INTERNATIONAL JOURNAL OF CONSERVATION SCIENCE
ISSN: 2067-533X Volume 1, Issue 3, July-September 2010: 175-182 www.ijcs.uaic.ro

EFFECT OF WATER TREATMENT RESIDUALS (ALUMINUM AND IRON IONS) ON HUMAN HEALTH AND DRINKING WATER DISTRIBUTION SYSTEMS.

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Abstract

The aluminum and iron contents in drinking water can mainly be derived from the water treatment process because these metal ions is commonly used as reactant for coagulation-flocculation. When the optimum physico-chemical condition of the treatment of raw water is not well established, the probability of the presence of residual coagulants in treated water increases. In most water treatment plants, variation in raw water quality and the quantity make necessary a good monitoring of the optimum condition of treatment. In spite of these, consumers are exposed to dangerous consumption of residual coagulant in drinking water. There are numerous studies about these metals associated with various health problems and obstructing water distribution networks. The paper represents a review of the main literature in the field.

Keywords: *Coagulant residual; Al (III); Fe(II, III); Human health; Metal deposit*

Introduction

In tropical regions, ground water and surface waters, especially river waters constitute the principal source of raw water for drinking water production. The river waters are soft, have low levels of alkalinity and mineralization. The production of drinking water is a major problem of developing countries and particularly those in tropical area of Africa. There are serious problems regarding the water supplies in the towns with high density of population, because the quantity and quality of groundwater are not sufficient to ensure the total potable water demands. It is necessary to use surface waters to produce potable water. Surface waters composition (natural organic matter, turbidity) depends of season and geographic area. Physico-chemical method is commonly used in drinking water treatment plants in order to remove turbidity and natural organic matter [1-2]. In the conventional treatment of drinking water, the purification has three steps: clarification, filtration and refining. During the clarification, coagulant is introduced into the water to destabilize suspended solids, dissolve organic matter and other colloidal substances. These substances are then aggregated to form floc which will settle down. This separation is followed by sand filtration.

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The disinfection step by chlorine, eliminate the pathogenic microorganisms. To improve the treatment process we will focus on the clarification. It aims to provide water free of suspended solids, dissolved organic matter and containing the least possible of residual coagulant. The most widely used coagulants in drinking water treatment plant are aluminum and iron salts [3-6]. During clarification most of the coagulant used is found in sewage sludge. However, it remains a low level in potable water when the hydrolysis of coagulant is incomplete. When some parameters such as the pH, quality of raw water are not well controlled [6], there is a risk to human exposition to residual aluminum or iron in drinking water or formation of solids that cab be deposited in the drinking water treatment network. In tropical regions, there is generally a low hydrolysis of coagulant because surface water has a low mineral content and low alkalinity [7].

In the town of Ngaoundere (Cameroon) it is widely observed that the tap water is clear but after a rest period there is a deposit. Scientific studies [7] showed that this deposit contains aluminum. The aluminum and iron contents in treated water can mainly be derived from two sources: (i) poor hydrolysis of metals during treatment, (ii) overdose when treating raw water. In most water treatment plants, the minimal coagulant concentration is determined by the jar test technique which produces problems due to excess (or insufficient) coagulant, particularly during periods of fast variation in raw water quality [8]. In our previous work [9], we showed that iron salt can be used for the removal of humic substances but the residual content of iron has not been studied. This review will focus on (i) the consequence of human exposure to aluminum and iron and (ii) risks of metal deposit in pipe for potable water distribution system.

Aluminum

a. General aspects

Chemical coagulation by hydrolysing aluminum salts (monomer or pre-polymerised) is the major technique used around the world in water treatment [3-6]. European Legislation has established a maximum contaminant level at 200 μg/L for aluminum content in drinking water [10]. These values originate from safety reasons for aluminium.

According to the estimation of the American Waste Water Association (AWWA), drinking water (probably drinking water treated) provides about 5% of the aluminium ingested by humans. Lack of scientific knowledge does not allow a clear assessment of the risk to the general population for aluminum residual in drinking water.

b. Health problems

Aluminum residual in drinking water enter the human body through the gastrointestinal tract. Dissolved aluminum in drinking water is classified as highly reactive (due to inorganic complex) to non-reactive (bound to inorganic complexes). Aluminum is more dissolved in the stomach when the pH is extremely low. The absorption in the stomach may be a risk. Chronic exposure to aluminum (toxic form: Al^{3+}, $Al(OH)_2^{+}$, $Al(OH)^{2+}$) is associated with various health problems.

In the human organism aluminum interferes with essential metals (oligo elements) and metalloids by altering their bioavailability. In the human body, aluminum ions complete in reactions with metal ions such as zinc, iron, calcium and chromium [11]. Once absorbed, aluminum reaches the blood and circulates mainly bound to transferrin and citrate and can cross the blood-brain barrier [12]. Aluminum accumulation in the brain is proposed to be associated with neurodegenerative diseases, including Alzheimer's dementia, Parkinson's disease, amyotrophic lateral sclerosis, and dialysis encephalopathy [13]. Neurotoxicity of aluminum is not caused by a single alteration, but it is probably a result of adverse effects at multiple cellular levels [14].

The work of Sies and Jones [15] shows that high aluminum concentration cause oxidative stress. Evidence of an oxidative stress status has been found in association with most neurodegenerative disorder in which aluminum is present in relative high amount [14]. Aluminum negatively impacts neurotransmission, either by directly inhibiting the enzymes responsible or by affecting the physical properties of synaptic membranes [13].

The toxicity of aluminum depends on the form (total aluminum, total dissolve aluminum, monomer organic aluminum, monomer inorganic aluminum, polymeric aluminum) in which it occurs, while the mechanisms of its action depend on the range of tolerance of an organism to aluminum concentration [16].

c. Drinking water distribution networks

Degradation of drinking water quality in distribution networks represents a problem for water supply in urban area. These can be an elevated concentration of residual aluminum, increase of residual turbidity or increase in microbial numbers; all of witch affect taste, odor and color of drinking water. In the drinking water pipeline, residual aluminum can cause the formation of micro-flocs, sources of soft deposition in the distribution networks.

Soft deposits inside the pipeline are source of biofilm consisting mostly of bacteria, sources of water born disease, which can be present in drinking water [17]. These deposits are sources of microbial nutrients.

At the solid liquid interface, soft deposit can also affect the hydraulic of the pipeline. Aluminosilicates frequently deposit onto plumbing materials in distribution systems. Such solids were previously believed to provide some degree of corrosion protection to pipes, despite consumer complaints related to post precipitation [18].

Previous research demonstrated that aluminum and silica form complex solids of opalescent appearance that can pass through the treatment plant (slow deposition) and precipitate in the distribution system [18].

Iron

a. General aspects

Iron is essential for human health but is also a toxic metal. Iron can be found dissolved in water that we use for drinking. Its positive contribution to human health is much studied, but the information about its toxicity is missing.

European Legislation has established a maximum contaminant level at 200 μg/L for iron content in drinking water [10], and the World Health Organization has fixed a sanitary security limit for iron at 2 mg/L [19]. These values originated from aesthetic reasons for iron (coloration of water).

b. Health problems

High content of residual iron in drinking water may be responsible for (i) the neutralization of the disinfectants used to kill micro organisms, (ii) the coloration of water, (iii) metallic tastes of water.

Excessive ingested iron can cause excessive levels of iron in the blood because high iron levels can damage the cells of the gastrointestinal tract, preventing them from regulating iron absorption. Humans experience iron toxicity above 20 milligrams of iron for every kilogram of mass, and 60 milligrams per kilogram is a lethal dose [20].

Sullivan [21, 22] was the first to propose and continued to reiterate that iron levels play a major role in producing atherosclerosis. His major basis for this proposal was that menstruating women, who have a reduced iron load, have strong protection against atherosclerosis, compared to men in the same age group.

Ong and Halliwell [23] suggest that iron can be involved in Alzheimer's disease. The important mechanism is the interaction of iron and cholesterol in promoting oxidative damage, causing both atherosclerosis and neurodegeneration.

Increasing evidence indicates that excessive iron in selective regions of the brain may be involved in the neurodegenerative disorders [24].

c. Drinking water distribution networks

The residual iron in drinking water can react with disinfectants (neutralizing them) and that causes proliferation of micro-organisms in distribution networks. The residual iron in the treated water can hydrolyze and settle throughout the pipeline system. Soft deposition can cause coloration of water and sometimes obstruction of the drinking water network.

The presence of residual iron in drinking water can also cause proliferation ferruginous bacteria sources of soft deposit. Growth of ferruginous bacteria can reduce the diameter of the pipe or cause corrosion [25]. Ochre deposition can cause a serious hydraulic clogging problem in drinking water network. The problem arises when the iron, as soluble Fe(II), is oxidized to insoluble Fe(III) either biotically via bacteria, or abiotically [26]. Oxyhydroxides of iron formed in the pipeline may be in crystalline form or amorphous form. Dehydration of amorphous form gives crystalline form. The crystallization is a complex electrochemical and physico-chemical phenomenon between the network surface and water composition.

In oxygenated medium, the divalent iron (Fe^{2+}) and in slightly alkaline medium the trivalent iron (Fe^{3+}) take the form of ferric oxide Fe_2O_3 with rust-colored or ferric hydroxide $Fe(OH)_3$. These forms are insoluble. Ferric oxide crystallizes as $Fe_2O_3 \cdot 2H_2O$, or like $Fe_2O_3 \cdot H_2O$. The increase in the iron and turbidity in drinking water would be attributable to the release of soft deposits into bulk water [27].

Outline of the deposition in the drinking water distribution system

In general, treated tap water is transported from water plant to taps via distribution pipelines. Use of Al(III) or Fe(II, III) salts as coagulants in water treatment may lead to increased concentrations of aluminum and iron ions in treated water.

Where residual concentrations are high, aluminum iron (III) may be deposited in the distribution system. Disturbance of the deposits by change in flow rate may increase levels of aluminum and iron ions at the tap and lead to undesirable color and turbidity and health problems [28]. The deposition in water distribution networks is a complex electrochemical and physicochemical phenomenon between a metal surface and germ of crystallization. The deposition in the pipe depends on chemical factors, such as pH, alkalinity, dissolving residual coagulant (aluminum and iron (II, III)), and physical properties, such as flow and velocity, as well as the pipe material [29].

In drinking water, we must distinguish the key elements that include the following chemical species: H_3O^+, HO^-, Ca^{2+}, HCO_3^-, CO_3^{2-} and characteristic elements: Mg^{2+}, Na^+, K^+, Cl^-, SO_4^{2-}, NO_3^- [30]. The concentrations of basic elements are linked by the equilibrium relationships from the law of mass action.

Total hardness of water is generally the concentration of dissolved salts of calcium and magnesium. The temporary hardness corresponds to calcium and magnesium binding to bicarbonate. The equations of the reaction can be represented as follows:

$$Ca_{(aq)}^{2+} + CO_{2(aq)} + H_2O \rightleftarrows Ca(HCO_3)_2$$

$$Mg_{(aq)}^{2+} + CO_{2(aq)} + H_2O \rightleftarrows Mg(HCO_3)_2$$

Calcium and magnesium dicarbonate, soluble, is in equilibrium with CO_2 and calcium carbonate, slightly soluble. The equations of the reaction can be represented as follows [31]:

$$Ca(HCO_3)_2 \rightleftarrows \underline{CaCO_3} \downarrow + CO_{2(aq)} + H_2O$$

$$Mg(HCO_3)_2 \rightleftarrows \underline{MgCO_3} \downarrow + CO_{2(aq)} + H_2O$$

Water deposition can be controlled by water quality parameters, such as pH, alkalinity and calcium hardness, which can induce the formation of calcium carbonate compounds in the internal wall of water pipe. The precipitation of calcium carbonate in a drinking water network is mainly due to the appearance of two phases: germination and growth. Germination involves the emergence of a stable solid phase in a solution in which was initially lacking [31]. The germ formed will grow by successive ions deposition. The germ of calcium carbonate can occur in the solid surface consisting of another compound, as is the case of iron pipes of water distribution networks in some countries. Soluble Fe(II,III) and Al(III) in water can be hydrolysis, and the presence of calcium carbonate deposition in drinking water distribution systems is the germ of crystallization of Fe(II,III) and Al(III) oxyhydroxides. Such solids deposited on the internal surface of water distribution system were believed to reduce the diameter of the pipe flow, leading to consumers complains related to postprecipitation [18]. The simplified equation for the formation of aluminum or iron deposition in drinking water distribution systems can be represented as follows [32]:

- Case of residual aluminum in water

$$2Al(OH)_3 \rightleftarrows 2AlO(OH) \rightleftarrows \underline{Al_2O_3} \downarrow + H_2O$$

- Case of residual iron(III) in water

$$2Fe(OH)_3 \rightleftarrows 2FeO(OH) \rightleftarrows \underline{Fe_2O_3} \downarrow + H_2O$$

Fig. 1 represents a schematic illustration of the phenomenon of Al(III) and Fe(II, III) deposition in drinking water network. In the case of water with high hardness the layer of calcium and magnesium carbonate deposited is thick and over it is formed in time another layer of calcium and magnesium carbonate mixed with Al(III) and Fe(II, III) oxhyhydroxide. In the case of water with low hardness there is just a thin film of calcium and magnesium carbonate, and the second layer consists in Al(III) and Fe(II,III) oxhydroxide as the main coumpound.

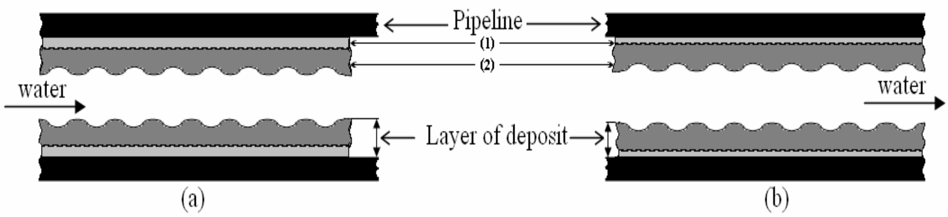

Fig. 1. Illustration of the phenomenon of aluminum or iron deposition in pipeline:
a. Case of water with high hardness, b. Case of water with low hardness,
(1) - Layer of calcium and magnesium carbonate deposit ,
(2) - Calcium and magnesium carbonate deposit/Oxyhydroxyde of Al(III) and Fe(II, III) deposit

Conclusions

In the light of progress of research on the effects of aluminum and under the terms of the precaution principle, water treatment plants using aluminum or iron - based coagulants should optimize their operations to minimize levels residual aluminum and/or iron in treated water. Aluminum has no known beneficial effect in humans. Many scientific articles are available, covering various aspects aluminum toxicity like the pathophysiology of neurodegenerative disorders (amyotrophic lateral sclerosis, Parkinson dementia, Alzheimer's disease). The high residual aluminum concentrations (above 0.4 mg/L) in some water can lead to the filing in the distribution network of gelatinous substance containing aluminum, which reduces flow in the network and alters the water quality.

The presence of residual iron in drinking water is not indicated for a number of reasons: (i) excessive iron in selective regions of the brain may be involved in the neurodegenerative disorders, (ii) ochre deposition can cause a serious hydraulic clogging problem in drinking water network.

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Funding Body

We thank AUF «EUGEN IONESCO» for granting a fellowship to J.M. Siéliéchi.

Received: July 11, 2010
Accepted: August 2, 2010