of certain cancers. The surface water in the study area contains high levels of organic contaminants. Gottlieb and Carr divided the study populations into three groups according to whether they received nonchlorinated groundwater, water containing less than 1.09 ppm chlorine, or water containing more than 1.09 ppm. The researchers found an association between use of chlorinated surface water and an increased incidence of rectal cancer. They also found possible increased risks for cancers of the brain and breast associated with chlorinated surface water.[29]

Ronald J. Kuzma and coworkers, Department of Environmental Health, University of Cincinnati, classified the 88 counties of Ohio according to whether the majority of the people in those counties received surface water or groundwater. As in the Louisiana study, surface water contained higher levels of organic contaminants than the groundwater. The researchers examined annual age-adjusted cancer mortality rates for the counties with respect to the type of water supply. The analysis showed that drinking surface water was associated with increased mortality resulting from cancer of the stomach and bladder among white males and cancer of the stomach in females. According to Kuzma and his colleagues, carcinogenic compounds in the surface water were a likely cause of the increased cancer mortality.[30]

Does chlorination of drinking water actually cause cancer in humans? Joseph A. Cotruvo, Office of Drinking Water, EPA, believes that the epidemiological evidence is still inconclusive. His agency is responsible for setting and enforcing drinking-water standards. Cotruvo points out that apparent links between chlorination and cancer may be confounded by factors such as population diversity and mobility. He nevertheless believes that current standards for controlling THMs in drinking water are fully warranted by the results of animal studies.[31]

Public and governmental concern led to the 1974 passage of the US Safe Drinking Water Act, which was amended in 1977.[32] This act authorized the EPA to establish national drinking-water standards. In 1979 the EPA promulgated the current regulations that permit a maximum of 0.10 mg total THMs per liter of water.[33]

According to Cotruvo, regulating THMs does not currently represent a source of controversy. He notes that water authorities throughout the US either have complied with the regulations or are attempting to do so. He noted that debate may be renewed in about a year and a half, when the current regulations come before Congress for revision.[34]

Conclusion

Chlorination has produced tremendous benefits for people—vastly improving public health by eliminating or reducing the incidence of waterborne diseases. At the same time, we must recognize that chlorination presents possible hazards to human health and the environment. Increased understanding of the effects of chlorine and chlorinated compounds, combined with improved technology for detecting hazards and preventing them in the future, makes it possible to "fine-tune" the processes by which we purify drinking water. By taking appropriate steps, we can further improve public health by preventing chronic exposure to low levels of hazardous substances. Risk-analysis studies are quite relevant here, as we must weigh the risk of chlorination or any other technology against the worse risk of contaminated water. We have discussed risk-analysis studies in previous essays.[35]

In Part 2 we will examine the alternatives to chlorination for purification of drinking water to see which, if any, of these methods hold promise for practical application. We will review organizations and agencies around the world that are concerned with chlorination and the broader issue of water treatment, and we will assess drinking-water treatment in different countries. Finally, a look at ISI's research fronts will show what the scientific literature has to say about chlorination and water treatment.

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Special note to readers: ISI® is introducing a new and improved version of its *Sci-Mate*® software this year. The new version of the *Sci-Mate Searcher* and *Manager* will be demonstrated at the American Chemical Society meeting in Chicago from September 8 to 13 and at the Information Space '85 exhibition in Bournemouth, UK, from September 16 to 19. The entire *Sci-Mate Software System*, including the *Searcher*, *Manager*, and the brand-new *Editor*, will be demonstrated at the American Society for Information Science (ASIS) meeting in Las Vegas, Nevada, from October 20 to 24; at Online '85 in New York, from November 4 to 6; and at the International Online meeting in London, from December 3 to 5. The modifications to *Sci-Mate* will be discussed in detail in an upcoming essay.