Control of Water Quality Deterioration in Water Distribution Systems:

Part 1: Presence of of Methane in IllinoisWell Water Supplies

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Summary

Part 1 of this series details the widespread presence of methane in Illinois ground waters and assesses its effect on distributed drinking water quality.

Evidence is present ed for the s timulation of microbial growth by methane.

Microbial Interference with Iron Removal Processes

Beginning in 1961, a continuing, long-term series of water treatment plant performance studies was initiated to evaluate problems related to process failure in iron removal plants treating ground waters in central Illinois (Komolrit, 1962). Toward the end of each filter cycle, ferrous ion concentrations in the filter effluents and finished waters were often found to exceed those initially present in the well water sources.

While aeration and filtration were found to be consistently successful both in introducing oxygen and precipitating iron oxides and carbonates in the alkaline Illinois well waters, high ferrous ion concentrations were commonly observed in the finished water effluents from pressure sand filters (Ghosh et al., 1966). During the filtration cycle, progressive depletions in dissolved oxygen concentrations, accompanied by increases in heterotrophic bacterial colony counts, were observed. Finally, when dissolved oxygen was totally consumed within the sand filter bed, ferrous ion was found to return to solution.

Owing to this initial evidence of microbially-mediated deterioration of water quality during filtration, a sequence of full-scale and pilot plant studies were conducted to detail the chemical influences of microbial activity and evaluate methods for controlling microbial growth and oxygen depletion in Illinois iron removal plants. Early studies employing the carbon-chloroform extract (CCE) procedure to quantify organic matter in ground waters indicated that Illinois ground waters contained substantial quantities of dissolved organic matter. Extracts of fractions of the organic residues recovered were found to be capable of sequestering ferrous ion and peptizing precipitated iron oxides (Robinson et al., 1967).

Comprehensive field and pilot plant studies also yielded evidence that microbial growth on sand filter beds mediated the chemical transformations leading to water quality deterioration. Oxygen depletion during the filtration cycle was the most readily-observed and sensitive index of the degree of microbial activity. Stoichiometric calculations confirmed that most of the oxygen introduced during aeration was consumed in the microbially-mediated process of nitrification. Nitrification resulted in the consumption of ammonium ion with the production of an equivalent amount of nitrite plus nitrate ion (O'Connor and Baliga, 1970).

Subsequently, field studies were conducted at the Rantoul, Illinois, water treatment plant to evaluate methods for controlling bacterial growth, nitrification and oxygen depletion in sand filters using high dosages of chlorine or potassium permanganate during the filter backwash cycle (Baliga and O'Connor, 1971).

Control of Microbial Nutrients: Methane

Buswell and Larson (1937) reported the widespread occurrence of methane in Illinois ground water. Methane gas was believed to be produced within glacially-covered beds of peat and organic-rich sediments. The abundance of methane in numerous regions of Illinois had led to the development of up to 460 glacial drift gas wells, reportedly yielding up to 4800 m^3 per day of gas, but varying widely in methane content. The gas wells were commonly used to recover methane for home heating and cooking.

In water supply wells, the outgassing of methane after ground water had been brought to the surface led

Implication of Methanogenic Organisms

Culturing of slime accumulations from Sullivan, Illinois, the University of Illinois experimental iron removal pilot plant and the Champaign-Urbana (Northern Illinois Water Corporation) water distribution system indicated the presence of a methanogenic organism which utilized methane as a carbon source (Gunsalas et al., 1972). The methane-oxidizing organism isolated produced an extensive capsule (consistent with the extensive slime growth observed) and exhibited taxonomic properties similar to *Methylomonas methanica*. The authors concluded that methane served as the primary energy source for the initial growth of methane-utilizing organisms which subsequently produced compounds (e.g., methanol) which led to further development of a diverse heterotrophic bacterial community in the distribution system. Microbial slime growth was particularly extensive at Sullivan where the well gas consisted of 67 percent methane.

According to Stanier et al. (1976), the early development of knowledge regarding the obligate methylotrophs (aerobic methane-oxidizing bacteria) was relatively slow. For decades, *Methylomonas methanica*, a polarly-flagellated rod, was the only known methane-oxidizer. Many new types have now been isolated. All have complex intruded membrane systems, resembling those found in certain nitrifying bacteria.

The obligate methylotrophs grow slowly (doubling times of 4 to 6 hours) and can use both ammonium and nitrate ion as their nitrogen source. Since trace amounts of nitrite ion are formed, the methane-oxidizers are considered to be mini-nitrifiers. Methane is their best growth substrate. If methanol is formed as a direct product of their metabolism, other facultative methylotrophs evolve. These are also polarly-flagellated rods. The best-known facultative methylotroph is *Hyphomicrobium*, a powerful denitrifier. This may be why nitrate may be again depleted once nitrification has occurred in a water distribution system. *Hyphomicrobium* is enriched by use of a medium containing methanol and nitrate. Oxidation of methane and methanol to completion yields carbon dioxide, resulting in lowered pH in a closed system.

Indices of Reducing Conditions in Ground Water and the Genesis of Methane

Particularly where organic matter is abundant, reducing conditions prevail in ground waters. Within meters of the ground surface, oxygen may be depleted or totally absent in soil pore water due to the respiration of aerobic heterotrophs. At greater depths, nitrate ion is commonly low or absent owing to the activity of denitrifying organisms which convert nitrate ion to nitrogen gas. As a result, nitrogen is often abundant in the mix of dissolved gases emanating from ground water after it is pumped to the surface.

Where sulfate-bearing minerals are present in the subsurface and reducing conditions are severe, hydrogen sulfide may be found in aqueous solution. Finally, where reducing conditions are particularly severe, fastidious methane-fermenting bacteria inexorably convert organic residues in the ground to methane. In such regions, the largest percentage of the well gas may consist of methane (O'Connor et al., 1999).

Early Studies of Methane Removal by Air Stripping

Methane in the Normal, Illinois Well Water Supply

Because of microbial slime growth in the Normal, Illinois, water system, an analysis (Neff, 1978) was conducted for methane in the first in a series of water supply wells to be drilled in a newer and deeper well field. Methane was determined to be "8.2 cubic feet per 1,000 gallons" or 16.4 g CH_4/m^3 while ammonium ion, the preferred nitrogen source for the methanogens, was found to be 5.6 g N/m^3.

Subsequent plant evaluation studies at Normal, IL have indicated that, since methane appears to contribute significantly to microbial growth, it should become a treatment plant and distribution system operational control parameter. Pilot aeration studies have indicated that effective aeration can reduce methane concentrations to less than 1 g CH_4/m^3 (O'Connor et al., 1999).

While it is not presently known how constant methane concentrations are in specific wells, it is evident that methane concentrations have remained high in Normal's eight deeper wells over two decades. Moreover, at least for the 16 wells in Normal's system, there appeared to be a direct relationship between non-purgeable organic carbon (NPOC) and ammonium ion concentrations. Non-purgeable organic carbon was approximately 2 g C/m^3 for each 1 g N/m^3 of ammonium ion.

Control of Microbial Nutrients: Ammonium Ion

A survey of the occurrence and control of nitrification in chloraminated water systems (AWWARF, 1995) included case studies from six surface water supplies and one ground water supply. Since most of the utilities surveyed were adding ammonium ion to form chloramines, "discontinuing the feed of ammonium ion and disinfecting with free chlorine only" was recommended to control nitrification.

However, in many groundwaters, naturally-occurring ammonium ion concentrations are high and consistently exceed those periodically found in surface waters. An option for ammonium ion removal available to those ground water treatment facilities which utilize cation exchange resin for softening is to reduce the exchanger throughput to allow for near-complete removal of ammonium ion (Brazos et al., 1996). Some softening (calcium and magnesium exchange) capacity is lost in this procedure.

The long-term effectiveness of breakpoint ammonia reduction treatment (BART) in controlling both distribution system nitrification and the progressive loss of chloramine residuals was successfully demonstrated at Willmar, Minnesota (Murphy et al., 1998). Comprehensive distribution system sampling was conducted for 12 months following the initiation of the ammonium ion oxidation procedure. Nitrification diminished within two months and persistent chloramine residuals were maintained. Nitrite ion concentrations in the distribution system were significantly reduced by the BART procedure.

Summary: Removal of Microbial Nutrients

Where microbial nutrients, such as methane and ammonium ion, contribute significantly to microbial growth during treatment and distribution, incorporation of specific processes for the removal of these microbial nutrients is indicated. In conjunction with nutrient removal, an expanded and more frequent

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