Effects of Aluminum in the Sludge of Sayedabad Water Treatment Plant on Environment

by

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It is hereby declared that this thesis or any part of it has not been submitted elsewhere for the award of any degree or diploma.

Fahmida Hoque Khan

Dedicated

To

My Beloved Parents

ABSTRACT

Sayedabad water treatment plant was established in 2002 to serve water demand of Dhaka city. Large scale use of aluminum in coagulation process during water treatment in Sayedabad Water Treatment Plant may generate significant quantities of aluminum rich treatment sludge and disposal of this sludge may lead to environmental pollution. The common practice is open water disposal or on land disposal. Aluminum leaching from such sludge may pose potential threat to both groundwater and surface water with long-term consequences. In this study, water quality in supernatant of excess alum sludge in receiving water bodies have been assessed. Besides, the leaching characteristics of aluminum sludge have been evaluated with a view to assessing the effects of aluminum sludge on environment. This study has also attempted to assess the uptake of aluminum from sludge by red amaranth plants.

The water quality parameters suggest that there is no significant impact on surface water due to disposal of supernatant of alum sludge. The spatial variation of Al concentration indicates there is little variation of aluminum (Al) concentration distribution at varying distances from the point of discharge.

The leachate concentration can be significant enough to percolate through alum sludge into groundwater. This percolated leachate concentration might raise the concentration of Al in groundwater above the allowable limit as suggested by World Health Organization guideline value and Bangladesh standard for drinking water.

The leachate from column leaching test indicates that distilled water and chloride have the highest tendency to leach Al from sludge. Significant amount of Al could be leached from alum sludge by distilled water, chloride, sulfate, nitrate and hydroxide anions. The leachate from chloride anion fluid media is higher than distilled water, nitrate, sulfate and hydroxide anion fluid media.

Aluminum uptake from sludge by red amaranth plants has been studied and it has been found that the red amaranth plants accumulate Al from soil and the concentration varies among the different parts of plants. Roots of red amaranth accumulated the highest concentration followed by stems and leaves. The quantity of accumulated Al decreases from roots to leaves gradually. The mean Al concentration in leaves is 2.4 times less than stem and 4.1 times less than root. The Al concentration of red amaranth plants grown in Al sludge was 451 ppm, which is about 3 times higher than the Al concentration of red amaranth plants grown in Al sludge free soil.

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CHAPTER 1

INTRODUCTION

1.1 Background

Aluminum makes up around 8 per cent of the Earth's surface, making it the third most common element. It is often used in cooking utensils, containers, appliances and building materials, as well as in the production of glass, paints, rubber and ceramics. Aluminum is used in several forms, such as aluminum hydroxide (in antacids), aluminum chlorohydrate (in deodorants), and the most common form, aluminum sulfate (in treating drinking water).

Micro-organisms present in drinking water include viruses, bacteria (e.g., E. coli), and protozoa (e.g., Cryptosporidium and the beaver fever causing organism, Giardia). At low levels, these organisms can cause sickness and disease (including severe diarrhoea) and are generally very difficult to remove from water. The parasites Giardia and Cryptosporidium are very resistant to most types of disinfection, including chlorination. Water treatment with aluminum sulfate is, however, effective at removing these parasites when used in a chemical treatment process called coagulation.

More than 90 per cent of our daily intake of aluminum comes from food, but this aluminum appears to be bound to other substances in the food and cannot be absorbed by the blood stream. In contrast, aluminum in water can be absorbed by humans because after water treatment, the aluminum is largely in an unbound form. Even so, the amount of aluminum absorbed from drinking water is usually very small. At low levels, aluminum in food, air, and water is not likely harmful to human health. However, at high concentrations there is evidence linking aluminum to effects on the nervous system, with possible connections to several diseases, such as Parkinson's, Alzheimer's and Lou Gehrig's disease. Patients suffering from these diseases tend to have high levels of aluminum in some areas of their brains. It is not known if aluminum is causing these diseases or if the aluminum starts accumulating in people that already have the diseases. There is also some concern that aluminum may cause skeletal problems. There is no evidence to suggest that aluminum affects reproduction, or that it causes cancer.

Recommendations for aluminum levels in drinking water have been made by several organizations and agencies, such as the American Water Works Association (AWWA) and the U.S. Environmental Protection Agency (USEPA). The AWWA recommends that concentrations of aluminum in drinking water should not exceed 0.05 parts per million (0.05 ppm or mg/L) and the USEPA recommends that the level not exceed 0.2 ppm. Future health guidelines are likely to centre on the amount of free aluminum rather than total aluminum, which includes aluminum that is likely not taken up by the human body.

Raw water is contaminated with many impurities that must be removed before it is safe to drink. A particular problem is the suspended solids - usually of inorganic clays and organic matter. The most common treatment method is based on the use of alum (aluminum sulfate, $Al_2(SO_4)_3$) to coagulate the suspended solids particles. This resultant sludge, separated from the cleaned water, contains around 0.1-0.5 per cent solids and requires further treatment before disposal. There are many options for this, but one of the most common is to further concentrate the sludge using thickeners and settling ponds, sometimes followed by centrifuges or filters. From there the material is often transported to a landfill site for disposal. The organic matter and fine clay particles in alum sludge are valuable resource that can be reclaimed and reused for a beneficial purpose. Disposal is a costly and wasteful way of dealing with alum sludge. Alum sludge is a desirable material for use in a soil blend.

1.2 Scope of the Study

Sayedabad Water Treatment Plant (SWTP) was established in 2002 to serve water demand of Dhaka city, from which 22.5 crore liter of water is supplied daily. After treatment, aluminum sludge is discharged into sludge drying beds. When these beds are over flown with sludge water, the excess sludge water is discharged into the nearby lake. Solid sludge, after filling up those sludge drying beds will then be dried into another site which may create hazards both in soil and crop. In this study, leaching characteristics of aluminum sludge collected from SWTP will be evaluated with a view to assessing its environmental quality. Besides, phyto-toxicity due to increased aluminum in soil/water and its impact on agricultural field is another major concern. This study focuses on the effects of aluminum sludge on environment.

Exposure to aluminum can come from food, air and water. Although everybody is exposed to it to some degree, aluminum is not a necessary substance for humans and too much of it may be harmful to your health. Most of daily aluminum intake is from food and water. The air, on the other hand, represents a relatively small portion of that daily intake. Aluminum in water is in a form that is more readily absorbed by the body and very high aluminum levels in water can be of concern. Aluminum is used in water treatment to remove disease-causing micro-organisms and other drinking water impurities that can affect health.

Excess aluminum is toxic to human body. Alum sludge if discharged without any treatment may easily be transmitted to plants through osmosis and then to human body as it consumes vegetables which may contain aluminum. The aluminum sludge of SWTP is discharged to the surrounding environment without any treatment. Vegetables are important food crops of Bangladesh and are rich in vitamins and minerals which are very essential for maintaining good health. As humans consume vegetables so it must be ensured that the vegetables are free from aluminum. The discharge of aluminum sludge into the environment must be carefully controlled and minimized. But it is unknown how much aluminum is transmitted to plants if discharged without any treatment. Therefore, it is of utmost importance to know the aluminum content in vegetables. With this view in mind, this study is also intended to find out the level of aluminum transmission from sludge to vegetables.

1.3 Aims and Objectives

The aim of the study is to find out the effects of aluminum sludge on environment with the following major objectives:

i) To assess the concentration of aluminum sludge in receiving water bodies

ii) To estimate the concentration of leaching aluminum in sludge through TCLP

iii) To determine the leachable aluminum from sludge through column leaching representing the natural leaching environment

iv) To assess uptake of aluminum from sludge by plants

1.4 Organization of the Thesis

The thesis consists of six chapters. Apart from this chapter, the remainder of the thesis has been divided into five chapters. Chapter 2 titled "Review of Literature" includes brief description of relevant literature regarding impacts of aluminum, theory of alum coagulation, reviews of toxicity of aluminum and different coagulant aids.

Chapter 3 titled "Overview of Sayedabad Water Treatment Plant" highlights about different steps of treatment processes in Sayedabad Water Treatment Plant.

Chapter 4 titled "Research Methodology" discusses in detail the methodology adopted in this work. Sample collection and physical and chemical characteristics of alum sludge are discussed. It also highlights about standard laboratory method of determining leachates, preparation and methods of analysis of red amaranth plants grown in with and without sludge.

Chapter 5 titles "Effects of Alum Sludge on Environment" presents analysis of water quality parameters, leachability of alum in alum sludge through TCLP and column leaching transmission of alum from sludge to red amaranth plants.

Finally Chapter 6 titled "Conclusions and Recommendations for Further Studies" provides a summary of the findings and recommendations for further studies in this field.

CHAPTER 2

REVIEW OF LITERATURE

2.1 Introduction

Alum is used in the drinking water treatment process to destabilize colloids for subsequent flocculation and water clarification. Alum sludge is the water treatment residual from this process and is considered to be a waste material. Varying amounts of aluminum are present naturally in groundwater and surface water, including those used as sources of drinking water. Because aluminum is ubiquitous in the environment and is used in a variety of products and processes, daily exposure of the general population to aluminum is inevitable. Many studies have been conducted on the various aspects of alum sludge e.g., use of alum as coagulant, disposal of alum sludge, alum phytotoxicity in aquatic and agronomic system, etc. A brief summary of the previous studies is presented below.

2.2 Alum as Coagulant

Aluminum salt or alum has been employed as a coagulant for water treatment since Roman times and today it is the most widely used coagulant. Alum is generally effective within the pH limits of 5.5 to 8.0 (Hammer, 1977). The dosages of aluminum used in water treatment are in the range of 5 to 50 mg/l (Hammer, 1977). Organic materials found in natural waters are, for the most part, humic substances, i.e., humic acid, fulvic acid. Humic substances are the cause of the yellow or brown color imparted to waters with high organic content. Organics, in the from humic substances, are undesirable in a potential water supply for a number of reasons, ranging from aesthetics to being the precursors of potentially carcinogenic trihalomethanes. Coagulation with aluminum sulfate, is an effective method for removing organic color which occur in the pH range of 5 to 6.5 (Edwards and Amirtharajah, 1985). Miller (1925) stated that in the coagulation of humic substances with aluminum, the aluminum ion is the active coagulant and in fact, since that time, the water industry has tended to use aluminum as a means of removing color. Hossain (1996) studied the mechanism of coagulation of colored water with aluminum sulfate. He found that the mechanism of coagulation depends on pH. At pH 4.5, the mechanism of coagulation is adsorption and charge neutralization (ACN) whereas at higher pH, the mechanism is different from ACN.

2.3 Disposal of Alum Sludge

Alum sludge is the water treatment residual (Al-WTR) from this process and is considered to be a waste material. Biosolids (sewage sludge) are a by-product of the wastewater treatment process. The disposal of Al-WTR alone would be beneficial to soils high in P, since the Al-WTR can adsorb soluble P. Likewise, the co-application of Al-WTR and biosolids may be advantageous to municipalities as a means of disposing of high P bearing biosolids in an environmentally sound manner. Because of Al-WTR's ability to adsorb P, Al-WTR could play a role in the removal of P in sewage treatment plant effluent (Ippolito et al., 1999).

Sludge is a valuable source of plant macro and micro nutrients (N, P, Cu, Fe, Mn, Zn) and organic matter, but it also contains heavy metals (Cd, Cr, Ni, Pb) that are potentially hazardous. Leaching of heavy metals is a concern because some metals accumulate in the soil, thus becoming toxic to plants and humans. Land application of biosolids (sewage sludge) can significantly increase heavy metal concentrations in agricultural soils (Sloan et al., 1998). For soil management and water quality purposes, it is important to determine the long-term fate of biosolid-applied heavy metals. Most metals in water treatment sludges occur predominantly in weakly mobile, non-bioavailable forms (Elliott et al., 1990). The initial leaching of heavy metals is attributed to their soluble or exchangeable forms and to the subsequent slow leaching to the solid compounds.

2.4 Aluminum Phytotoxicity in Aquatic and Agronomic Systems

pH of soil is an indicator of the relative availability of nutrients. Low pH of soil stress is a major growth limitation to crop production in many regions. At low pH, it is not often the H^+ ion activity that limits growth, but the toxicity and/or deficiency of other elements. On the basis of existing literature, Al toxicity is one of the yield-limiting factors that have been identified in acid soils. It is very difficult to determine the direct effects of H^+ ion toxicity on plant growth in acid soils because of the changing interrelationships that occur between pH and Al concentrations in the soil solution, and the changing availability of essential nutrients (Fageria et al., 1990). Aluminum concentration can be sufficiently high in acid soils with pH values of 5.5 or below to be toxic to plants. The aluminum species which are responsible for the phytotoxic effect appear to be a small fraction of the total aluminum in the soil solution (Sawhney et al., 1996). Average aluminum concentration in natural soil is 71,000 ppm and common range lies between 10,000-30,000 ppm (USEPA, 1983).

2.5 Toxicity of Alum Sludge

Toxicity of aluminum sulfate, alum, appears to be dependent on pH levels, and with other compounds, concentrations of alum may have an effect at many levels. Each mg/L of alum reacts with 0.5 mg/L of total alkalinity and reduces pH (Boyd, 1979). A toxicity study at 10 water treatment plants found algae growth was limited by alum sludge (George et al., 1995). In the algae assay, 48% of the samples inhibited growth in pH level between 5 and 9 and monomeric aluminum (mAl) concentrations between < 0.04 and 11.8 mg/L. When mAl was < 0.04 mg/L, there was an inhibitory effect 33% of the time. Hall and Hall (1989) also examined the effects of alum on daphnia and fathead minnows. They observed that mortality was the greatest when aqueous aluminum levels were the highest.

Studies with juvenile striped bass indicate that this species is extremely sensitive to several forms of aqueous aluminum (Driscoll et al., 1980; Palawski et al., 1985;

Skogheim and Rosseland, 1986; Rosseland et al., 1992). An in situ study with larval striped bass found 90 to 99% mortality in river water with 480 to 4100 mg/l Al and pH levels between 6.0 and 6.8 (Hall et al., 1985). Klauda et al. (1989) reported that 15 mg/l of monomeric aluminum (mAl) was the critical environmental value to protect larval river herring at pH levels below 6.2. A later study supported the theory that mAl, the inorganic fraction, was potentially the most toxic to early life stages of migratory fish (Hall et al., 1993).

Studies are now showing that variability of physical environmental conditions may play a role in aluminum toxicity. Polymers created from aluminum and water collect on gills and limit respiration (Oughton, 1992). Chemical changes through the polymerization process occur when waters with different pH, temperature, and ionic strength are mixed, or when waste water is discharged into a river system (Witters et al., 1996). The ability for Al polymerization to occur in natural water, as a function of increasing pH, was described by Driscoll and Schecher in 1988. In the laboratory, Witters et al. (1996) found that when pH was increased from 4.6 to 6.4, polymerization was increased because of the high molecular weight of the total aluminum.

The form in which aluminum appears varies with pH so its toxicity is also pH dependant. This is acknowledged by ANZECC (1992) which includes a water quality guideline for protection of aquatic ecosystems for total aluminum of 0.1 mg/l Al at pH >6.5, and < 0.005 mg/l Al for water with pH <6.5. Discharge of aluminum-rich sludge from filter backwashes has been associated with fish mortalities. In such streams concentrations of 1 mg/l Al and above may be necessary before fish mortalities result from short-duration exposure under basic conditions.

The toxicity of aluminum at low pH is complex, and interpretation is not assisted by the variety of fish species used in determining its toxicity. The toxic effects observed have varied with pH, species, life-stage, hardness and composition of the test media. These variations result from the complex chemistry of aluminum over a range of low pH values.

Physiological differences between test species may also interact with the aluminum present, thus accentuating or reducing its toxicity.

Aluminum forms complexes readily with organic matter which can modify its toxicity. For example, trout lived for 10 days in water at pH 4.7 with 0.18 mg/l Al plus humic substances (74-80 per cent of the Al was organically bound), whereas trout died within 2-3 days in the absence of humic substances – 98 per cent of the Al was inorganic (Wilters et al., 1990).

Generally it is considered that aluminum is more toxic to fish than to smaller animals. It is believed that aluminum coagulates the mucous on their gills, causing osmoregulatory and respiratory problems. Although aluminum may not be toxic to some plankton and small invertebrates, these species can be coagulated with alum and be removed from the water column in slow moving waters. The USEPA (1988) has developed an ambient water quality criterion for aluminum requiring that the instream and soluble aluminum level not exceed 0.087 mg/l on a 4-day average.

Wang et al. (1998) studied the effects of alum-treated waste water sludge on barley growth. Alum sludge derived from a municipal wastewater plant was used as a soil amendment in a greenhouse study with barley (Hordeum Vulgare) as the test crop. Barley growth decreased as the Al^{3+} activity in the sludged soil solution increased, but for a given Al^{3+} the phytotoxicity of Al was markedly pH dependent.

2.6 Aluminum Coagulants

2.6.1 Toxic Action

Aluminum coagulants contain high concentrations of ionic aluminum, the toxic form. Toxicity is very dependant on pH and increases at lower pH. At high pH, most aluminum is present in solid form and is not bio-available. Below pH 6 it is mostly in the dissolved, bio-available form. The bioavailability and toxicity of aluminum is generally greatest in acid solutions and generally most toxic over the pH range 4.4 -5.4 with a maximum toxicity around 5.0-5.2 (ANZECC, 2000). However, at pH of 6.5 to 8.0, which is the normal range for natural waters, there is generally considered to be little threat of toxicity. Where pH of natural waters is outside this range, instream values including fish and invertebrate diversity would tend to be limited in any event, and environmental sensitivity reduced.

In fish the toxic effect is manifest as an accumulation of mucous on gill surfaces, which impairs their function. Aluminum has been implicated in fish mortalities in acidified waters (Baker, 1982). A review by Spry and Weiner (1991) concluded that in low-pH water (i.e. 6-6.5 or less): "both sub-lethal and lethal toxicity of aluminum has been clearly demonstrated in both laboratory and field studies at environmental concentrations". Complexing agents including humic substances reduce the bioavailability of aluminum to organisms resulting in lower toxicity. Larcombe (1999) reported that even at doses in excess of requirements, the dissolved toxic aluminum is reduced in the receiving environment very rapidly to a very low concentration with no serious toxicity implications.

2.6.2 Bioaccumulation

Conflicting results have been reported on the effect of pH and uptake of aluminum, and hence its bioavailability, in freshwater organisms (ANZECC, 2000). Under a normal pH range of 6.5 to 9 for freshwaters and 6.5 to 8 for marine waters, aluminum can be considered to carry a low risk of bioaccumulation. Runoff from acid soils will tend to enhance bioaccumulation risk if the effect is to reduce pH to below 6.5. Conversely, runoff from alkaline soils will tend to increase pH and thereby reduce such risk. However, in addition to mediating influences such as DOM, other factors need to be taken into account on a site specific basis.

2.6.3 Persistence

The biogeochemical cycle of aluminum is complex, yet poorly understood (ANZECC 2000). Research into effects of alum sludge discharged from wastewater treatment plants indicates that dissolved aluminum can be released from sludge under highly acidic conditions (George et. al., 1991). However overseas research indicates that alum sludge is stable under normal pH conditions and pollutants have little or no affinity for release. Furthermore, aluminum is reported to be tightly bound in alum treated sediment under both reduced and oxidized conditions and at pH ranges between 5-7 (Larcombe, 1999). Larcombe (1999) reports that alum floc is not toxic to benthic organisms and small planktonic crustaceans present in sediment pond water showed no toxicity during coagulation and settlement periods. Aluminum toxicity is reduced by calcium and dissolved organic carbon. The use of lime for soil stabilisation would have the effect of increasing water pH and thereby decrease aluminum toxicity. There is also evidence of acclimation (i.e. increase in resistance or tolerance) of fish to aluminum (Spry and Weiner, 1991). Fish in waters with high background concentrations of aluminum may therefore have enhanced resistance to episodic increases.

2.7 Sanitary Significance of Al

2.7.1 Exposure

Because aluminum is ubiquitous in the environment and is used in a variety of products and processes, daily exposure of the general population to aluminum is inevitable. Varying amounts of aluminum are present naturally in groundwater and surface water, including those used as sources of drinking water. Miller et al. (1984) reported that aluminum is more likely to exist in surface water than in groundwater; only 9% of groundwaters had detectable amounts of aluminum (detection limit 0.014 mg/L), whereas 78% of surface waters had detectable aluminum.

Concentrations of aluminum in food range widely (means range from <0.001 to 69.5 mg/100 g), depending on the nature of the foodstuffs (Pennington, 1988). The highest levels are found in nuts, grains and dairy products, particularly processed cheeses. There is also potential for exposure from the ingestion of aluminum contained in over-the-counter drugs, including antacids (Graves et al., 1990) and buffered acetylsalicylic acid (aspirin); based on the recommended dose, the range of aluminum exposure from antacids has been given as 840–5000 mg/d (Lione, 1985) and as 120–7200 mg/d (Nieboer et al., 1995) and that from buffered aspirin has been given as 126–728 mg/d (Lione, 1985) and as 200–1000 mg/d (Nieboer et al., 1995). Aluminum leaching from cooking utensils, containers and packaging made of aluminum may also contribute to dietary exposure (Lione et al., 1984).

The total intake of aluminum from all food sources (excluding over-the-counter drugs) for an adult is estimated to be 6 mg/d in the United Kingdom (Ministry of Agriculture, Fisheries and Food, 1985) and 8–9 and 7 mg/d (adult men and women, respectively) in the United States (Pennington et al., 1995) although higher daily intakes have been estimated (Greger, 1993). Estimates of aluminum intakes ranged from 0.7 mg/d for six- to 11-month-old infants to 11.5 mg/d for 14- to 16-year-old males (Pennington et al., 1995).

Assuming a daily contribution of 8 mg (average of 7–9 mg/d) from food, 0.0042 mg (maximum daily intake in Ontario) from air and 0.26 mg (global mean level 0.17 mg/L, daily intake 1.5 L) from water, an adult would take in about 8.26 mg of aluminum per day. In other words, approximately 97 per cent of the normal daily intake for an adult is from food and the remainder is from drinking water; the contribution from ambient air is insignificant.

2.7.2 Health Considerations

Gardner and Gunn (1995) reported increased levels of aluminum in urine after tea drinking; Other investigators have confirmed the low bioavailability of aluminum in tea

(Forster et al., 1995). Although drinking tea with milk or lemon juice over a short period does not contribute significantly to the total aluminum burden (Butterworth et al., 1992) absorption of aluminum in heavy tea drinkers, particularly those with enhanced absorption, may not be insignificant because of the relatively high aluminum content of tea (Nieboer et al., 1995).

In the human diet, citric acid may be the most important factor determining the absorption of aluminum. Several studies have found that the presence of citrate in food or beverages significantly increases the absorption of aluminum from dietary sources (Greger and Powers, 1992).

The highest levels of aluminum in mammalian tissues are found in the skeleton, lungs, kidneys, spleen, thyroid and parathyroid glands. Experience with dialysis patients has shown that aluminum has the potential to accumulate in the skeleton and brain (Crapper et al., 1980). The normal blood aluminum levels in humans are reported to be between about 1 and 16 μg/L (Weberg and Berstad, 1986).

In the brain, aluminum levels increase with age, and the highest levels of aluminum are found in the grey matter. Even in persons with normal renal function, the ingestion of aluminum-containing antacids can cause an elevation of the brain aluminum levels from 0.6 μg/g wet weight to 1.1 μg/g wet weight (Zumkley et al., 1987). Dollinger et al. (1986) found high levels of aluminum in the brains (1.05 μg/g wet weight or 5.25 μg/g dry weight) of 10 patients who were given 70 mL of a high-aluminum-content antacid per day (dose not reported) for 10 days, compared with 10 patients (aluminum in brain 0.412 μg/g wet weight or 2.60 μg/g dry weight) who were given an equal amount of low-aluminum-content antacid for 10 days.

In humans, absorbed aluminum is excreted from the body via the kidneys (Alfrey, 1986). In individuals with healthy kidneys, any aluminum absorbed is eliminated from the body before deleterious effects can occur. In patients with kidney dysfunction or in normal persons under high aluminum load, the buildup of aluminum can lead to toxic effects (Sedman et al., 1984).

2.7.3 Toxicity in Humans

On acute exposure, aluminum is of low toxicity. In humans, oral doses up to 7200 mg/d (100 mg/kg bw per day) are routinely tolerated without any signs of harmful short-term effects. However, two healthy individuals who drank water accidentally contaminated with an aluminum sulfate solution experienced ulceration of the lips and mouth (Eastwood et al., 1984). Intake of large amounts of aluminum can lead to a wide range of toxic effects, including microcytic anaemia (Parkinson et al., 1995) osteomalacia (Alfrey et al., 1986)' glucose intolerance of uraemia (Banks et al., 1987) and cardiac arrest (Starkey, 1987) Elderly persons with elevated serum aluminum levels exhibit impaired complex visual-motor co-ordination and poor long-term memory (Bowdler et al, 1979).

Patients with dialysis dementia were shown to have markedly elevated serum aluminum levels with increased concentrations in many tissues, including the cerebral cortex (Alfrey et al., 1976) Investigators reported a correlation between the aluminum concentration in water used to prepare the dialysate fluid and the incidence of dialysis dementia (Savory and Wills, 1984).

Aluminum has also been suggested as having a causal role in the onset of Alzheimer's disease (AD). Memory lapses, disorientation, confusion and frequent depression are the first recognizable symptoms that mark the beginning of progressive mental deterioration in patients with AD. Numerous other causes have been suggested for AD.

Crapper et al. (1986) found that the average aluminum content of control human brains (1.9 ± 0.7 mg/kg dry weight) was less than that of AD-affected brains (3.8 mg/kg dry weight), and Xu et al. (1992) reported small but significant increases of aluminum in brain tissues of AD patients compared with age-matched controls.

2.8 Summary

Alum sludge is the water treatment residual from flocculation and water clarification. From the above review of literature, it is seen that many studies have been conducted on the various aspects of alum sludge e.g., use of alum as coagulant, disposal of alum sludge, alum phytotoxicity in aquatic and agronomic system, sanitary significance of alum etc. This study is intended to focus on the effects of aluminum sludge on environment with a view to assess the concentration of aluminum sludge in receiving water bodies, to estimate the concentration of leaching aluminum in sludge through TCLP, to determine the leachable aluminum from sludge through column leaching representing the natural leaching environment and to assess the uptake of aluminum from sludge by plants.

CHAPTER 3

OVERVIEW OF SAYEDABAD WATER TREATMENT PLANT

3.1 Introduction

Dhaka Sayedabad Potable Water Treatment Plant (PWTP) is a project financed by French government and the World Bank to provide an adequate supply of clean water to the citizens of Dhaka - a megapolis of some 10 million people which is growing at the fast rate of 6% or more per annum.

3.2 Overview of Sayedabad Water Treatment Plant

The construction of Sayedabad PWTP started in 1999 and has been successfully completed in June 2002. The plant is designed for a nominal daily production of nearly 230,000 m^3 per day. This is the largest surface water treatment plant in Bangladesh. This plant is the first phase of a global project which is expected to produce 920,000 m^3 per day. The expected phase II will produce another 230,000 m^3 per day and should be started shortly with an expected completion within 3 years.

3.3 Raw Water Supply and Network

The Sayedabad Water Treatment Plant is treating the water of Sitalakhya River which is flowing from north to south on the eastern limit of Dhaka City. A pumping station located on the river side in Sarulia is lifting the water level to an elevation of 5 meters. From that point the raw water is flowing gravitationally to the Sayedabad WTP. The raw water is traveling on total distance of nearly 8.2 km, from Sarulia to Sayedabad, through a network of culverts and canal.

3.4 Raw Water Intake and Network

The raw water intake and network is shown in Fig. 3.1. The raw water comes from Shitalakhya river (Fig. 3.2). It is them pumped and transported to Dhaka-Narayanganj-Demra (DND) canal through a culvert. The pumping station (Fig. 3.3) is situated in Sarulina near the confluence of the Sitalakhya and Balu river. Four vertical pumps are used for the purpose (Fig. 3.4). The raw water is then transported to Sayedabd water treatment plant through DND canal (Fig. 3.5).

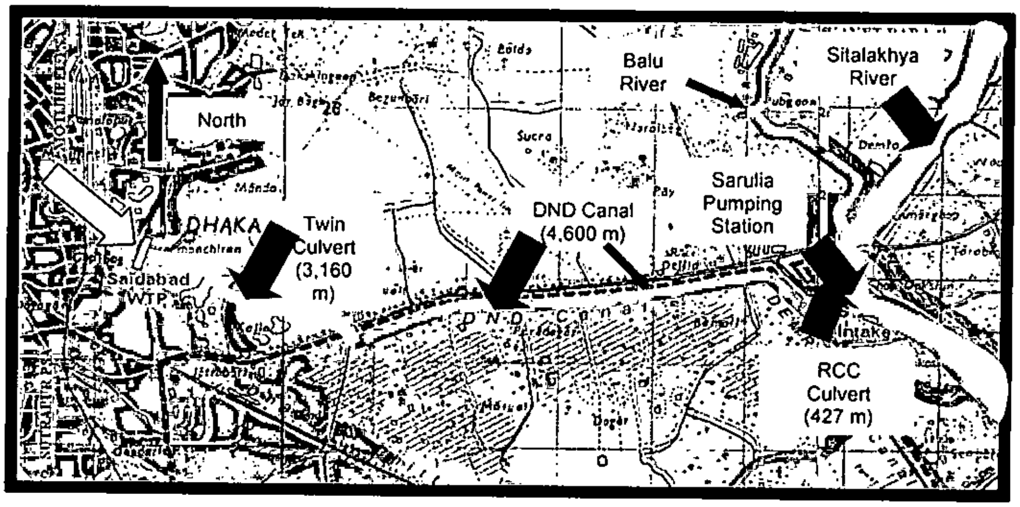

Figure 3.1 Raw water intake and network.

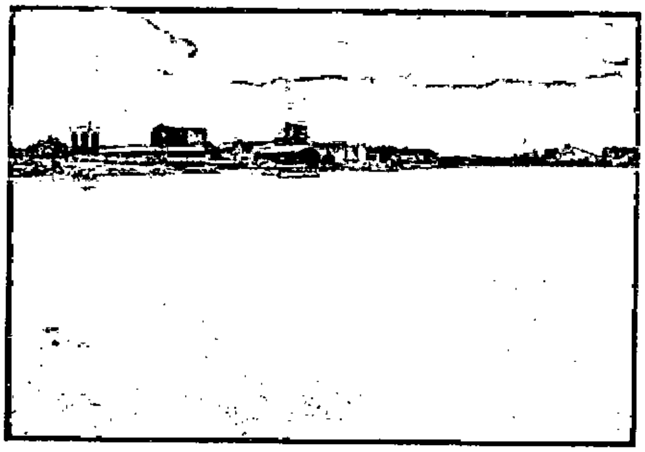

Figure 3.2 Sitalakhya river.

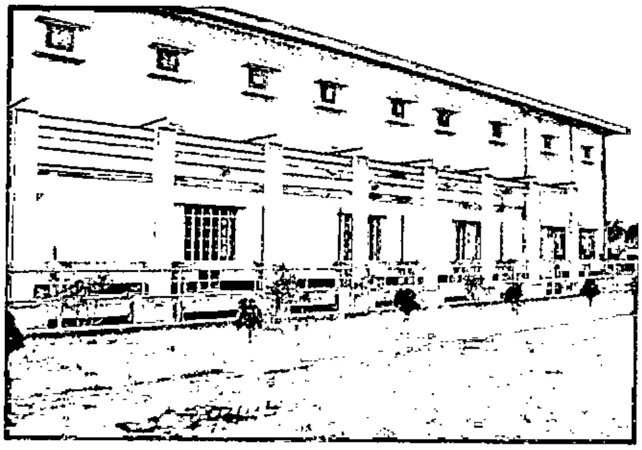

Figure 3.3 Sarulia pumping station.

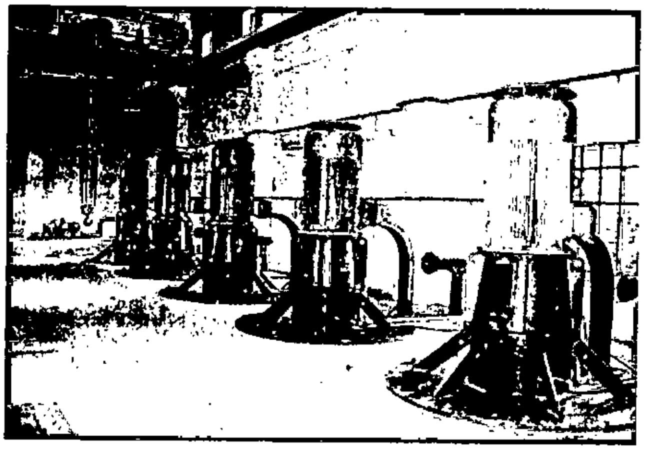

Figure 3.4 Vertical pumps.

Figure 3.5 DND canal.

3.5 Layout of Sayedabad Water Treatment Plant

The layout of Sayeadabad water treatment plant is shown in Fig. 3.6. The different steps which are followed in treating the raw water are explained below.

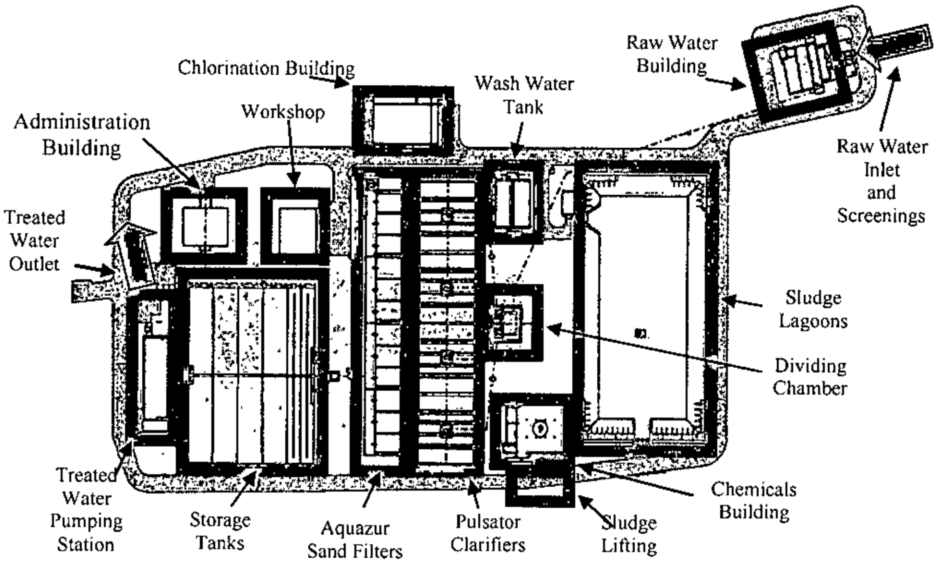

Figure 3.6 Layout of Sayedabad water treatment plant.

3.6 Different Steps of Sayedabad Water Treatment Plant

3.6.1 Step One: Raw Water Screening

Two independent inlet culverts (Fig. 3. 7) with enough capacity for the actual phase I and for the next phase II are used to transport raw water of DND canal to the treatment plant. Stationary screens are installed to remove larger size trashes and to protect the downstream equipment.

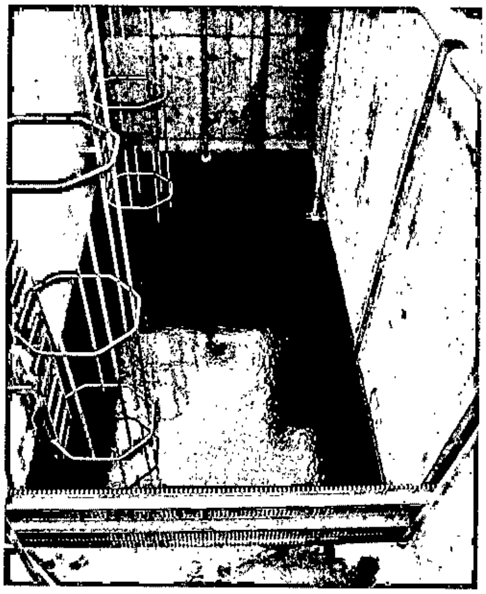

Figure 3.7 Inlet culvert.

3.6.2 Step Two: Raw Water Pumping Station

Five vertical pumps are available for pumping raw water (Fig. 3.8). At maximum production 3 pumps are running simultaneously, 2 are on standby. The pumps are lifting the raw water to a level of 10 meters into an outlet tank. After this step the flow downstream the process is fully gravitational. There is a pre-chlorination injection in the outlet pipe of the raw water building.

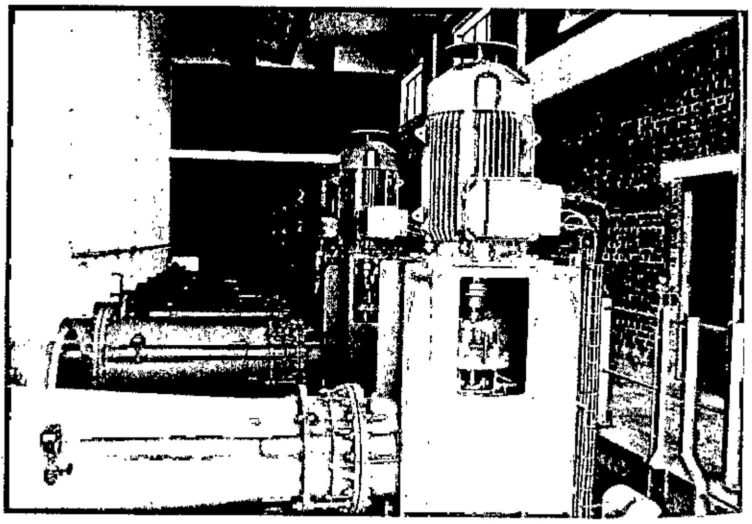

Figure 3.8 Pumping station for raw water.

3.6.3 Step Three: Dividing Chamber

The water is then flown gravitationally from the raw water building up to the dividing chamber (Fig. 3.9). The diameter of the pipe is 1500 mm. The dividing chamber is the starting point of the process. This is the injection point of lime which is used to raise the pH of water and also the injection point of aluminum sulfate which is used for the coagulation-floculation of particles in the water. Successive water falls are insuring the mixing of chemicals and the aeration of the water. An ultrasonic flowmeter is continuously measuring the raw water flow sent to the downstream process and equipment.

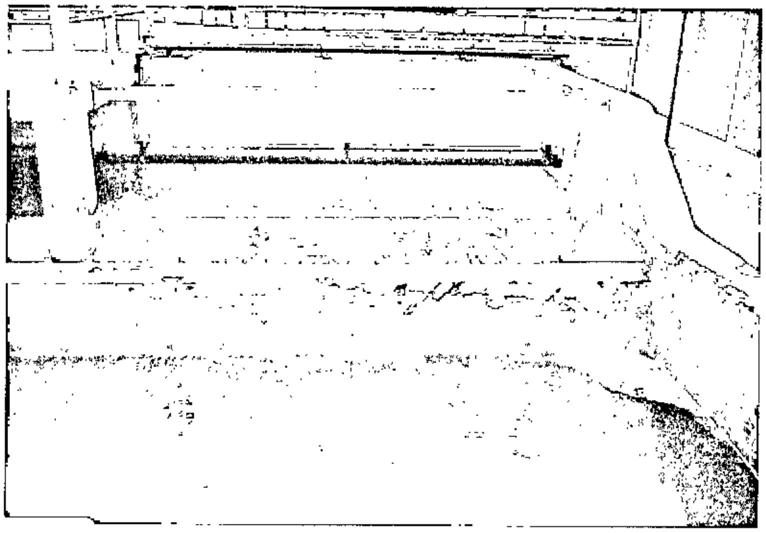

Figure 3.9 Raw water flowing gravitationally to the dividing chamber.

3.6.4 Step Four

3.6.4.1 Clarification

From the dividing chamber the flow is divided equally to supply the four clarifiers which are operated simultaneously and in parallel. Four pulsator clarifiers (Fig. 3.10) are in function. The purpose of clarification is to provide the necessary optimal conditions to favor the sedimentation of the particles and suspended solids present in the raw water. The pulsator provides a piston type sedimentation as the particles are settling down while the flow is going up. In order to increase the sedimentation efficiency a coagulation-floculation of the raw water has to be performed.

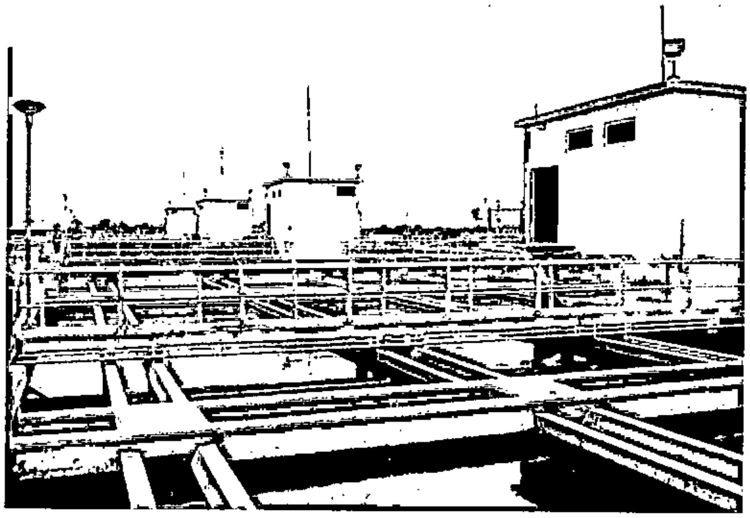

Figure 3.10 Pulsator clarifiers.

3.6.4.2 Coagulation/Floculation

The particles and suspended solids in natural water tend to carry a residual negative charge which is repulsing them and which is restraining the sedimentation (Fig. 3.11). When a strong cationic salt (alum) is injected the negative charges of the particles are destabilized, compressed and therefore the repulsive forces are reduced.

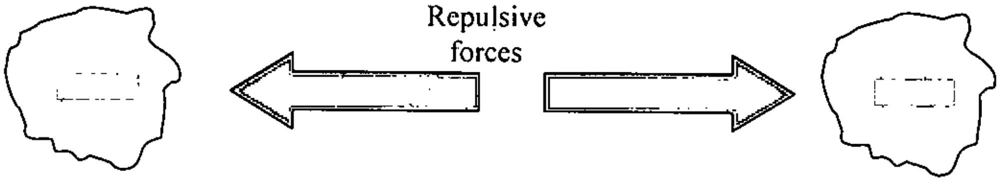

Figure 4.11 Repulsive forces of particles and suspended solids in natural water.

When the right quantity of coagulant is applied to the raw water the particles charges is eliminated and agglomeration is possible. The flocculation is the agglomeration of particles in larger sizes with better and faster settling capabilities (Fig. 3.12). This is achieved under more quiescent mixing conditions inside the pulsator.

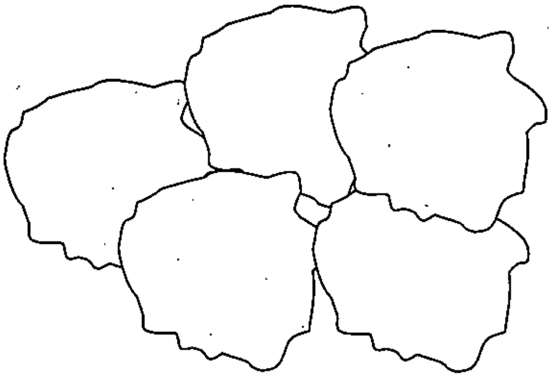

Figure 3.12 Process of flocculation.

3.6.4.3 Sedimentation

In a pulsator the water flows from the bottom to the top at a specific speed. In order to sediment, the particles have to fight the kinetic energy of the water velocity. The maximum tolerable energy to enable the sedimentation of particles is corresponding to about 3 m/h. The energy of a particle is solely gravitational and depends on its density and its structure or shape. The pulsator configuration is designed to apply different levels of kinetic energies, or rising water speed, to the particles. This configuration is at the base of the pulsator which is the suspended sludge bed.

3.6.4.4 Pulsator Configuration

The pulsator configuration is shown in Fig. 3.13. The vacuum chamber is located in the middle of the clarifier. It is the inlet of the pulsator. Perforated and installed in the bottom of the pulsator, they distribute uniformly the flow through the whole floor surface of the clarifier. This is the maximum free surface area or the elevation where the water speed is minimal. This zone is trapping the lighter weight particles. In that zone the wider free flowing surface with lower rising speed is allowing the particles sedimentation. The sludge is building up in that zone. Its function is to restrict the free flowing area and therefore to increase the speed of water making impossible for the particles to sediment. This is the collecting area of the sludge excess produced.

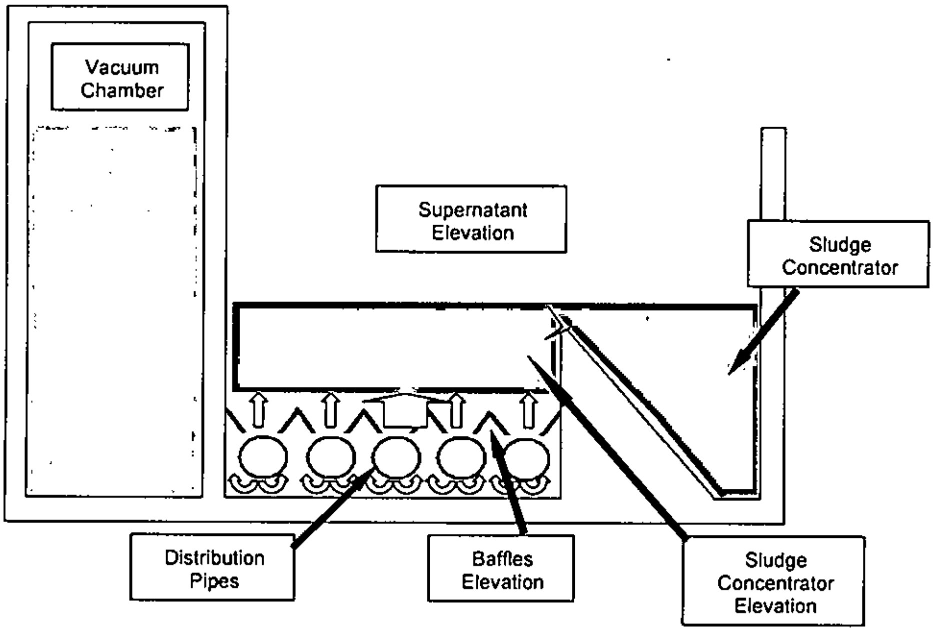

Figure 3.13 Pulsator configuration.

3.6.4.5 Pulsator Hydraulic Pattern

The pulsator hydraulic pattern is shown in Fig. 3.14.

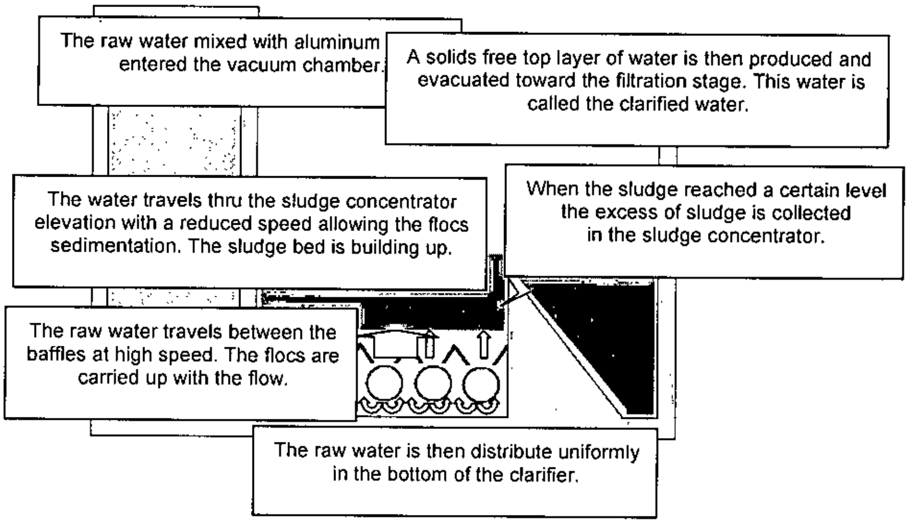

Figure 3.14 Pulsator hydraulic pattern.

3.6.4.6 Pulsator Pulsation System

The pulsator pulsation system is shown in Fig. 3.15. The function of the pulsations is to generate slow motions in the sludge bed by forcing successive and continuous expansions and contractions. The pulsations are achieved with the use of a fan that create a depression in the vacuum chamber. The level of the water in the chamber is rising with the vacuum up to a preset level. The flow in the clarifier is reduced and the sludge goes in contraction. When the chamber is put back to atmospheric pressure by the opening of an air vent valve the water is flushed at high speed in the clarifier and sludge bed goes in expansion. This movement in sludge bed is insuring optimal flocculating conditions (quiescent mixing) and the homogeneity of the sludge bed.

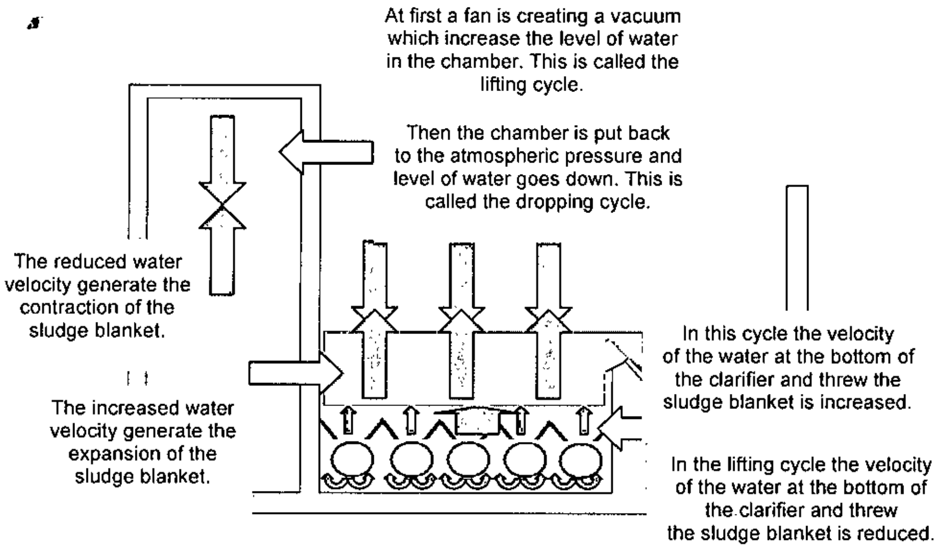

Figure 3.15 Pulsator pulsation system.

3.6.4.7 Filtration with Aquazur V filters

The filtration with AQUAZUR V filters is shown in Fig. 3.16. Aquazur sand filters provide a deep filtration throughout an even depth of graded sand (0.95 m) increasing the filtration cycle time (48 hours). Each filter is individually controlled. A greater water depth is avoiding any risk of degassing. The configuration allows for shorter washing cycle duration. The filter back-washes are performed in three main steps: air blowing, unclogging and rinsing. The wash water is sent to a recovery tank before being recycled at the dividing chamber.

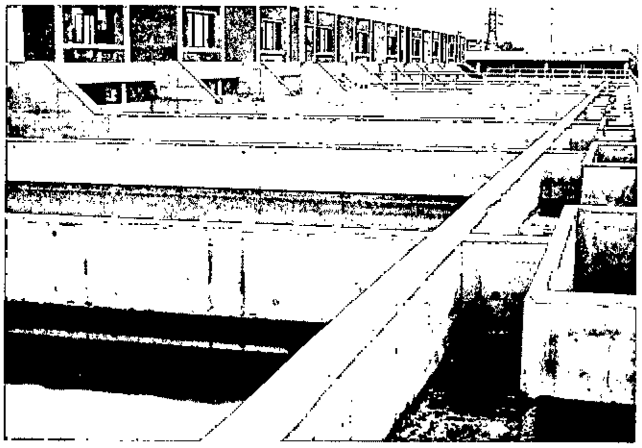

Figure 3.16 Filtration with AQUAZUR V filters.

3.6.5 Step Five: Aquazur V Filter Back-Washes Main Steps

The main steps consist of Air and Water Rinsing, Water Rinsing, End of Rinsing and Back to Filtration Mode. They are shown in Figures 3.17 to 3.20 respectively.

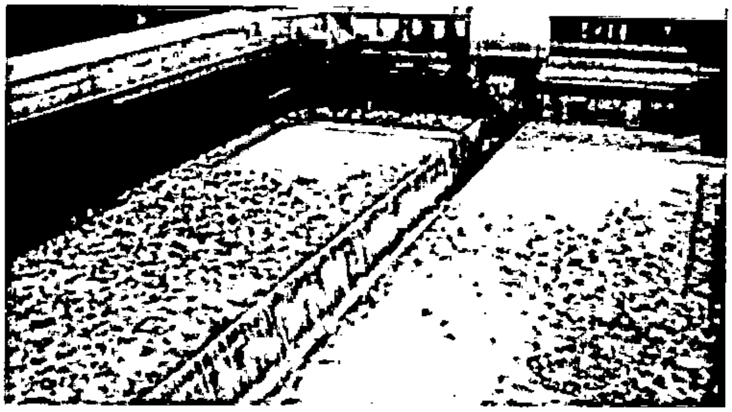

Figure 3.17 First Step: air and water rinsing.

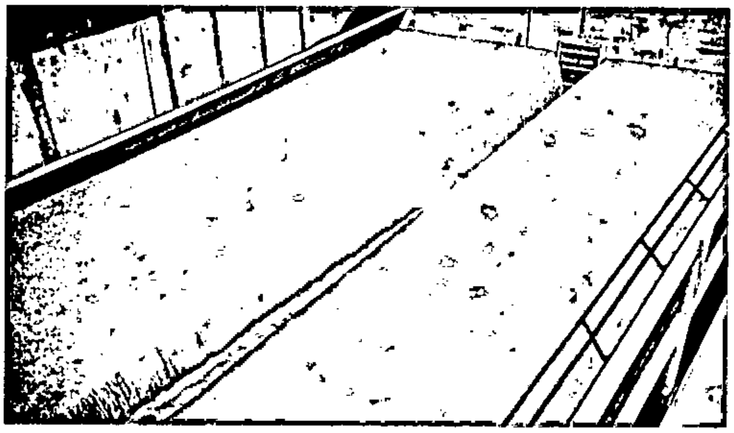

Figure 3.18 Second step: water rinsing.

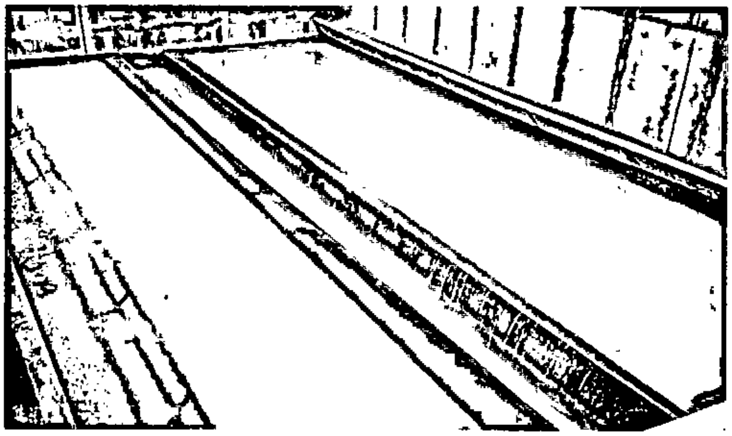

Figure 3.19 Thirst Step: end of rinsing.

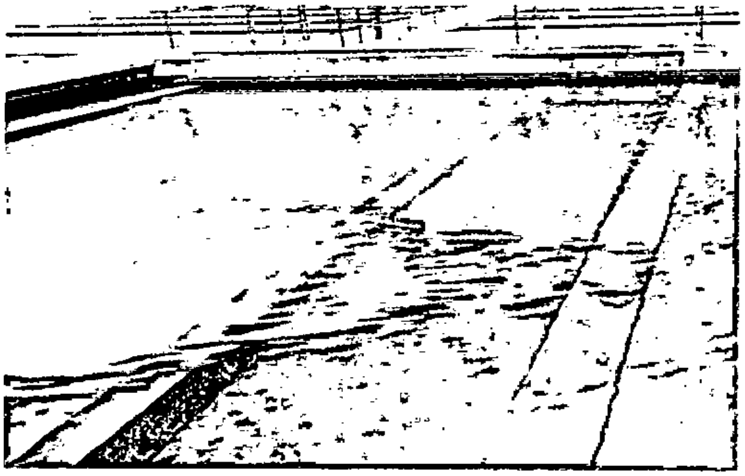

Figure 3.20 Final step: back to filtration mode.

3.6.6 Step Six: Final Water Quality Adjustment and Treated Water Storage

After the filtration a final pH adjustment is performed along with a final disinfection. The final disinfection is achieved with chlorine injection. The water then goes in two contact tanks to provide the appropriate reaction time for the lime and chlorine before being stored in two storage tanks that can provide an autonomy of 2 hours. The contact tanks and the storage tanks can be isolated from each others. The final pH adjustment is achieved with lime injection

3.6.7 Step Seven: Treated Water Pumping Station

The treatment and processing of water is completed and the potable water is sent to the distribution network with the Treated Water Pumping Station (Fig. 3.21). Seven centrifugal pumps are available. Five are fixed speed pumps and two are variable speed pumps controlled with a frequency converter. The pumping station and its automatic controls are designed to maintain a constant pressure of 3 to 6 bars in the distribution network. The delivery pressure is maintained constant with online pressure sensors and a computer manages the speed of the variable pumps. A sampling pump sends water to the onsite laboratory for daily internal water controls and analysis. A sensor measures the free chlorine level on a continuous basis.

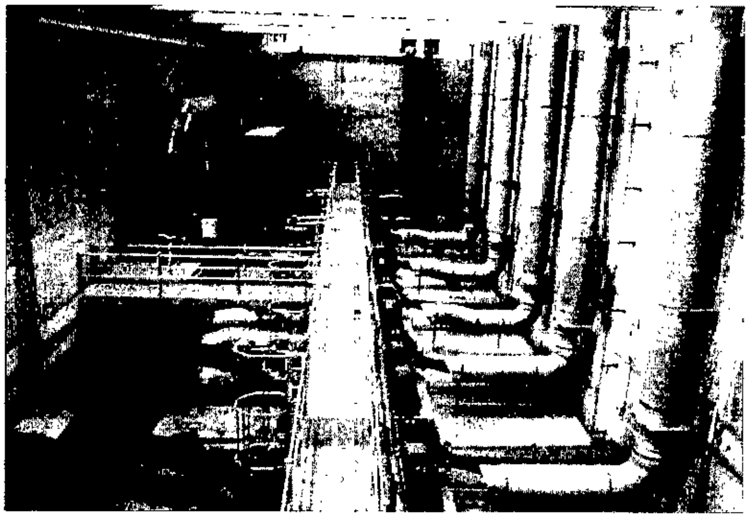

Figure 3.21 Treated water pumping station.

3.7 Sources and Conditions of Alum Sludge in SWTP

Alum sludge is generated from the process of flocculation and water clarification where alum is used as a coagulant to destabilize colloids. Waste water containing alum sludge is discharge automatically in every 15 minutes. There is provision for manual discharge as well when required. The alum sludge is ultimately discharge into sludge drying beds. There are six sludge drying beds in SWTP. Currently only two beds are working. The size of each bed is 150 ft x 30 ft. When these beds are over flown with sludge water, excess sludge water is discharge into the nearby water bodies The beds are not yet filled up with solid sludge. Gravels having 6 inch diameter have been placed above 3 ft sand bed so that water from sludge can easily infiltrate to the ground. The infiltration rate is 185 m^3/hr which is less than the standard infiltration rate of 126 m^3/hr. This implies that the alum sludge is getting exposed to both surface water and groundwater.

CHAPTER 4

RESEARCH METHODOLOGY

4.1 Introduction

The methodology adopted in this study can be divided into definite steps which included collection of alum sludge from sludge drying bed, collection of water from nearby receiving surface water bodies, where supernatant of alum sludge is disposed of. Sample preservation schemes, procedures for analysis of the sludge water receiving supernatant of alum sludge for pre-selected physical and chemical characteristics procedures for analysis of leaching of alum sludge through TCLP and column leaching experiment, procedures for determining aluminum concentration in red amaranth sample produced with and without alum sludge. The methodologies are elaborately discussed in subsequent sections in this chapter.

4.2 Collection of Alum Sludge

Alum sludge were collected from sludge drying beds of Sayedabad Water Treatment Plant located within Dhaka City Corporation. Two types of samples were collected from this plant viz. type-I: sludge from sludge drying bed (Fig. 4.1) and type-II: water from nearby lake (Fig. 4.2) which is used for discharging the excess sludge when the sludge drying beds are filled up with sludge water. Solid sludge samples were accumulated sludge collected from sludge drying beds. The sludge samples were collected at the point of generation. Collection of sludge sample from the point of generation (i.e. end point of the treatment plant) is essential to ensure that it had not undergone significant changes after generation. Plastic containers were selected for storing sludge samples because of possibility of reaction with the sludge compared to glass containers.

The containers were sealed to prohibit loss of moisture. The digested sludge samples were processed in disposable small plastic bottles and kept in cool dry place before analysis. The preservation period did not exceed the recommended maximum permissible periods.

Figure 4.1 Collection of Alum sludge.

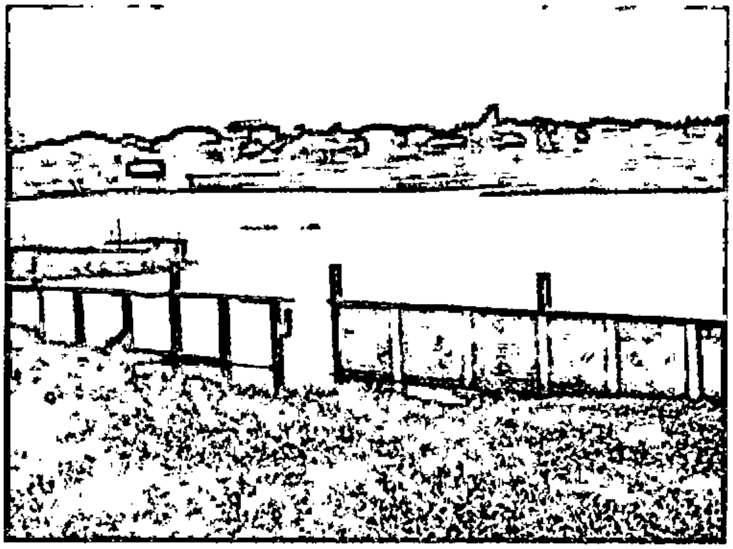

Figure 4.2 Nearby receiving water bodies.

4.3 Collection of Surface Water from Nearby Receiving Water Bodies

Water from nearby lakes (Fig. 4.2) for excess sludge disposal were collected at varying distances (0 feet, 50 feet, 100 feet, 200 feet, 500 feet, 1000 feet) up and down from the point of generation to determine distribution of aluminum concentration and basic physical and chemical characteristics after sludge disposal. Plastic containers were used for storing water from the nearby lake. The containers were also sealed to prohibit loss of moisture.

4.4 Characterization of Supernatant of Excess Alum Sludge

Basic physico-chemical characteristics including moisture content, pH, color, turbidity, alkalinity, ammonia, COD, TDS, sulfate, nitrate have been determined for nearby surface water whereby supernatant of alum sludge is discharged.

4.4.1 Physical Characterization

Moisture content of sludge samples was determined by following dry method. Sludge samples of 25 gm were taken at ambient temperature. Therefore, samples were taken at 105^0C for about 24 hours. After oven drying the samples were placed in a desiccator for half an hour for cooling. After half an hour weight of the sample was taken again. The reduced weight was divided with dry weight and moisture content was found in terms of percentage. The moisture content of sludge was found to be 91.94 per cent.

4.4.2 Characterization of Surface Water

Basic chemical characteristics of surface water including pH, chloride, sulfate, nitrate, ammonia, alkalinity etc. were determined for surface water receiving excess sludge from sludge drying beds as they are likely to influence the leaching characteristics of the sludge in the open environment.

4.5 Toxicity Characteristics Leaching Procedures (TCLP)

The TCLP is a standard USEPA test procedure. It is a soil sample extraction method for chemical analysis. The TCLP analysis simulates landfill condition. Over time, water and other liquids percolate through landfills. The percolating liquid often reacts with the solid waste in the landfill and may pose public and environmental health risks because of the contaminants it absorbs. The TCLP analysis determines which of the contaminants are present in the leachate and their concentration. The leachate is analyzed for appropriate analytes. It is designed to determine the mobility of both organic and inorganic analytes present in liquid, solid and multiphase waters. So it is applicable for a wide range of waters. The test also aims to labeling waters toxic/non-toxic following regulatory limits on leaching set forth by EPA (USEPA, 1992b)

In this test, alum sludge was dried with 105^{0}C temperature for 24 hours. These samples were then crushed and perchloric acid was added. Each sample was taken as 25 gm in 500 ml extraction fluid. Acetic acid (0.57%) was used as standard extraction fluid. The arrangement of TCLP agitation apparatus is shown in Fig. 4.3. The aluminum content was determined by Atomic Absorption Spectrophotometer. Each sample was tested at least twice to obtain average value. Later leaching concentration was converted into mg/kg.

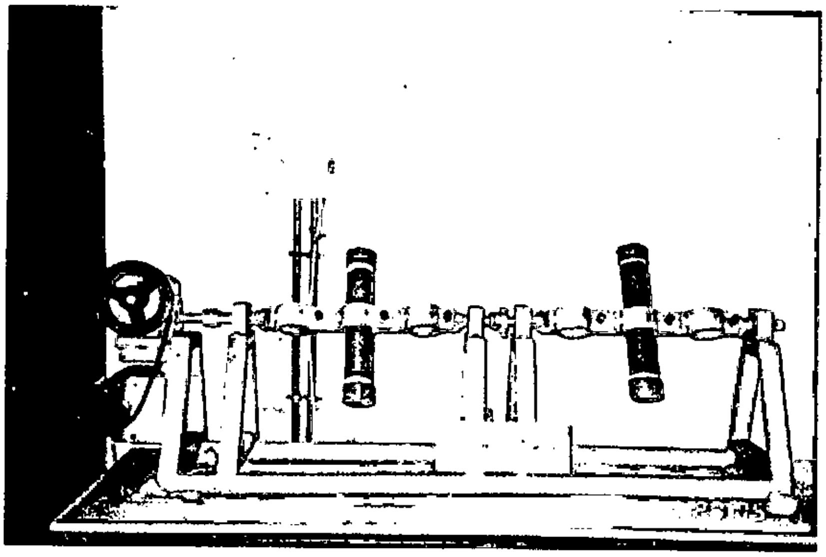

Figure 4.3 TCLP apparatus.

4.6 Column Leaching of Alum Sludge

4.6.1 Introduction

TCLP may not be suitable for assessment of long term leaching of aluminum from alum sludge because such leaching may be kinetically restricted. Thus modification of TCLP to represent the natural leaching environment comparable to real disposal condition is necessary. Column leaching is aimed at carrying out long term leaching from alum sludge. Hence column leaching may be kinetically directed. Column leaching takes longer time than TCLP but it provides better performance in assessing leaching concentration from alum sludge. Leaching is dependent on duration of contact area and pH. In the absence of standard method for long time leaching, column leaching technique with available resources serve as a fruitful alternative employing improvisation in model setup and operation. As leaching in natural condition takes a long time which is not possible to simulate in the laboratory, column leaching experiment has been taken as a standard method.

4.6.2 Experimental Set-up

The fluids of varying composition were allowed to drip through 100 ml burette from nine liter plastic water container placed at higher elevation on a table being connected by plastic tube as shown in Fig. 4.4.

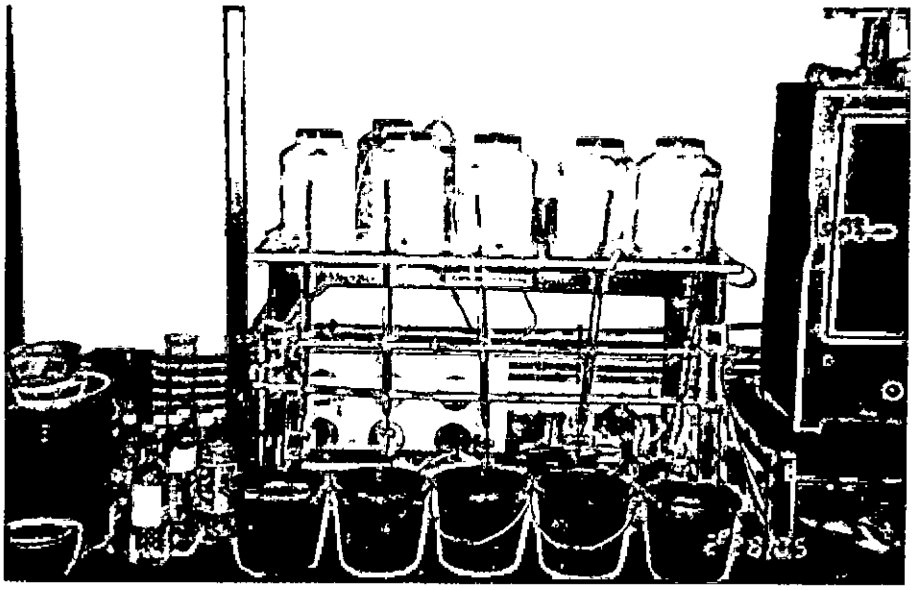

Figure 4.4 Column leaching apparatus.

Five burettes were filled up with 35 gm dry sludge sample (105^{0}C oven dry) each. Improved filter arrangement was made at the bottom of burette with stone chips comprising size 4.8 mm and 16 mm. Extraction fluid media were prepared by trial and error mixing proportions with a view to adjust pH within normal range in between 6 to 7.5 as shown in Table 4.1. Extraction fluids were collected at ten liter water bucket placed beneath each burette. Continuous flow of fluids was maintained by regular refilling of container in each week as and when necessary. Uninterrupted constant flow rate of fluids were maintained by adjusting dripping cock of individual burette. Leaching fluid samples were collected in 100 ml plastic container. Samples were taken every day for first 7 days. After 7 days, it was taken in 1 day by every 7, 15 days being dependent on flow rate of fluid media. Concentrated HCl of 1 ml was added to each collected extraction fluid to preserve it for up to six months to room temperature. Before conducting aluminum content experiment, the sample should be adjusted to pH 3.5 to 4.5 within 5N NaOH. Al content was then determined by Spectrophotometer (Fig. 4.5).

Table 4.1 Constituents of varying extraction fluids

Extraction fluid	pH	Source (salt)	Molecular weight	Concentration (mg/l)	Strength (%)	Amount taken (mg/l)
SO_4^{2-}	6.42	Na_2SO_4	142.0	400	99.0	598
NO_3^-	6.84	$Ni(NO_3)_2.6H_2O$	290.8	10	97.0	24
Cl^-	6.12	NaCl	58.4	500	99.5	829
OH^-	7.31	NaOH	40.0	10	98.0	24

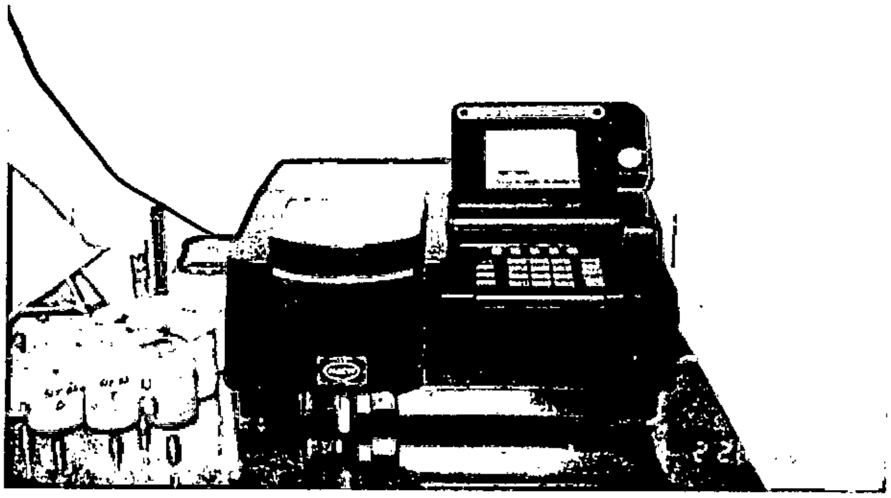

Figure 4.5 Determination of Al content using Spectrophotometer.

As TCLP may not be suitable for assessment of long term leaching of aluminum from aluminum sludge, column leaching would be provided at carrying out leaching longer period as long as sludge constituent continue to leach with sulfate, nitrate, chloride and hydroxide. The results of aluminum present in sludge to that of original sludge at initial stage would then be compared.

A total of five columns were set up and leaching of aluminum was evaluated under continuous flow with five different extraction fluids (one for each column). The extraction fluids were: i) distilled water, ii) nitrate, iii) sulfate, iv) chloride and v) hydroxide. The column experiments have been continuing for periods ranging from three to four months. Leaching is dependent on duration of contact, contact area and pH.

4.7 Al in Red Amaranth Plant

Red amaranth plant has been selected in order to assess the uptake of aluminum from sludge by plant. The red amaranth plant is widely available and human consumable vegetable which can be easily grown in a short period of time and in a variety of soil. The root, stem and leaf of red amaranth plant can be easily differentiated to assess the uptake of Al.

The alum sludge was collected from sludge drying bed of Sayedabad Water Treatment Plant. Two small plots were prepared in front of Civil Engineering Building of BUET. The sludge was then spread over one plot and the other plot was kept with natural soil. The red amaranth (Amaranthus Gangeticu) seeds were sown in both the plots to compare the Al concentration (Fig. 4.6 and 4.7). It took about one month for the red amaranth plants to be grown in. The number of red amaranth plants was 25-30 in both the fields. The average height of red amaranth plants grown in with alum sludge was more (6 inches) than that of red amaranth plant grown in without alum sludge (5.2 inches). The plants were wiped out and taken to the laboratory for the determination of Al concentration.

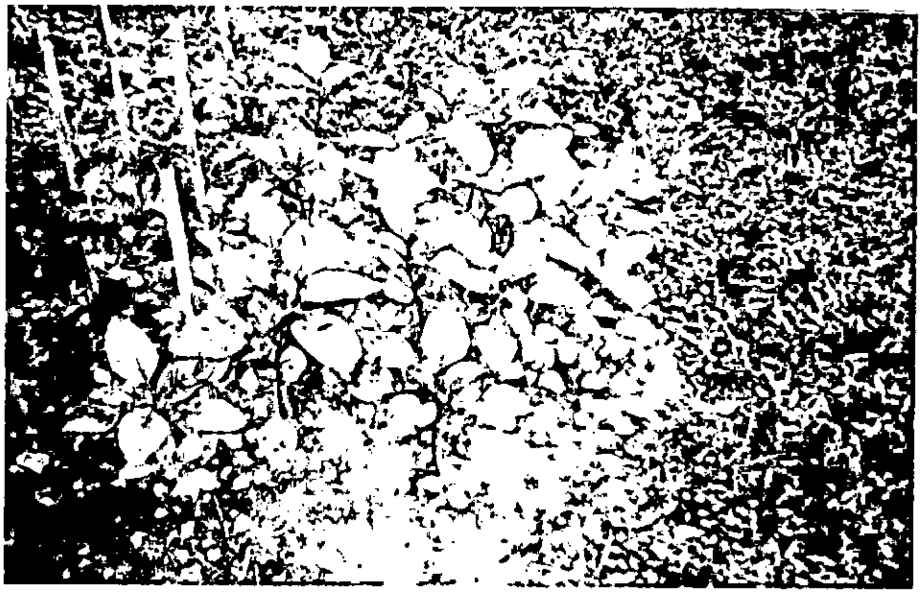

Figure 4.6 Red amaranth plant grown in with alum sludge.

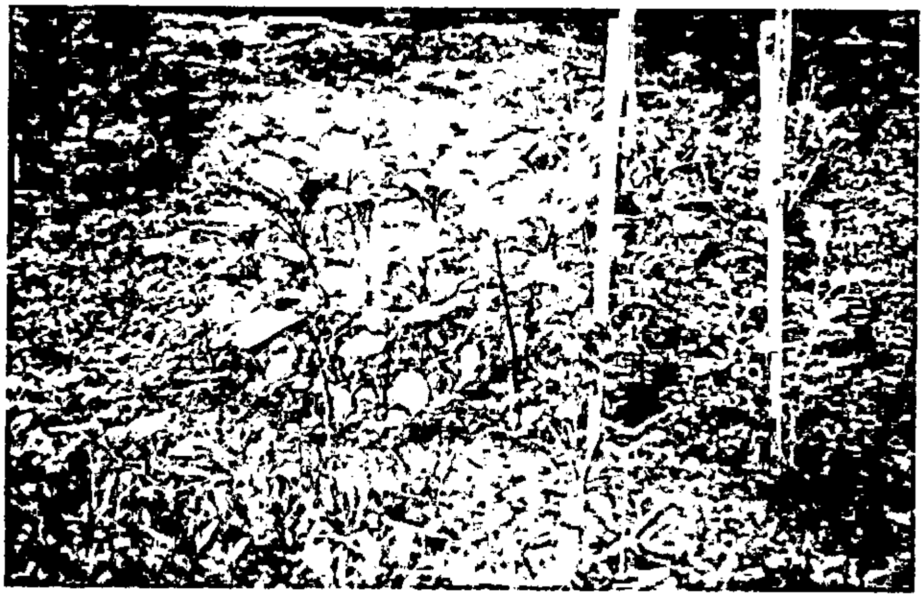

Figure 4.7 Red amaranth plant grown in without alum sludge.

Before analysis, the red amaranth plant samples were divided into three parts: (i) root, (ii) stem and (iii) leaf. For analysis of total aluminum, the different parts/segments of the red amaranth plants were digested. Briefly the digestion procedure consists of the following steps: (i) wash crop samples with distilled water, (ii) divide the crop sample into parts (as described above), (iii) take weight of each part of the sample, (iv) oven-dry the sample at 65^0C for 24 hours and take weight of the oven-dried sample, (v) tale approximately 2 grams of dry crop sample in a volumetric flask and make it moist by adding a few milliliters of deionized water, then add 25 ml nitric acid to the flask and keep it overnight, (vi) heat the flask for two hours to boiling, then after cooling add 10 ml of perchloric acid to the flask and heat again (to boiling) for an hour, (vii) if color of the sample turns yellow, digestion is assumed to be complete, (viii) if color of the sample turns dark, add 2 to 3 ml of nitric acid to the flask and apply heat; repeat the process until the color turns yellow. Aluminum analysis of different segments of red amaranth plant was carried out with atomic absorption spectrophotometer (Farid et al., 2003).

CHAPTER 5

EFFECTS OF ALUM SLUDGE ON ENVIRONMENT

5.1 Introduction

The analysis and discussion of this study begins with the analysis of water quality parameters in nearby receiving water bodies where supernatant of excess alum sludge disposal occurs. The subsequent sections include the analysis of leachability of alum sludge through TCLP and column leaching experiment, analyzing level of aluminum transmission from alum sludge to red amaranth plant, analyzing spatial variation of aluminum in nearby receiving water bodies and finally focusing on mass balance analysis.

5.2 Water Quality of Receiving Water Bodies

Surface water quality-based limits are derived for the water body's critical condition, which represents the receiving water and waste discharge condition with the highest potential for adverse impact on the aquatic biota, human health, and existing or characteristic water body uses. The water quality standards for surface waters require that the effluent not cause toxic effects in the receiving waters.

5.2.1 Water Quality of Receiving Water Bodies

The results of the analysis of surface water parameters are shown in Table 5.1. These limits or standards are set to protect human health and ensure that water is of good quality.

Table 5.1 Surface water quality parameters of nearby alum sludge receiving water bodies

Parameter	Units	Concentration in nearby water bodies	WHO guideline value (2004)	Bangladesh drinking water standards
pH	-	7.84	6.5-8.5	6.5-8.5
Color	Pt-Co unit	55	15	15
Turbidity	NTU	16.2	5	10
Alkalinity	mg/l	6.9	-	-
Ammonia (NH_3)	mg/l	Nil	1.5	0.5
Chemical Oxygen Demand (COD)	mg/l	6.5	-	4
Total Dissolved Solid (TDS)	mg/l	197	1000	1000
Sulfate (SO_4^{2-})	mg/l	18	250	400
Nitrate (NO_3^-)	mg/l	3.8	50	10
Calcium	ppm	24.69	-	75
Al	mg/l	0.026	0.2	0.2

The water quality data for turbidity suggests that turbidity is likely to be a concern whereas the pH is within the range of the limit. The color is observed to possess high concentration. The observed concentration of ammonia, sulfate, nitrate and total dissolved solids are low. Concentration of chemical oxygen demand is moderate. The observed concentration of calcium and aluminum are also low in surface water receiving supernatant of alum sludge. Hence, on surface water there is no significant impact due to disposal of supernatant of alum sludge.

Aluminum coagulants contain high concentrations of ionic aluminum, the toxic form. Toxicity is very dependant on pH and increases at lower pH. At high pH, most aluminum is present in solid form and is not bio-available. Below pH 6 it is mostly in the dissolved, bio-available form. The bioavailability and toxicity of aluminum is generally greatest in acid solutions and generally most toxic over the pH range 4.4 - 5.4 with a maximum toxicity around pH 5.0-5.2 (ANZECC, 2000). However, at pH of 6.5 to 8.5, which is the normal range for natural waters, there is generally considered to be little threat of toxicity. Where pH of natural waters is outside this range, instream values including fish and invertebrate diversity would tend to be limited in any event, and environmental sensitivity reduced. As the pH is within the range of 6.5-8.5, it can be concluded that there is little threat of bioavailability and toxicity of aluminum in the receiving water bodies.

5.2.2 Spatial Variation of Al in Nearby Receiving Water Bodies

For measuring spatial variation of aluminum in nearby surface water where supernatant of alum sludge disposal occurs, sample was collected from different locations. Samples were collected at 10 ft, 50 ft, 100 ft in the north direction and 10 ft, 50 ft, 100 ft, 200 ft, 500 ft, 1000 ft in the south direction from the point of generation. Test results shown in Table 5.2 show that Al content at the point of generation in nearby water bodies is the highest (0.026 ppm) while the lowest amount (0.0025 ppm) is found at 1000 ft in the south from the point of generation. Al content in the north direction from the point of generation ranges from 0.020 ppm at 10 ft to 0.010 ppm at 100 ft. Al content in the south direction varies from 0.006 ppm at 10 ft to 0.0025 ppm at 1000 ft. Al concentration at varying distances from the point of generation is shown in Fig. 5.1. It shows that the distribution of Al decreases with distances along the north-south directions from the point of generation. Aluminum concentration at the point of generation is 0.026 ppm which is lower than Bangladesh drinking water standard and WHO guideline value (0.2 ppm). It is apparent that there is little variation of Al concentration distribution at varying distances from the point of generation. Hence, there is no significant effect on surface water due to disposal of supernatant of alum sludge.

Table 5.2 Spatial Variation in Al Concentration

Distance (feet)	0	10	50	100	-10	-50	-100	-200	-500	-1000
Al Concentration (ppm)	0.026	0.02	0.015	0.01	0.006	0.006	0.004	0.0035	0.0027	0.0025

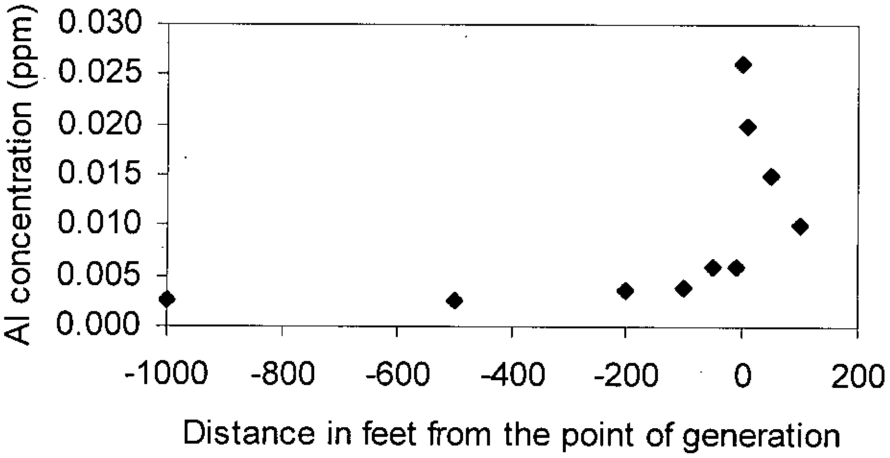

Figure 5.1 Spatial variation of Al concentration in nearby water bodies.

5.3 Concentration of Al in TCLP Extracts of Sludge

TCLP is comprised of four fundamental procedures: sample preparation for leaching, sample leaching, preparation of leachate for analysis and leachate analysis. The TCLP test was preformed on raw sludge samples. The leaching test was performed on alum sludge collected from sludge drying beds. During the TCLP, constituents are extracted from the wash to simulate the leaching actions that occur in landfills.

According to USEPA, in some waters post-precipitation of aluminum may take place after treatment. This could cause increased turbidity and aluminum water quality slugs under certain treatment and distribution changes. USEPA also agrees with the World Health Organization (WHO, 1984) that discoloration of drinking water in distribution systems may occur when the aluminum level exceeds 0.1 mg/l in the finished water. WHO further adopts a guidance level of 0.2 mg/l in recognition of difficulty in meeting the lower level in some situations.

Table 5.3 shows that Al concentration in the TCLP extracts of alum sludge is 45000 mg/kg. This leachate concentration can be significant enough to percolate through alum sludge into groundwater. For aluminum, drinking water standard is 0.2 mg/l according to WHO guideline value, 2004 and Bangladesh drinking water standards. Leaching values might be large enough to exceed the allowable limit of Al concentration in groundwater and surface water. The percolated leachate concentration might raise the concentration of Al in groundwater above the allowable limit as suggested by WHO guideline value and Bangladesh standards for drinking water. As a result, it might affect human health and food chain.

Table 5.3 Concentration of Al in TCLP test and digestion method

TCLP (mg/kg)	Total Al (mg/kg)
45000	48000

5.4 Leachable Aluminum from Sludge through Column Leaching

TCLP may not be suitable for assessment of long term leaching of aluminum from alum sludge because such leaching may be kinetically restricted. Thus modification of TCLP to represent the leaching environment comparable to real disposal conditions is necessary. Hence column leaching was carried out to simulate natural leaching environment.

Results from Figure 5.2 indicate that leaching of Al from alum sludge varies in different media conditions. Results from Table 5.2 indicate that residual alum concentration after column leaching is more in case of extraction fluid containing nitrate and distilled water than fluids containing sulfate, hydroxide and chloride anion which reflects highest leaching in case of extraction fluid containing chloride anion followed by fluids containing hydroxide, sulfate, distilled water and nitrate.

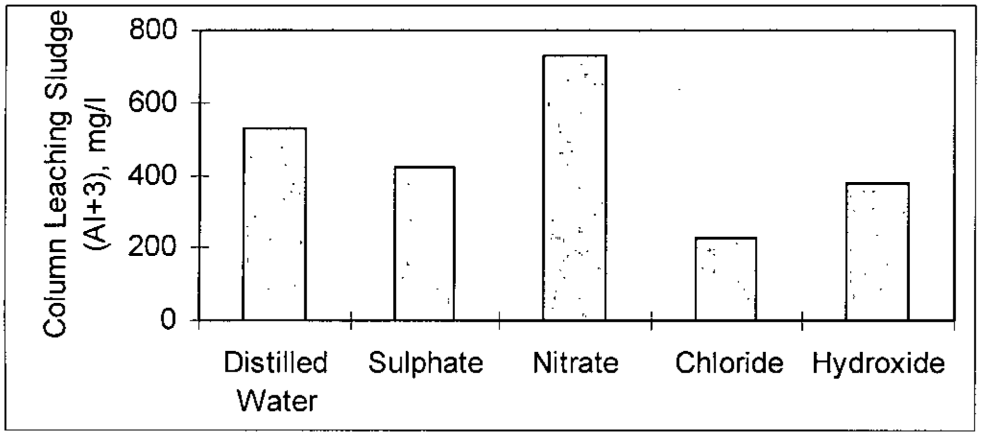

Figure 5.2 Leaching of Al in different fluid media used in column leaching test.

Table 5.4 Results of Al concentration in extraction fluid of column leaching

Fluid media	Distilled water	Na_2SO_4 solution	$Ni(NO_3)_2.6H_2O$ solution	NaCl solution	NaOH solution
Alum concentration in TCLP extracts (mg/kg)	21200	16800	29200	9000	15200

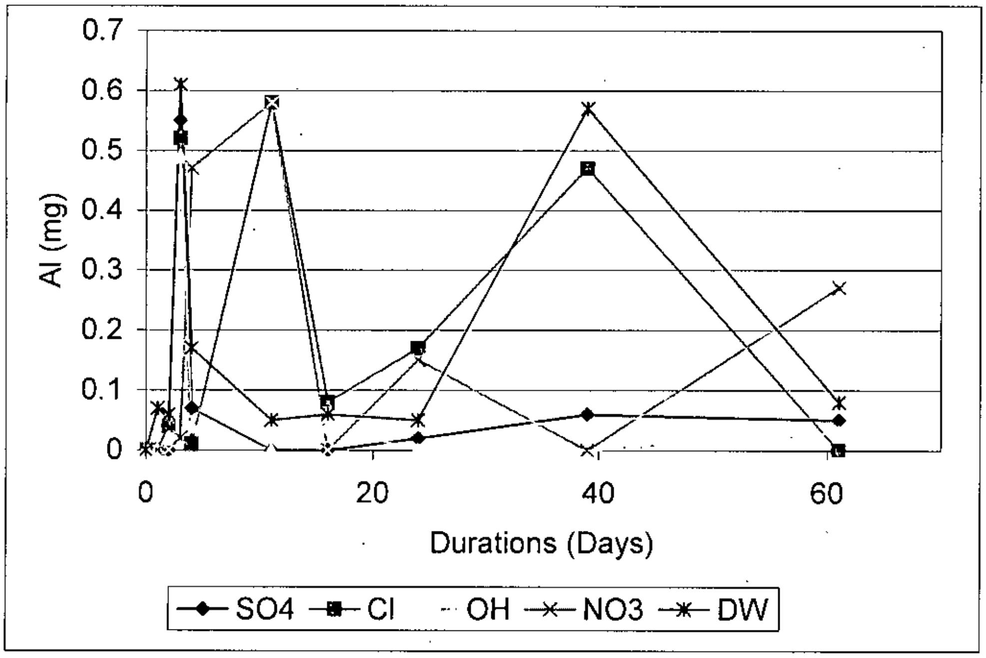

Figure 5.3 Column leaching in varying fluid media (individual strength value).

The results of column leaching tests from Figure 5.3 indicate that leaching of Al in extraction fluid containing distilled water was maximum. It shows that of all the extractions, distilled water showed highest value followed by chloride, sulfate, hydroxide and nitrate. This can be explained on the basis of ionic strength effect. The highest leaching by distilled water appears to be due to the lack of dissolved ions in distilled water. As water is an aggressive liquid, leaching of aluminum from distilled water is much higher than that of fluid containing anion bounds (Cl^-, SO_4^{2-}, NO_3^-, OH^-) at initial stage. However, leaching from distilled water continues to decrease as duration of leaching progresses.

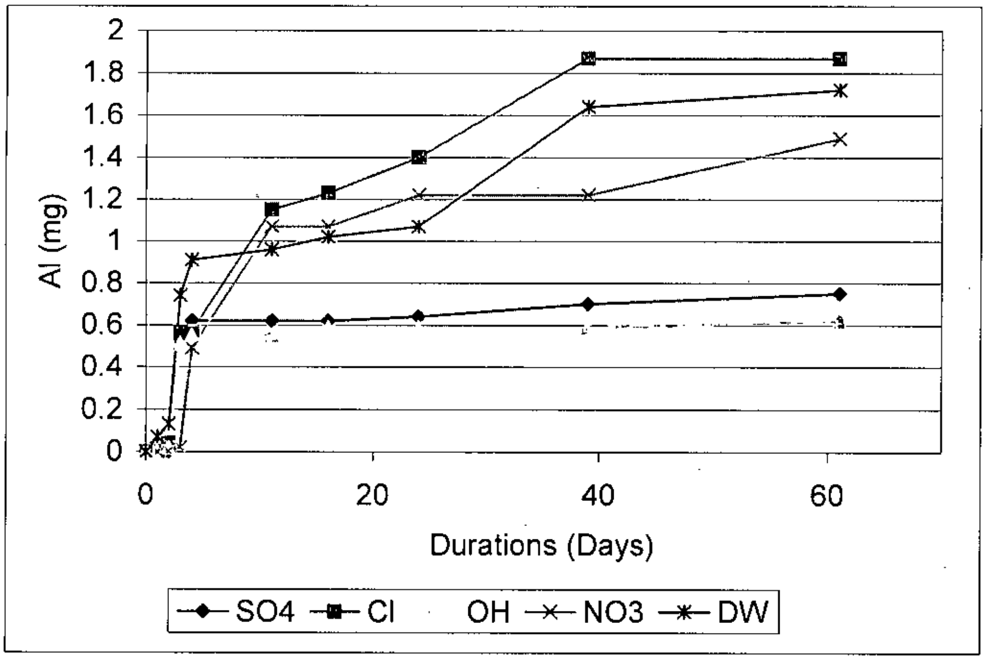

Figure 5.4 Column leaching in varying fluid media (cumulative value).

Cumulative leachate concentrations from Figure 5.4 indicate that leachate concentration from OH⁻ anion continues to increase in much lower rate and maintains a steady state. It is observed that leachate concentration from chloride anion fluid media is higher than distilled water, nitrate, sulfate and hydroxide anion fluid media. The higher the ionic strength, the greater is the leachate concentration.

The leaching constants for different fluid media were derived from gradients of the fitted straight lines of the plot of Al concentration versus duration as shown in Figures 5.5 to 5.9. The horizontal axis has been taken as the logarithm of (T+1), where T is the duration in days.

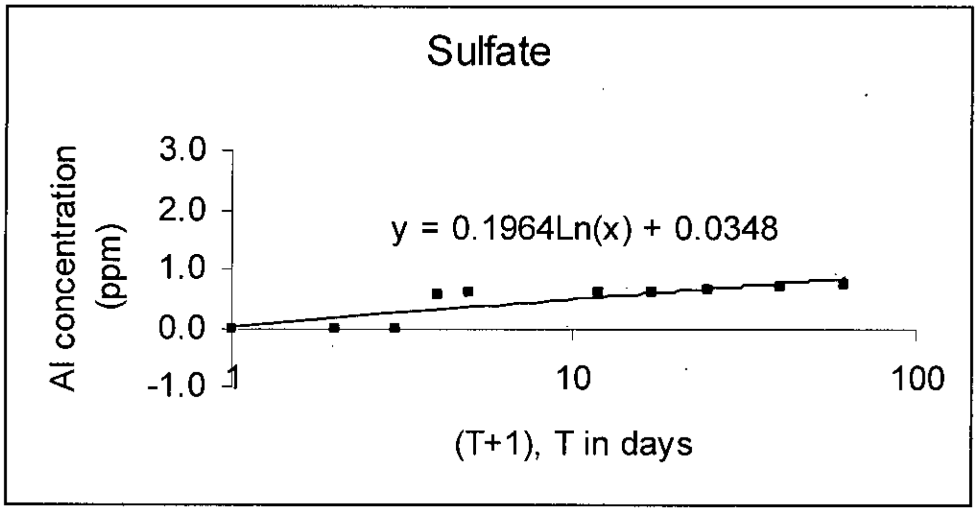

Figure 5.5 Cumulative aluminum leaching concentration with elapsed time for sulfate anion fluid media.

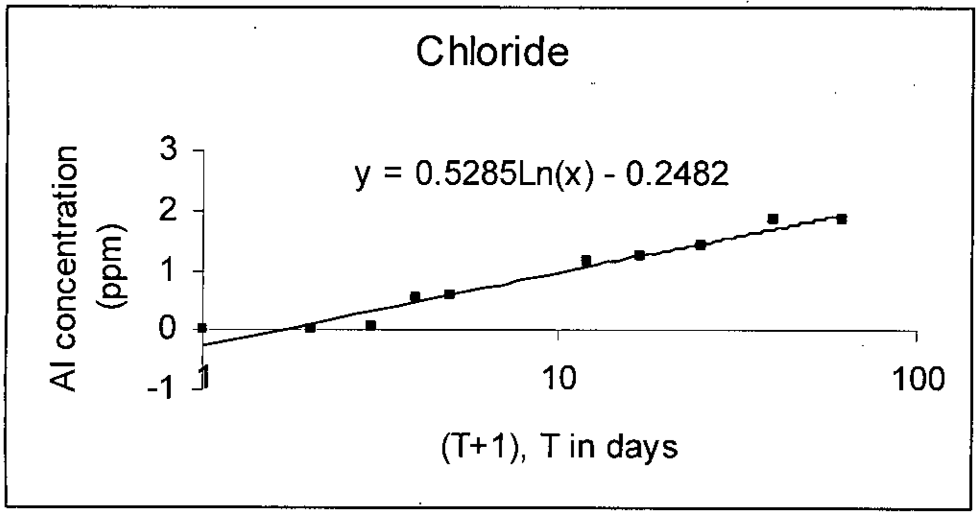

Figure 5.6 Cumulative aluminum leaching concentration with elapsed time for chloride anion fluid media.

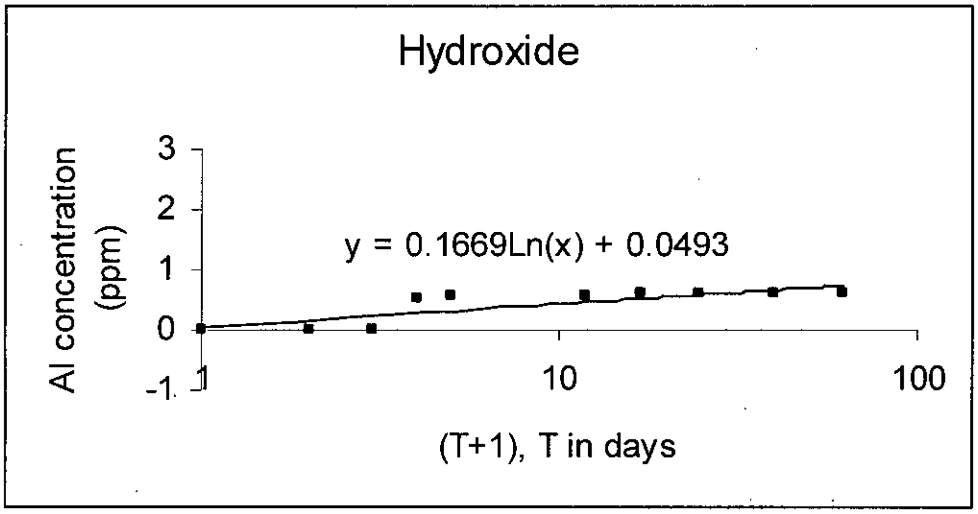

Figure 5.7 Cumulative aluminum leaching concentration with elapsed time for hydroxide anion fluid media.

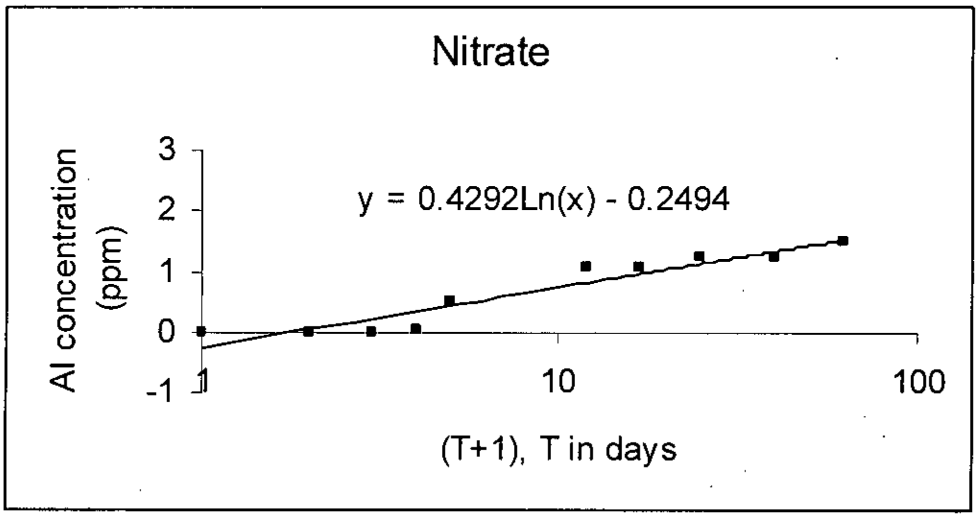

Figure 5.8 Cumulative aluminum leaching concentration with elapsed time for nitrate anion fluid media.

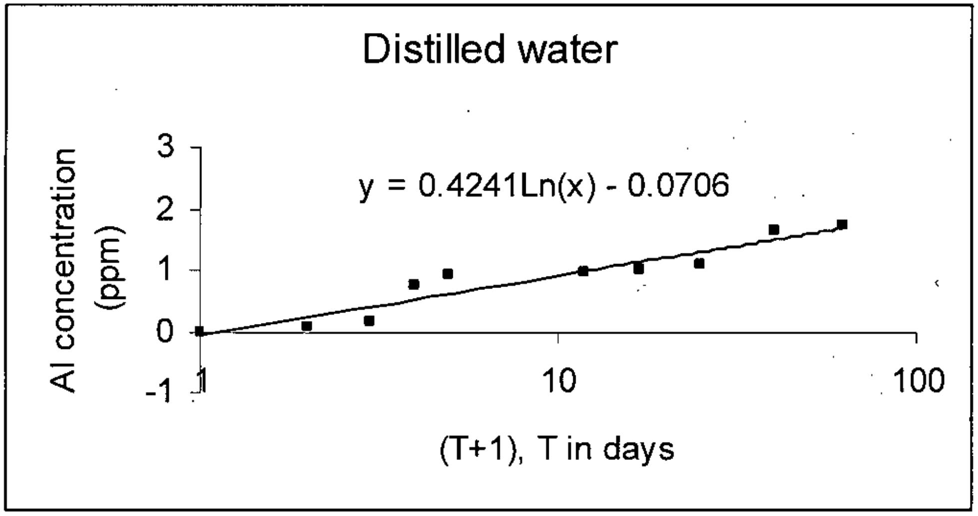

Figure 5.9 Cumulative aluminum leaching concentration with elapsed time for distilled water.

The intercepts of the fitted line indicate the initial Al concentration. Leaching constants for varying fluid media in column leaching is shown in Table 5.5.

Table 5.5 Leaching constants for varying fluid media in column leaching

Fluid media	Leaching constant
Na_2SO_4 solution	0.4522
NaCl solution	1.2168
NaOH solution	0.3842
$Ni(NO_3)_2.6H_2O$ solution	0.9844
Distilled water	0.9766

It is seen from Table 5.5 that the leaching constant varies with ionic strength. The higher the ionic strength, the higher the leaching constant and hence higher the leachability. The unit of leaching constant is ppm/day. The leaching constant from column leaching experiment shows that chloride anion extraction fluid causes higher leachate concentration than other types of extraction fluid media.

5.5 Uptake of Al from Sludge by Red Amaranth Plant

Vegetables are important food crops of Bangladesh and are rich in vitamins and minerals which are very essential for maintaining good health. So far the author's knowledge goes, no work has been done in Bangladesh to find out the effects of using aluminum contained sludge on crop production and its carry over effect on food chain. For this reason, this study was undertaken to find out the level of Al transmission from sludge to red amaranth plants. Al content of sludge was also determined to indicate spread of alum compared to natural soil. Average aluminum concentration in natural soil is 71,000 ppm or mg/kg and common range lies between 10,000 - 300,000 ppm or mg/kg (USEPA, 1983).

In order to assess the spread the aluminum from sludge to plants, red amaranth plants grown in with and without sludge were analyzed for their aluminum content. The concentrations of Al in soil and sludge and in different parts of red amaranth with and without sludge are presented in Table 5.6. From the test results presented here, it is clear that red amaranth plants accumulate Al from soil and the concentration varies among the different parts of plants.

Al concentrations in root, stem and leaf of studied red amaranth were different. The Al concentrations accumulated in roots, stems and leaves of red amaranth plants grown in with and without alum sludge are shown in Fig. 5.10. Roots of red amaranth accumulated the highest concentration followed by stem and leaf. The quantity of accumulated Al decreases from root to leaf gradually. The mean Al concentration in leaf is 2.42 times less than stem and 4.1 times less than root. Al in sludge is 3.54 times higher than alum in natural soil. The alum concentration of red amaranth plants produced in Al sludge was 451 ppm which is about 3 times higher than the alum concentration of red amaranth plants produced in Al sludge free soil.

From this study it can be concluded that the content and range of Al were found higher in the red amaranth plants growth with alum sludge than those grown with alum free soil. The trend of Al accumulation was higher in root and lower in leaf.

The red amaranth plants accumulated Al through the alum sludge. The reason may be due to the fact that Al present in sludge is easily mobilized and hence readily taken up by plants.

From Table 5.6, it is found that the average aluminum content in stem and leaf is 10188 mg/kg. If a human takes 100 gm of red amaranth vegetables in a day, the intake of aluminum would be 1018.8 mg. On acute exposure, aluminum is of low toxicity. In humans, oral doses up to 7200 mg/d (100 mg/kg bw per day) are routinely tolerated without any signs of harmful short-term effects (Eastwood et al., 1984). The intake of aluminum through red amaranth vegetables is utterly low compared to the oral doses which can be tolerated. So consumption of red amaranth plants grown in with alum sludge does not pose any threat to human body.

Table 5.6 Aluminum concentration in different parts of plants with and without sludge

Part of plant	Al (ppm) [without sludge]	Al (ppm) [with sludge]
Root	99.50	246.92
Stem	45.92	144.24
Leaf	8.60	59.51

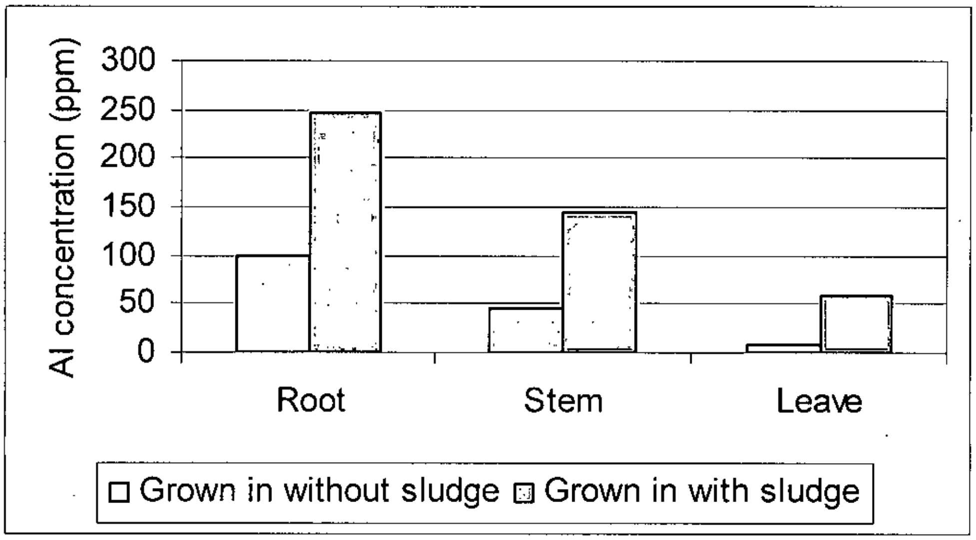

Figure 5.10 Variation of Al concentration in different parts of plants with and without Al sludge.

5.6 Mass Balance Analysis

The concentration of Al is low in case of leached extracts of aluminum from column leaching experiments. Al content in residual samples after carrying out column leaching experiment reflects similar result like Al content of raw sludge. It is also observed that Al content of raw sludge is sum of leached aluminum and residual aluminum in the sludge after column leaching experiment as shown in Table 5.7.

Table 5.7 Mass balance analysis

Al content (mg/kg)	Sulfate	Nitrate	Chloride	Hydroxide	Distilled water
Raw sludge	48000	48000	48000	48000	48000
Column leaching extracts	19.2	76.7	84.0	25.7	90.3
Al after column leaching	47890	47826	47801	47869	47781
Deviation (%)	1.9	2.0	2.4	2.2	2.7

The deviation in percent ranges from 1.9 to 2.7. Mass balance analysis indicates a considerable difference in Al content in alum sludge between residual aluminum and leached aluminum with varying extraction fluids in column leaching experiment.

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

Large scale use of aluminum in coagulation process during water treatment in Sayedabad Water Treatment Plant may generate significant quantities of aluminum rich treatment sludge and disposal of this sludge may lead to environmental pollution. The common practice is open water disposal or on land disposal. Aluminum leaching from such sludge may pose potential threat to both groundwater and surface water with long-term consequences. In this study, the effects of aluminum in the sludge of Sayedabad Water Treatment Plant on environment have been studied.

The water quality parameters suggest that turbidity is likely to be a concern whereas the pH is within the range of the limit. The color is observed to possess high concentration. The observed concentration of ammonia, sulfate, nitrate and total dissolved solids are low. The concentration of chemical oxygen demand is moderate. The observed concentration of calcium and aluminum are also low in surface water receiving supernatant of alum sludge. There is no significant impact on surface water due to disposal of supernatant of alum sludge.

Spatial variation of Al concentration indicates that concentration of aluminum decreases with the increase of distances from the point of generation. Further it shows that at the point of generation, concentration of aluminum is 0.026 ppm which is less than WHO guideline values and Bangladesh standards for drinking water. The distribution of Al decreases with distances along the north-south directions from the point of generation. There is little variation of Al concentration distribution at varying distances from the point of generation.

The leachate concentration from TCLP can be significant enough to percolate through alum sludge into groundwater. This percolated leachate concentration might raise the concentration of Al in groundwater above the allowable limit as suggested by WHO guideline values and Bangladesh standards for drinking water.

Leachate concentration from column leaching test indicates that distilled water and chloride anion show prominence. Significant amount of Al could be leached from alum sludge by distilled water, chloride, sulfate, nitrate and hydroxide anions. The leachate concentration from chloride anion fluid media is higher than distilled water, nitrate, sulfate and hydroxide anion fluid media. Mass balance analysis indicates that aluminum content of raw sludge is sum of leached aluminum and residual aluminum in the sludge after column leaching experiment.

This study has attempted to find out the level of Al transmission from sludge to red amaranth plants. Red amaranth plants accumulate Al from soil and the concentration varies among the different parts of plants. Roots of red amaranth accumulated the highest concentration followed by stems and leaves. The quantity of accumulated Al decreases from roots to leaves gradually. The mean Al concentration in leaves is 2.42 times less than stems and 4.1 times less than roots. The alum concentration of red amaranth plants produced in Al sludge was 451 ppm which is about 3 times higher than the alum concentration of red amaranth plants produced in Al sludge free soil.

6.2 Recommendations for Future Studies

The present study analyzed the concentration of aluminum only in red amaranth plants grown with alum sludge. Alum uptake of different crops could be physiologically different. Future studies should aim at determining the bioavailability of aluminum through different crops grown with alum sludge not covered in this study. More studies are needed to develop a better understanding of Al accumulation in soil and food chain and its possible impacts.

Mass balance analysis was carried out for aluminum extracts in column leaching experiment. Similar analysis can be undertaken for aluminum in TCLP extracts. An analysis out of these two mass balance analyses may be carried out with a view to draw a correlation between mass balance analysis with TCLP and column leaching.

For the purpose of research, spatial variation of aluminum concentration was conducted at near receiving water bodies where supernatant of alum sludge disposal occurs. From experiment, it was observed that aluminum concentrations are diluted with distances which implies no significant impacts on surface water. Further study may be carried out to determine effects on groundwater due to disposal of supernatant of alum sludge to surface water.

Accumulation of Al may also vary from place to place. The reason may be due to variation in soil properties, soil fertility and concentration. These issues can be addressed in future studies.

The present study did not include analysis of the comparison of the trend of accumulation of Al in leafy vegetables and fruity vegetables. Further studies should focus on this issue.

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