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**NAME:** Miss Rachaneepen Pensit

#### THIS THESIS HAS BEEN ACCEPTED BY

*Wilai Chiemchaisri*

**THESIS ADVISOR**

( Assistant Professor Wilai Chiemchaisri, D.Tech.Sc. )

*Pipat Pooripanyakun*

**COMMITTEE MEMBER**

( Mr. Pipat Pooripanyakun, M.Eng. )

*Sutharin P.*

**COMMITTEE MEMBER**

( Assistant Professor Sutharin Pathomvanich, Ph.D. )

*Mongkol Damrongsri*

**DEPARTMENT HEAD**

( Mr. Mongkol Damrongsri, Dr.Eng. )

**APPROVED BY THE GRADUATE SCHOOL ON**

31 March 2006

*Vinai Artkongharn*

**DEAN**

( Associate Professor Vinai Artkongharn, M.A. )

THESIS

LEACHABILITY OF ALUMINUM IONS FROM  
WATER TREATMENT SLUDGE

RACHANEEPEN PENSIT

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The objective of this study was to investigate effects of environmental factors on aluminum leachability from water treatment sludge using three different soil acidities (Nonthaburi soil, Rangsit soil, and Korat soil). The experiment was divided in two parts: 1) batch leaching experiment, to study effects of soil texture, temperature, and moisture content on aluminum leaching in three different soil-sludge mixing ratios (25:75, 50:50, and 75:25 %DW) 2) column leaching experiment, to study effect of rain pH on aluminum leaching using synthetic acid rain.

In batch experiment, it was found that the Rangsit soil, the extremely acid soil, under soil-sludge mixing ratio of 75:25 %DW gave highest amount of aluminum leaching (13.32-15.10 mg/l). With this mixing ratio and under temperature variation (5, 20, 28-32, and 35-40 °C) it was found that the 28-32 °C incubation gave highest amount of aluminum leaching (13.93-14.85 mg/l). At this temperature, when the moisture content was controlled at 30%, highest aluminum leaching level (14.20-14.77 mg/l) was found as compared to the others (5%, 10%, and 20% MC).

In column leaching experiment, it was found that there were no significant differences in aluminum leaching levels for rain pH of 6.7 and 7 (0.9-1.1 mg/l). However, under acid rain conditions (pH 5.5 and pH 3) aluminum concentration was increased sharply (pH 5.5 : 2.52 mg/l and pH 3 : 3.10 mg/l). It indicates that rain pH had similar effect on aluminum leaching as soil pH. The lower the pH value, the higher amount of aluminum leaching had appeared in the soil-sludge amendment.

P. Rachaneepen

Student's signature

Wilai Chiemchaisri

Thesis Advisor's signature

27/03/06

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## LIST OF ABBREVIATIONS

%	=	Percent
%DW	=	Percent dry weight
%MC	=	Percent moisture content
"	=	Inch
°C	=	Degree Celsius
°F	=	Degree Fahrenheit
$\mu\text{g/l}$	=	Microgram per liter
AAS	=	Atomic absorption spectroscopy
Al Conc.	=	Aluminum concentration
aq	=	Aqueous
ASTM	=	American Society for Testing and Materials
BOD	=	Biochemical oxygen demand
BOD <sub>5</sub>	=	Biochemical oxygen demand at day 5 of incubation period
Bq/l	=	Becquerel per liter
CEC	=	Cation exchangeable capacity
cm	=	Centimeter
cmol/kg	=	Centimole per kilogram
COD	=	Chemical oxygen demand
dS/m	=	Deci-Siemens per meter
EC	=	Electrical conductivity
ECEC	=	Effective cation exchange capacity
EEG	=	Electroencephalogram
g	=	Gram
$\text{g/cm}^3$	=	Grams per cubic centimeter
g/kg	=	Gram per kilogram
g/mol	=	Gram per mole
$\text{g}_{\text{O}_2}/\text{g}_{\text{soil}}/\text{day}$	=	Gram oxygen per gram soil per day

**LIST OF ABBREVIATIONS (Continued)**

hrs.	=	Hours
ISO	=	International organization of standardization
J/(mol·K)	=	Joule per mole Kelvin
K	=	Kelvin
kJ/mol	=	Kilo-Joule per mole
L	=	Liter
L/S	=	Liquid to solid ratio
LC <sub>50</sub>	=	Lethal Concentration 50, the concentration of a chemical which kills 50% of a sample population
M	=	Molarity
MC	=	Moisture content
m/kg	=	Meter per kilogram
m <sup>3</sup> /day	=	Cubic meter per day
meq/100 g	=	Milli-equivalent per 100 gram
mg Al/kg	=	Milligram aluminum per kilogram
mg Al/l	=	Milligram aluminum per liter
mg Ca/l	=	Milligram calcium per liter
mg/kg	=	Milligram per kilogram
mg/l	=	Milligram per liter
Million cu.m.	=	Million cubic meter
ml	=	Milliliter
ml/min	=	Milliliter per minute
mm	=	Millimeter
mol/l	=	Mole per liter
N	=	Normality
nm	=	Nanometer

**LIST OF ABBREVIATIONS (Continued)**

No.	=	Number
NTU	=	Nephelometric turbidity unit
pH	=	Acidity or alkalinity of a solution
pm	=	Picometer
ppm	=	Parts per million
rpm	=	Rotations per minute
vs.	=	Versus

# LEACHABILITY OF ALUMINUM IONS FROM WATER TREATMENT SLUDGE

## INTRODUCTION

### Background of the Study

Alum or hydrated aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 14\text{H}_2\text{O}$ ) is widely used for water purification to flocculate dissolved and suspended particles settling out of the water. This process normally produces sludge containing high aluminum content. The sludge may cause agricultural and health problems if the aluminum leaches into soil or ground/surface water resulting from sludge disposal by landfilling.

The Metropolitan Waterworks Authority (MWA) has been used alum as coagulant in water supply production for a long time as the chemical is inexpensive and has high efficiency in coagulating. There are 4 water treatment plants operating in the Bangkok Metropolitan Region: Bangkhen, Samsen, Thonburi and Mahasawat, with different production capacities. The Bangkhen water treatment plant has the highest capacity of 3,200,000 m<sup>3</sup>/day and produces 2,987,000 m<sup>3</sup> water/day (MWA, 2004). The sludges produced per day are about 345 tons or 126,000 tons/year (Keawruenrom et al., 2001).

Currently, sludge of MWA water treatment is disposed only by landfilling. Because of high amount of sludge produced from water treatment, there are several studies about possibility in using water treatment sludge in many different ways, one of the most popular sludge applications is to use water the sludge to substitute chemical fertilizer in agriculture as the sludge itself has high nutrition compounds (Summer, 1977). As the sludge has high aluminum content this may cause aluminum accumulation in soil due to the sludge disposal by landfilling. When aluminum leachs into agricultural areas and water resources, it may cause aluminum accumulation in plants which leads to problems regarding to aluminum toxicity in plants and agronomic crops. However, amount of aluminum leaching into the soil or body of water is dependant on many factors such as chemical characteristics and properties of the soil for

examples dissolvable aluminum contained in sludge, soil pH, soil redox potential, organic matter contained in soil, soil texture/soil acidity and the volume of sludge added into the soil, etc. (World Health Organization, 1997).

Aluminum is normally in nontoxic insoluble form in soil, under acidic condition (such as acidic soil) the acidic condition enhances the insoluble-form aluminum to be soluble and leach out in toxic form effect to plants and agronomic corps and this will be more severe when pH value less than 5 (Lindsay, 1979).

It is known that aluminum is toxic to plant growth. However, there are more studies on effects of aluminum on humans. There are reports on the effect of aluminum on kidneys and the possibility of aluminum toxicity associated with Alzheimer (World Health Organization, 1997), memory impairment, and neurological diseases (Nayak, 2002). By these reasons, this study aims to investigate the potential tropical environmental factors on the leachability of aluminum from water treatment sludge in soil. If the results show any potential factors that enhance aluminum leachability from sludge into the environment by landfilling, the landfill site should be required to select carefully the sludge or prior treatment of sludge before disposing to avoid risk of contamination due to aluminum leaching.

### **Objectives of the Study**

The overall objectives of the study are:

1. To investigate the potential environmental factors affecting aluminum leachability from water treatment sludge from the Metropolitan Water Work Authority, MWA (Bangkhen water treatment plant).
2. To study the effect of soil acidity on aluminum leaching.

### **Scopes of the Study**

1. The water treatment sludge samples from Bangkok water treatment plant were analyzed to evaluate aluminum content.
2. The environmental factors studied were tropical climate factors such as soil texture (soil pH), temperature, soil moisture content, rain pH, etc.
3. The experiments were conducted at Laboratory of Environmental Engineering Department, Kasetsart University, Bangkok, Thailand.
4. Batch leaching experiments were conducted to investigate effect of soil acidity, temperature, and soil moisture content on aluminum leaching.
5. Column leaching experiments of soils, sludge and amended sludge samples were conducted to study the potential leachability of aluminum and its remobilization after amended with soils.

## LITERATURE REVIEW

### **1. The Metropolitan Waterworks Authority (MWA)**

King Rama V, in his concern about clean water supply for the Thai people, had established the waterworks in Thailand. The first water work was founded in Bangkok and could supply clean water to the Bangkok household in early 1914 under the King Rama VI era, the water work was known as 'Karn Prapa Krungthep'. On August 16, 1967, the Bangkok Water Supply became a state enterprise organization and changed its name to Metropolitan Waterworks Authority, (MWA). MWA's missions are to search for raw water sources used in tap water production, produce and supply clean tap water in Bangkok, Nonthaburi and Samutsakorn area, and perform works relating and benefiting MWA. (MWA, 2004).

Today, MWA has four water treatment plants (Bangkhen, Thonburi, Samsen, and Mahasawat) with production capacity of 4,660,000 m<sup>3</sup>/day and produces 4,015,000 m<sup>3</sup>/day of potable water (MWA, 2004). Amount of clean water production of each water treatment plant are shown in Table 1. Drinking water quality standard of MWA is show in Table 2. Locations of the water treatment plants are shown in Figure 1. A flow chart of water production and distribution systems are shown in Figure 2 and the general concepts of the water treatment are illustrated in Figure 3.

Among the four water treatment plants, Bangkhen plant is the biggest with the highest production capacity and also produces large amount of water treatment sludge each year. The Bangkhen water treatment plant produces water and supplies most of Bangkok metropolitan area (29 districts): Don Muang, Bangkhen, Nonthaburi, Pakgred, Bangsue, Chatuchak, Praya Thai, Din Daeng, Huay Kwang, Pra Nakorn, Pom Prab Satrupai, Sampanthawong, Bangrak, Pratumwan, Sathorn, Bangkorlaem, Yanawa, Klongtoey, Suan Luang, Lad Prao, Bangkok Yai, Bueng-gum, Ladgrabang, Pravet,Prakhanong, Prapadaeng, Samutprakarn, Bangkok Yai, Ratburana and Jomtien (MWA, 2004).

The waste from these water treatment plants are sludges. Sludge from Bangkok water treatment plant solely is about 126,000 tons/year (Keawruenrom et al., 2001).

Table 1 Water production from MWA water treatment plants

Water Treatment Plant	Water Production/day (Million cu.m.)
Bangkok	3.20
Samsen	0.70
Thonburi	0.17
Mahasawat	0.80

Source: MWA (2004)

Table 2 Drinking water quality standard of MWA

Parameters	Units	Guideline Value
1. Bacteriological quality		
- Total coliform bacteria	Presence-Absence/100 ml	None
- E. coli	Presence-Absence/100 ml	None
2. Physical & Chemical Property		
- Apperance color	True colour unit	15
- Turbidity	NTU	5
- Taste and odor	-	Not offensive
- Arsenic	mg/l	0.01
- Cadmium	mg/l	0.003
- Chromium	mg/l	0.05
- Cyanide	mg/l	0.07
- Lead	mg/l	0.01

Table 2 (Continued)

Parameters	Units	Guideline Value
- Mercury	mg/l	0.001
- Selenium	mg/l	0.01
- Fluoride	mg/l	1.5
- Chloride	mg/l	250
- Copper	mg/l	1
- Iron	mg/l	0.3
- Manganese	mg/l	0.1
- Aluminium	mg/l	0.1
- Sodium	mg/l	200
- Sulfate	mg/l	250
- Zinc	mg/l	3
- Hydrogen sulfide	mg/l	0.05
- Total dissolved solids	mg/l	1,000
- Nitrate as NO <sub>3</sub> <sup>-</sup>	mg/l	50
- Nitrite as NO <sub>2</sub> <sup>-</sup>	mg/l	3
- Free residual chlorine	mg/l	> 0.2
- Benzene	μg/l	10
- Carbon Tetrachloride	μg/l	2
- Dichloromethane	μg/l	20
- 1,2-Dichloroethane	μg/l	30
- Benzo [a] pyrene	μg/l	0.7
-Microcystin-LR	μg/l	1
3. Pesticides		
- Aldrin/Dieldrin	μg/l	0.03
-Chlordane	μg/l	0.2
- DDT	μg/l	2
-2,4-D	μg/l	30

Table 2 (Continued)

Parameters	Units	Guideline Value
- Heptachlor and Heptachlor epoxide	$\mu\text{g/l}$	0.03
- Hexachlorobenzene	$\mu\text{g/l}$	1
- Lindane	$\mu\text{g/l}$	2
- Methoxychlor	$\mu\text{g/l}$	20
- Pentachlorophenol	$\mu\text{g/l}$	9
4. Trihalomethanes (sum of the ratio)	-	1
- Chloroform ( $\text{CHCl}_3$ )	$\mu\text{g/l}$	200
- Bromodichloromethane ( $\text{CHBrCl}_2$ )	$\mu\text{g/l}$	60
- Dibromochloromethane ( $\text{CHBr}_2\text{Cl}$ )	$\mu\text{g/l}$	100
- Bromoform ( $\text{CHBr}_3$ )	$\mu\text{g/l}$	100
5. Radioactive		
- Gross alpha activity	Bq/l	0.1
- Gross beta activity	Bq/l	1

Source: MWA (2004)

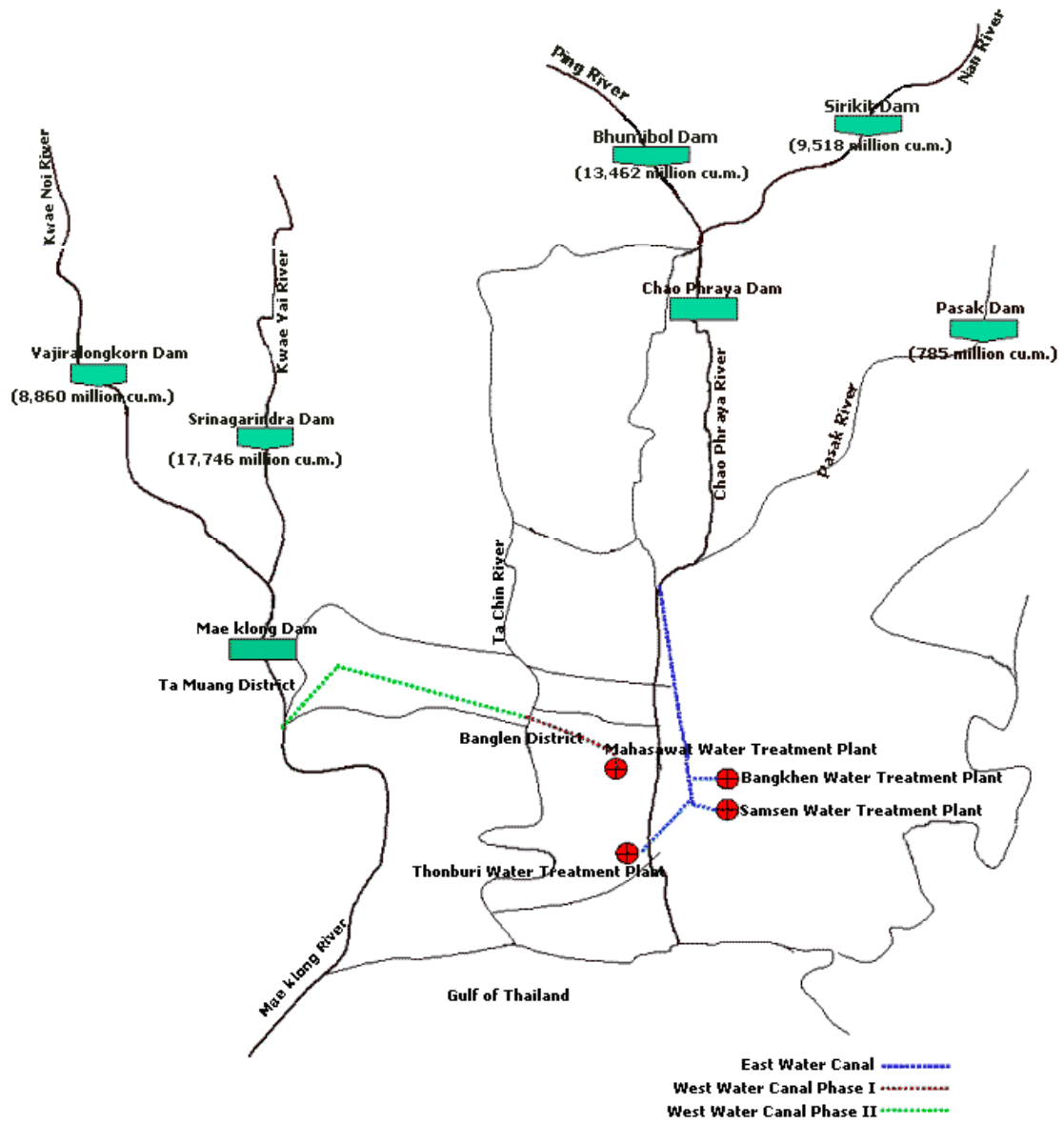


Figure 1 Location of MWA water treatment plants

Source: MWA (2004)

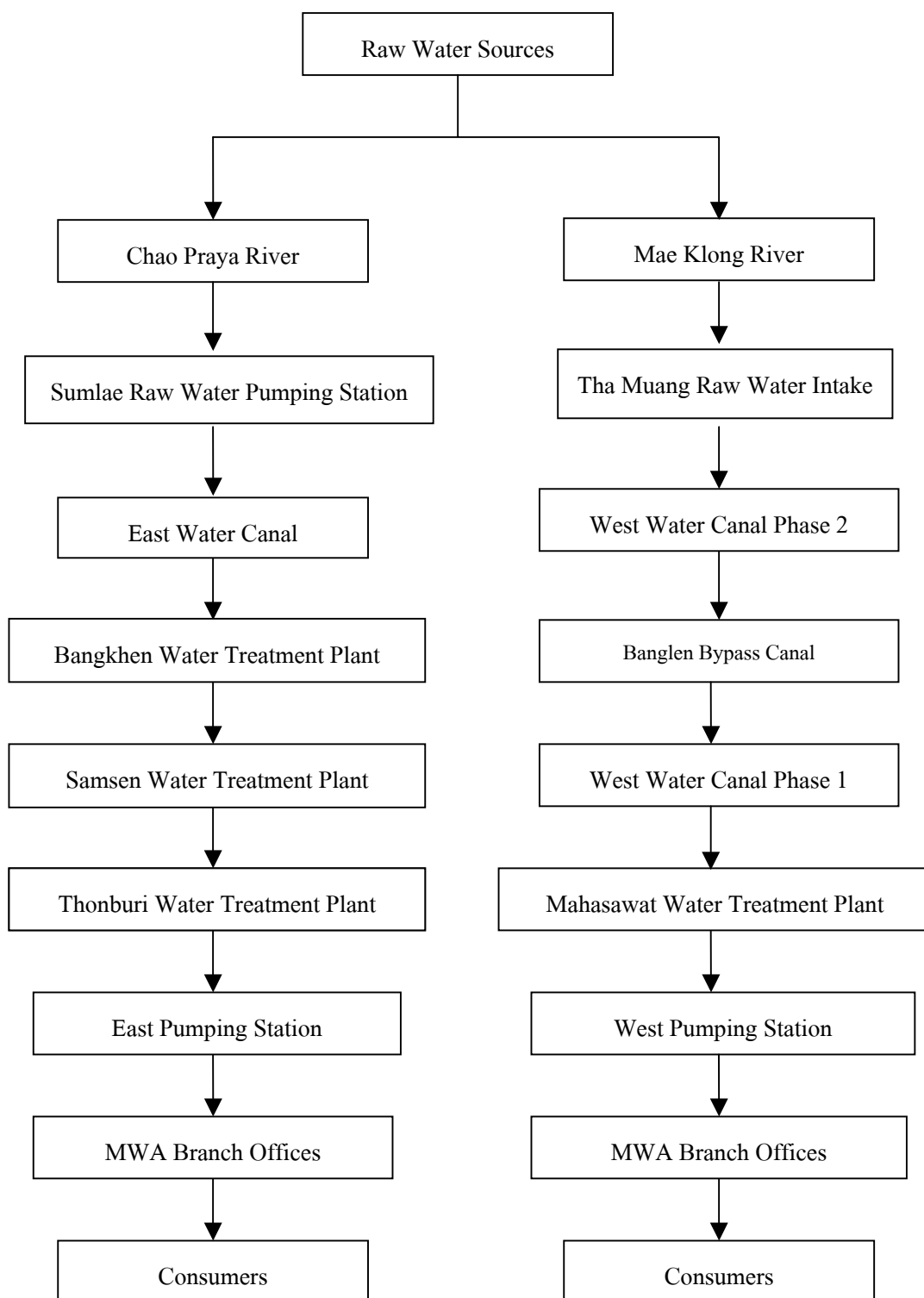


Figure 2 MWA tap water production and distribution system

Source: MWA (2004)

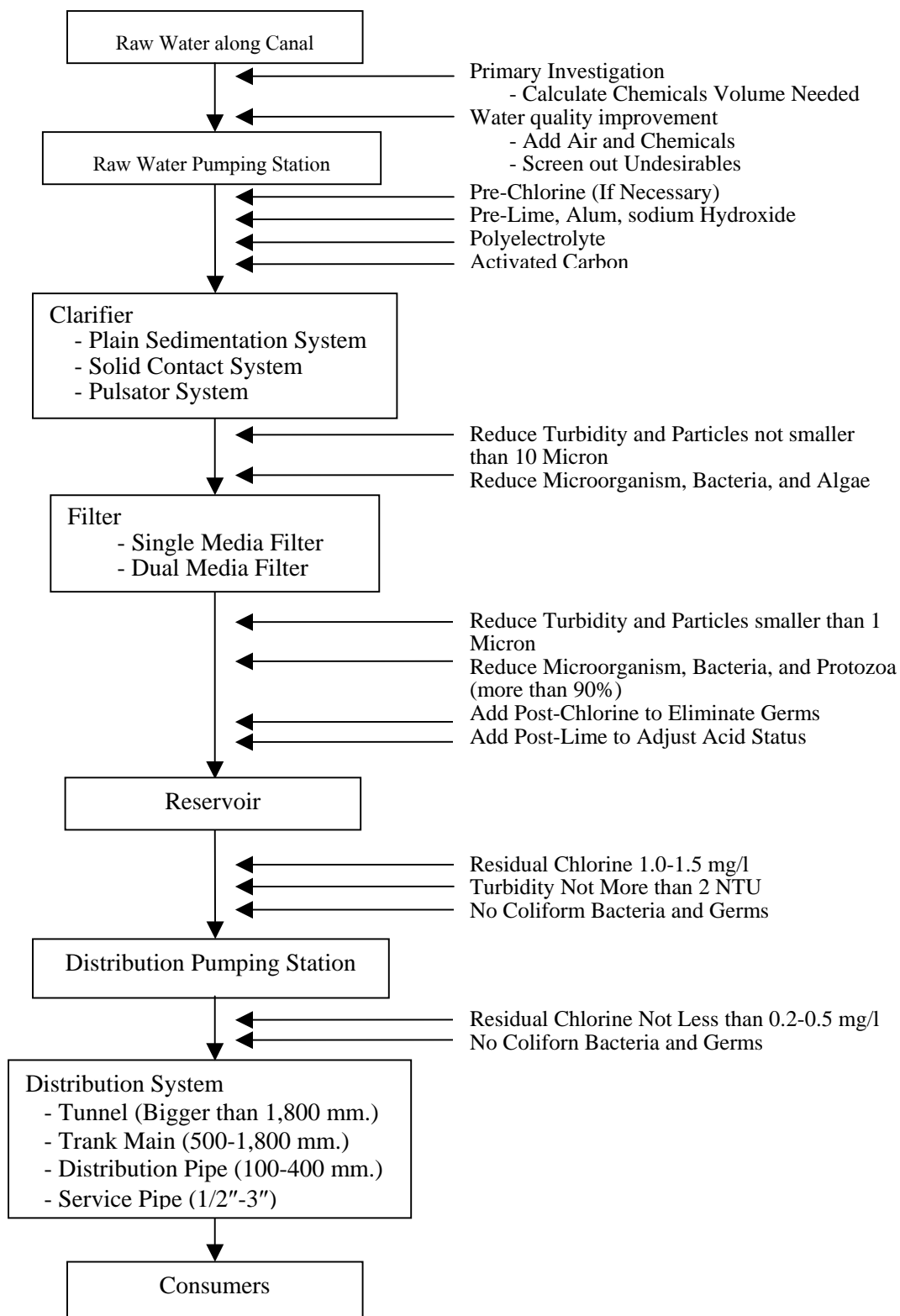


Figure 3 General concepts of the water treatment

Source: MWA (2004)

## **2. Sludge**

Sludge is semi-solid sediment produced in sedimentation process or other turbidity removal process of water treatment. Sludge property varies from place to place depends on characteristics of raw water brought into the system. Different raw water characteristics require different type and quantity of chemical reagent used in water treatment process.

Quality of surface water can be assessed by chemical-based parameters, including biological oxygen demand (BOD), and by biological indicators that take into account the presence of indicator organisms and the diversities of life found in water. A 1975 EU Directive concerning the quality of surface water intended for the abstraction of drinking water in the Member States (75/440/EEC) classifying surface waters into three classes. It specified that;

- Category A1: water requires a simple physical treatment and disinfection
- Category A2: water requires a normal physical treatment, chemical treatment, and disinfection
- Category A3: water requires an intensive physical and chemical treatment, extended treatment, and disinfection (Binnie et al., 2002).

### **2.1. Sludge type**

Sludge can be divided into 2 main types;

1. Coagulant or polymeric sludge: sludge from turbidity removal process (coagulation and flocculation). This type of sludge contains hydroxide of coagulant used, organic matters and solids separated from water by coagulation and flocculation process.
2. Softening sludge: sludge from softening process (process reduces calcium and magnesium components from raw water). This type of sludge contains calcium carbonate and

other components, such as magnesium hydroxide, lime, organic matters, etc. (Ruengtananrak, 2004)

## **2.2. Sludge Disposal**

Landfilling or land application is one of the most common disposal methods for water treatment sludge. This method allows sunlight and soil microorganisms to biodegrade the organic matters while the soil binds up the metals contains in the sludge (New Zealand Water and Wastes Association, 2005).

Hence, it is important to know characteristics of the sludge to help making decision on selection the proper sludge disposal method and treatment to assure that the disposal of sludge will not cause environmental or health problems.

## **2.3. MWA Water Treatment Sludge Characteristics**

MWA water treatment plants use raw water from Chao Praya River and Mae Klong River resources (changed from Chao Praya River and Tha-jeen River since March 2002) (MWA Quality Control Division, 2004).

In a production process, MWA uses alum to flocculate suspended solids, thus the sludge produced has an average aluminum content of about 124 g/kg soil. The sludge disposal method is landfilling (Keawruenrom et al., 2001).

Sludge characteristics consist of physical, chemical and biological properties as described as follows;

### **2.3.1. Physical Properties**

The physical compositions of sludge, such as specific gravity or particle density, viscosity, particle size distribution (suspended matter, colloidal, total solids, dissolved solid, etc.), degree of hydration, and compressibility are principally depending on raw water properties and treatment process being used.

In general, dry MWA water treatment sludge is hard to extremely hard while in moisten condition it is very firm, very sticky and very plastic in wet condition. These show that the sludge has fine to very fine particles consistency. The sludge is silty clay sediments and quite homogeneous. (Keawruenrom et al., 2001).

The physical properties of Bangkhen water treatment sludge are reported in Table 3.

### **2.3.2. Chemical Properties**

The chemical composition of the sludge depends on the raw water characteristics and treatment process. The chemical properties of sludge are important regarding a potential leachability of aluminum into the soil, aluminum toxicity and the sludge disposal and utilization alternatives.

Bangkhen water treatment sludge has total aluminum content in range of 119-126 g/kg (Table 4). The sludge is slightly acidic with pH value in range of 6.3-6.5, organic matter in range of 41.1-47.3 g/kg. Result from chemical properties analysis of the sludge shows that the sludge consists of plant nutrients such as nitrogen, phosphorus, and potassium (Table 5).



**Table 4** Previous studies of total chemical analysis of Bangkok water treatment sludge

Sludge	Si	Al	Ca	Mg	Na	K	Ti	Fe	Mn	P	S	Cu	Zn
Sample	(-----g/kg-----)							(-----mg/kg-----)					
Basin A, B, C and E, first sampling													
Basin A													
Range	250 - 254	123 - 126	4	6-7	2	16	4	47	777- 911	881- 907	868- 935	30- 32	108- 111
Basin B													
Range	251 - 253	121 - 124	5	6-7	2	15	4	48	870- 982	982- 106	869- 930	26- 32	98- 110
Basin C													
Range	250 - 253	121 - 124	5	6-7	2	15- 16	4	47- 48	824- 937	947- 104	826- 913	24- 32	99- 107
Basin E													
Range	252 - 255	123 - 124	5-6	7	2	15- 16	4	46- 48	810- 849	906- 954	841- 931	28- 33	104- 111
Basin D and E, second sampling													
Basin D													
Range	255 - 262	119 - 122	4-5	7	2	16	4	45- 46	778- 832	851- 901	845- 107	31- 33	105- 109
Basin E													
Range	251 - 259	123 - 125	5- 11	7	2	16	4	46- 48	799- 846	897- 931	868- 914	30- 35	109- 111

Source: Keawruenrom et al. (2001)

**Table 5** Chemical properties of Bangkhen water treatment sludge

Chemical properties	Range
pH	6.3-6.5
Organic Matter (g/kg)	41.1-47.3
Total Nitrogen (g/kg)	1.08-1.35
Available Phosphorus (mg/kg)	11.50-12.75
Available Potassium (mg/kg)	77-225
Cation Exchangable Capacity, CEC (cmol/kg)	20.3-22.8
Total Aluminum (g/kg)	117-124
Extractable Aluminum (cmol/kg)	0.03 (0.033 ppm)
Electrical Conductivity, EC (dS/m)	0.81-0.99

Source: Sengsri (2003)

### 2.3.3. Biological Properties

The prime objective of water treatment is to produce clean water free from pathogenic microorganisms. Most biological compositions in sludge are fungi, protozoa, bacteria, and algae from raw water. The most important biological characteristic of sludge is the presence of pathogenic organisms which are harmful to humans and animals when exposed to the sludge.

### 3. Alum

Alum or aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3$  or  $\text{Al}_2(\text{SO}_4)_3 \cdot 14\text{H}_2\text{O}$  or  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , Figure 4) is a white crystalline compound used in water purification process to remove small particles suspended in the water (Pickett et al., 2000). The alum draws small particles together to form floc which settles to the bottom of the tank. The settled floc called sludge.

Chemical name of alum is aluminum sulfate hexadecahydrate with molecular weight of 630.4 g/mol. Alum is in solid phase and soluble in water (Table 6).



Figure 4 Aluminum sulfate crystal

Table 6 Properties of aluminum sulfate

	General
Chemical name	Aluminium sulfate hexadecahydrate
Other names	Cake alum Filter alum Papermaker's alum alumogenite Aluminum sulfate Aluminium sulphate
Molecular formula	$\text{Al}_2(\text{SO}_4)_3 \cdot 14\text{H}_2\text{O}$ , $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$
Molecular weight	630.40 g/mol
Appearance	white crystalline solid

Table 6 (Continued)

General	
Chemical name	Aluminium sulfate hexadecahydrate
Density and phase	2.71 g/cm <sup>3</sup> , solid
Solubility in water	31.3 g/100 ml (0 °C)
In ethanol	Slightly soluble (anhydrate) Insoluble (hydrate)

Source: Wikipedia (2005)

When adding alum into water, the reaction of alum with water is as follows:



This simplified reaction shows that as alum is added to the water it will react with the alkalinity and produce aluminum hydroxide (Graham et al.,1960), but in the actual treatment process, equilibrium does not reached and various intermediates are formed. However, in the pH range of 6-8, in which most plants coagulate, probably the insoluble aluminum hydroxide complex of  $\text{Al}(\text{H}_2\text{O})_3(\text{OH})_3$  predominates (Borchardt et al., 1981).

In water treatment plant, sludge is most commonly produced in coagulation process. Aluminum sulfate or alum is generally used to accomplish coagulation. Alum sludge leaving the sedimentation basin generally has suspended solid content of 1%. Around 20-40% of the solids are volatile and the remainders are inorganic clays or silts. The BOD<sub>5</sub> of alum sludge is usually 100 mg/l. However, the chemical oxygen demand (COD) of the sludge is considerably higher. The pH of alum sludge is generally in the 5.5-7.5 range. Alum sludge from the sedimentation basin may include large numbers of microorganisms, but generally does not exhibit an unpleasant odor (Borchardt et al., 1981).

Alum sludge appearance is depending on solid concentration. Low solid concentration has liquidous sludge appearance while high solid concentration has solid appearance as shown in Table 7.

In general, alum sludge has pH value in range of 6-8 with 0.1-4.0% total solids, 15-25% organic matters, and aluminum (in form of  $Al_2O_3$ ) 15-40%. Chemical properties of the sludge are shown in Table 8. However, physical and chemical properties of alum sludge may vary from place to place depend on characteristics of water resource. Physical and chemical characteristics of wet alum sludge from different water resource are reported in Table 9.

Table 7 Alum sludge appearance

Solid Concentration in Sludge (Weight %)	Alum Sludge Appearance
0-10	Liquid
10-15	Viscous Liquid
15-20	Pasty
20-25	Semi-Solid
25-30	Soft Solid
30-50	Crumbly Cake

Source: Kawamura (2000)

**Table 8** Chemical properties of alum sludge

BOD (mg/l)	COD (mg/l)	pH	Total Solids (%)	Al <sub>2</sub> O <sub>3</sub> (%)	SiO <sub>2</sub> and Inert (%)	Organic Matter (%)
30-300	30-5,000	6-8	0.1-4.0	15-40	35-70	15-25

Source: Kawamura (2000)

**Table 9** Physical and chemical characteristics of wet alum sludge

Location	Water Source	Suspended Solids (%)	BOD <sub>5</sub> (mg/l)	COD (mg/l)	Specific Resistance (m/kg)
General	-	0.3-1.5	30-100	500-10,000	-
General	-	0.2-2.0	-	30-5,000	$20 \times 10^{12}$
Milwaukee, WI	Lake	0.78	-	-	$5 \times 10^{12}$
Auburn, AL	-	0.19	-	-	$5.5 \times 10^{12}$
Unknown	-	0.59	-	-	$55 \times 10^{12}$
Rochester, NY	Lake	0.36	36-77	500-1,000	-
San Francisco Bay, CA	Reservoir	1.0	100	2,300	-
Erie County, MI	Lake	0.16	-	-	-
Moberly, MO	Reservoir	-	-	-	$16 \times 10^{12}$
Unknown	-	0.6-1.5	-	-	$3.0 \times 10^{12}$
Unknown	-	-	-	-	$40 \times 10^{12}$
Radford, VA	River	3.0	-	-	$1.6 \times 10^{12}$

Table 9 (Continued)

Location	Water Source	Suspended Solids (%)	BOD <sub>5</sub> (mg/l)	COD (mg/l)	Specific Resistance (m/kg)
Timbersville, VA	River	1.4	-	-	$5.4 \times 10^{12}$
Harrisonburg, VA	River	2.1	-	-	$3.6 \times 10^{12}$
Blacksburg, VA	River	3.1	-	-	$4.1 \times 10^{12}$

Source: Borchardt et al. (1981)

Dry sludge contains aluminum at least 20% by weight. Therefore, this should be raises some concern about leaching of aluminum when sludge is disposed in a landfill.

#### **4. Aluminum**

Aluminum or aluminum is the third most abundant element in the earth's crust, ranking behind oxygen and silicon (Table 10) (Press and Siever, 1974). About 8.1% of the earth's crust is composed of aluminum. In form of compound, aluminum compound is the fourth most abundant compound in earth's crust (6.99% by weight) as shown in Table 11).

Table 10 List of the eight most abundant elements in the earth's crust

Element	Approximate % by weight
Oxygen	46.6
Silicon	27.7
Aluminum	8.1
Iron	5.0
Calcium	3.6
Sodium	2.8
Potassium	2.6
Magnesium	2.1
All others	1.5

Source: HyperPhysics (2005)

**Table 11** Ten most abundant compounds in the earth's crust

Compound	Formula	Abundance percent by weight	Abundance parts per million by weight
Silicon dioxide	SiO <sub>2</sub>	42.86%	428,600
Magnesium oxide	MgO	35.07%	350,700
Ferrous oxide	FeO	8.97%	89,700
Aluminum oxide	Al <sub>2</sub> O <sub>3</sub>	6.99%	69,900
Calcium oxide	CaO	4.37%	43,700
Sodium oxide	Na <sub>2</sub> O	0.45%	4,500
Ferric oxide	Fe <sub>2</sub> O <sub>3</sub>	0.36%	3,600
Titanium dioxide	TiO <sub>2</sub>	0.33%	3,300
Chromic oxide	Cr <sub>2</sub> O <sub>3</sub>	0.18%	1,800
Manganese dioxide	MnO <sub>2</sub>	0.14%	1,400

Source: Heiserman (1992)

Chemical symbol for aluminum is Al. It is a silvery-white metal with a face-centered cubic crystalline structure. Aluminum has atomic number 13, atomic mass 26.981538, and valence 3. Melting point of aluminum is 1,220°F (660°C), boiling point is about 4,440°F (2,450°C), and density 2.708 g/cm<sup>3</sup>. Aluminum is both ductile and malleable with a very good electrical conductivity. Pure aluminum is a silvery-white metal. It is remarkable for oxidation resistance due to the passivation phenomenon (Columbia University Press, 2005; Lange, 1952). Aluminum properties are shown in Table 12.

Table 12 Properties of aluminum

General	
Name, Symbol, Number	Aluminum, Al, 13
Chemical series	poor metals
Appearance	Silvery
Atomic mass	26.981538(2) g/mol
Electron configuration	[Ne] 3s <sup>2</sup> 3p <sup>1</sup>
Electrons per shell	2, 8, 3 x
Physical properties	
Phase	solid
Density (near room temperature)	2.70 g/cm <sup>3</sup>
Liquid density (at melting point)	2.375 g/cm <sup>3</sup>
Melting point	933.47 K (660.32 °C, 1220.58 °F)
Boiling point	2792 K (2519 °C, 4566 °F)
Heat of fusion	10.71 kJ/mol
Heat of vaporization	294.0 kJ/mol
Heat capacity	(25 °C) 24.200 J/(mol·K)
Atomic properties	
Crystal structure	cubic face centered
Ionization energies (more)	1 <sup>st</sup> : 577.5 kJ/mol 2 <sup>nd</sup> : 1816.7 kJ/mol 3 <sup>rd</sup> : 2744.8 kJ/mol
Atomic radius	125 pm

Source: Wikipedia (2005)

Typical range of aluminum in soil is from 1 to 30% (10,000 to 300,000 mg Al/kg) (Lindsay, 1979; Dragun, 1988). Although aluminum is abundant, it is never found as pure metal but as an important constituent of many minerals (Chutima, 1980).

In nature, aluminum occurs as compounds found primarily in a form of aluminum silicates and oxides. Aluminosilicate occurs as feldspars, mica, and clay minerals. Aluminum oxide (alumina) occurs as corundum and emery (Hesse, 1972; McLean, 1965). Aluminum in rocks and soil is normally in nontoxic insoluble form. However, under acidic condition aluminum becomes soluble and dissolves into water. Aluminum mobilization increases as soil pH decrease (Lindsay, 1979).

The main aluminum ore is bauxite (typically,  $\text{Al}_2\text{O}_3 \cdot 2\text{H}_2\text{O}$ ) which is a source of over 99% of metallic aluminum. Bauxite is a mixture of hydrated aluminum oxides such as gibbsite ( $\text{Al}(\text{OH})_3$ ), diaspore ( $\text{AlO}(\text{OH})$ ), and boehmite ( $\text{AlO}(\text{OH})$ ) and usually containing oxides of iron and silicon in varying quantities. Bauxite is claylike and earthy and ranges in color from white to deep brown, reddish-brown, tan, tan-yellow, or red according to the nature and quantity of its components as shown in Figure 5 (Columbia University Press, 2005).



Figure 5 Bauxite

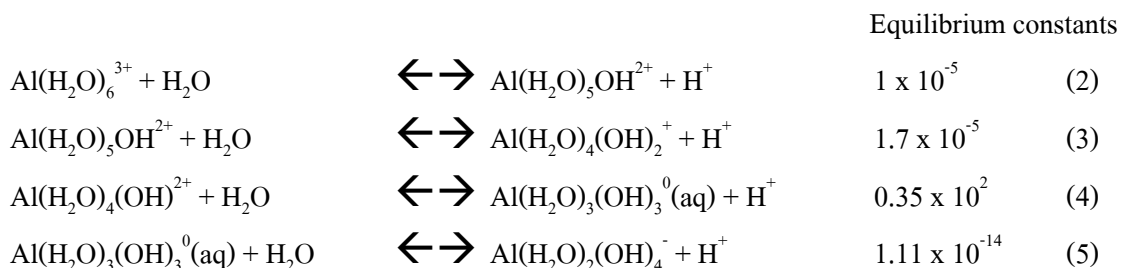
Source: Mineral Information Institute (2005)

Weak acids such as dilute  $\text{H}_2\text{CO}_3$  can decompose the silicate and montmorillonite layers resulting in releasing of aluminum (Jackson, 1963). Thus, natural decomposition of organic matter and minerals in soil enhances the weathering of rocks releasing aluminum into soil solution (McBride, 1994).

#### 4.1 Species of Aluminum in Soil Solution

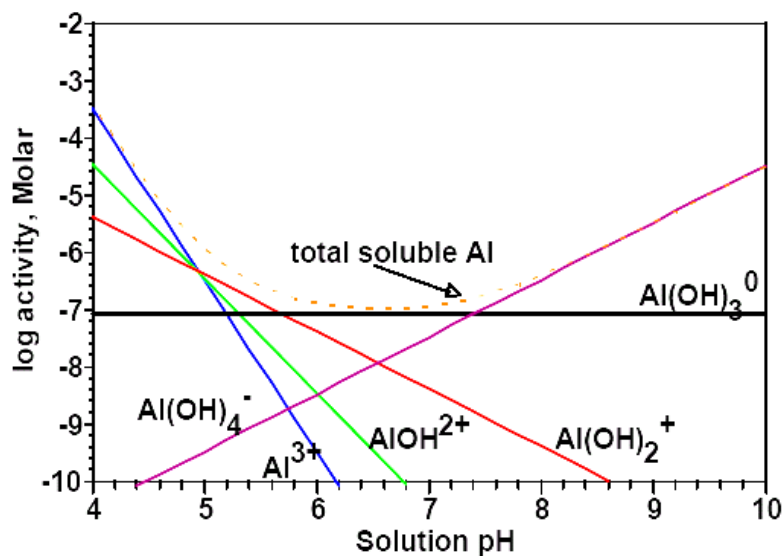
In soil solution, hydrated aluminum ion ( $\text{Al}(\text{H}_2\text{O})_6^{3+}$ ) is an acid as it contains hydrogen ions removable from the six water molecules ( $-\text{OH}_2$ ) surrounding the aluminum. The  $\text{Al}(\text{H}_2\text{O})_6^{3+}$  occurs in acid solutions at pH 5.0 and below (Jackson, 1963; McLean et al., 1965).

When  $\text{Al}(\text{H}_2\text{O})_6^{3+}$  hydrolyzes with water molecule, it will release proton (hydrogen ion). The steps of proton dissociation of hydrated aluminum ion in dilute solution may be represented by the following equations (Jackson, 1963; Black, 1968; Lindsay, 1979; McBride, 1994; Bohn, 1979):



The concentrations of these species as a function of pH are shown in Figure 6.

The  $\text{Al}(\text{H}_2\text{O})_6^{3+}$  is normally written without water molecules as  $\text{Al}^{3+}$ .



**Figure 6** Solubility diagram of the most significant species of aluminum in aqueous solution of  $\text{AlCl}_3$

Source: McBride (1994)

When soil pH is lowered much below 5.5, aluminosilicate clays and aluminum hydroxide minerals begin to dissolve, releasing aluminum-hydroxy cations and  $\text{Al(H}_2\text{O)}_6^{3+}$ . Therefore, as the pH is lowered, the concentration of soluble aluminum increases (McBride, 1994).

#### 4.2 Effects of Aluminum on Environment

When aluminum is mobilized in lakes and streams, it has been shown to be toxic to aquatic life. Aluminum forms compounds with organic and inorganic ligands that are soluble in acidic and basic solutions. The acidic extracts were most toxic. Aluminum appeared to be directly toxic to algae, as opposed to indirectly inhibiting algal growth by phosphate removal (George et al., 1991). Therefore, aluminum in water could harm the balance of aquatic ecology.

Aluminum is not needed for plant growth and in acid soils it can be toxic to plants (Bates and Johnston, 1991). For plants, it is an important growth-limiting factor in acidic soils (Foy et al., 1999).

At low soil pH, aluminum solubility increases and this can result to limited plant growth (Delhaize, 2004). Under acidic conditions, aluminum is more soluble and at high aluminum concentration levels the aluminum will damages root system by shorten and swollen it (Hue et al. 2004).

Aluminum toxicity commonly occurs in Oxisols (very highly weathered soils found primarily in the intertropical regions of the world containing not more than 10% weatherable minerals and less than 10% base saturation with high concentration of iron (III) and aluminum oxides and hydroxides (McDaniel, 2006; Wikipedia, 2005)) and Ultisols (strongly leached, acid forest soils with relatively low native fertility found primarily in humid temperate and tropical area of the world containing less than 10% weatherable minerals in the extreme top layer of soil and less the 35% base saturation throughout the soil (McDaniel, 2006; Wikipedia, 2005)) as well as in other heavily leached soils such as lateritic soils of the humid tropics. It is an important growth limiting factor on upland soils with a pH of less than 5 (Delhaize, 2004). Relationship between soil pH and percent saturated aluminum in soil is shown in Figure 7.

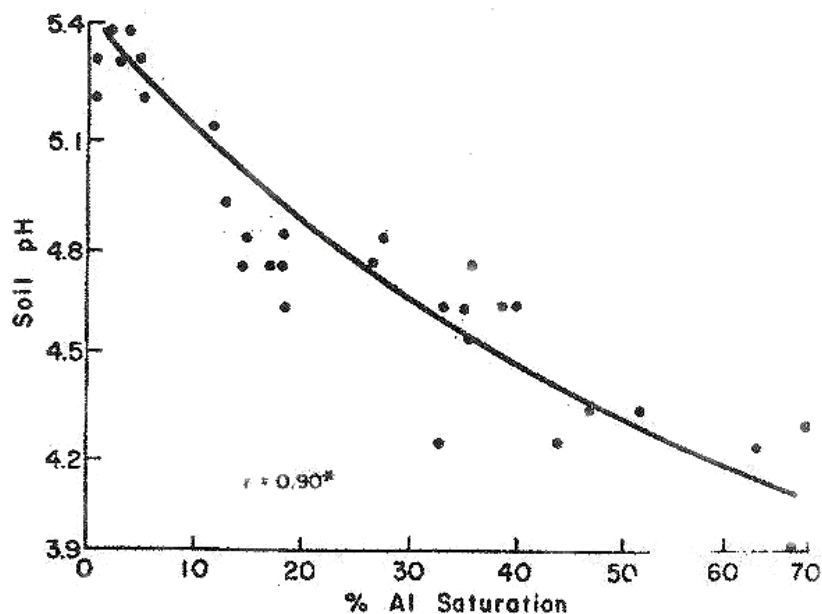


Figure 7 Relationship between soil pH and percent saturated aluminum in Oxisols and Ultisols in Puerto Rico

Source: Abruna et al. (1975)

## 5. Aluminum Toxicity

Even if not all forms of aluminum are toxic however, the soluble forms such as divalent, monovalent and trivalent are toxic (Mengel and Kirkby, 1987).  $\text{Al}^{3+}$  is a dominant species for aluminum hybridization in solution under acidic condition, while  $\text{Al}(\text{OH})^{2+}$  and  $\text{Al}(\text{OH})_2^+$  species are dominants at pH between 5 and 7 (Delhaize, 2004).

Aluminum toxicity has been linked to soil pH by considering the amount of soluble aluminum rather than the total aluminum concentration in the soil (Jackson, 1963; McLean et al; 1965). Aluminum toxicity generally occurs in soils below pH 5.5 (Watcharothayan et al., 1984) and becomes particularly severe at lower pH values, when the solubility of aluminum increases sharply and dissolved in monomeric forms (predominantly  $\text{Al}^{3+}$  and  $\text{Al}(\text{OH})^{2+}$ ) (Matsumoto et al, 1980). Effect of aluminum toxicity is known to pose a serious agricultural problem throughout the

world and it has been implicated as one of the major causes of forest decline in Europe and North America. And in natural water, aluminum ion also causes acute toxicity to many aquatic organisms (Wang, 2005).

### 5.1 Toxic Effect on Plants

Plant roots are sensitive to  $Al^{3+}$  ions that occur in solution in acidic soils (Bennet and Breen, 1991). Aluminum toxicity is the primary factor limiting plant growth on acidic soils. Toxicity of aluminum may interfere with the acquisition and metabolism of mineral nutrients, disrupt cell membranes, and alter enzyme activities (McLean and Gilbert, 1927; Matsumoto et al., 1976).

High aluminum concentration in soil solution can lead to toxicity to the plant root system and reduced nutrient uptake by forest vegetation. Poorly plant growth in very acid soils ( $pH < 5$ ) is generally affected by aluminum toxicity, which causes plant root to become short, thick, and stubby (Nosko et al. 1988). Aluminum is highly toxic to the growing region of root tips but has little effect on other parts of the root (Fleming and Foy, 1968).

Aluminum accumulates principally in the root tips of the main root and lateral root tissue, with small quantities in the cortex and epidermal cells. Monomeric forms of aluminum prohibit cell fission (Matsumoto et al., 1976). The accumulation of aluminum in root tips at sites of cell division and cell elongation inhibits root growth that causes root damaging such as stunning of root growth and interfere the plants ability to absorb necessary nutrients. This can slow down overall of plant growth rate. Long-term effect of plant exposed to aluminum is shoot growth inhibiting because of nutrient (Mg, Ca, P) deficiency, and phytohormone imbalances (McLean and Gilbert, 1927).

Effect of aluminum on plant's root growth is shown in Table 13 and effect of aluminum concentration on root length is shown in Table 14.

**Table 13** Effect of aluminum on maize and millet root growth in Puerto Rico

Soil Type	pH value	Exchangeable Al		Saturated Al %	Root Weight (mg/pot)	
		meq/100 g	ppm <sup>a</sup>		Maize	Millet
Humatas	4.8	4	4.44	40	931	400
	4.5	6	6.67	57	874	296
	3.9	11	12.22	87	209	19
Coto	4.8	3	3.33	52	687	345
	4.5	4	4.44	70	630	126
	4.0	5	5.56	87	389	128

Source: Brenes and Pearson (1973)

<sup>a</sup>: converted by the author of this thesis

**Table 14** Effect of aluminum concentration on plant's root length

Aluminum toxicity reduces root growth	
Al (ppm)	Cotton Root Length
0	16 cm
0.25	11 cm
0.50	8 cm

Source: Clemson Crop and Soil Environmental Science Extension (1999)

At pH 5.0 and lower, root injury is observed at pH 5.0 and lateral root development is suppressed (Hutchinson et al. 1986). In some cases root tips are killed. The roots become discolored brown or a dull gray (McLean and Gilbert, 1927). At these pH levels, high aluminum

concentration suppress cations (Ca and Mg) uptake. At low pH and low Ca concentrations, damage to root membranes is accentuated (Long and Foy, 1970; MacLean and Chiasson, 1966).

A soil with potential aluminum toxicity has aluminum saturation over 30%, soil pH (H<sub>2</sub>O) less than 5.0, and aluminum in soil solution more than 1-2 mg Al/l (Dobermann and Fairhurst, 2000). The toxic effects of aluminum to plants are observed in association with soluble aluminum (Al<sup>3+</sup>) (van Breemen et al., 1989).

## **5.2 Toxic Effects on Aquatic Life**

It has been determined that fish tend to be more sensitive to aluminum toxicity than aquatic invertebrates (Sparling et al., 1997; Hunter and Tannahill, 1980). Aluminum can be acutely toxic to fish at pH levels that are not normally considered toxic to humans. Aluminum concentration as low as 6.2 mg/l is known that it is able to kill fish.

Effects of aluminum toxicity on aquatic life are shown in Table 15.

There are two ways in which aluminum become harmful to fish. First, to reduce the ions exchange through the gills and subsequently causes a salt depletion. Aluminum also precipitates in the gills and interfere oxygen and other ions transportation, so that the fish literally dies of suffocation. Second, the fish will release mucus against the aluminum in their gills. The building up of this mucus will clog the gills and prohibit oxygen and salt transportation (Thoreau, 2005).

**Table 15** Effect of aluminum toxicity on aquatic life

Tested Animal	Water Type	pH value	Total Al $\mu\text{g/l}$	Al-Speciation	Effect
Brook trout	Synthetic	5.20	20	Free, monomeric	Mean survival 99%
<i>Salvelinus fontinalis</i>	Soft water	5.20	420	ditto	Mean survival 28%
	Soft water	4.40	480	ditto	Mean survival 42%
	Soft water	5.20	500	fluoride	Mean survival 45%
Brow trout	Tap				
<i>Yolk sac fry</i>	0.5 mg Ca/l	5.40	500		Survival about 30%
<i>Salmo trutta</i>	2 mg Ca/l	5.40	500		Survival 0%
	2 mg Ca/l	5.40	250		Survival over 90%
	0.5 mg Ca/l	5.40	250		Survival about 20%
White sucker					
<i>Embryo</i>	Tap	5.2	160	As monomeric 140	Hatching 0%
<i>Larvae</i>	Tap	5.4	40	As monomeric 50	Survival 0%
<i>Postlavae</i>	Tap	5.0	90	As monomeric 80	Survival 11%
<i>Postlavae</i>	Tap	5.4	70	As monomeric 60	Survival 80%
Gold fish	Hard	5.5	10500		Lethal 1 to 10 h
<i>Carassius</i>	Hard	6.8	10500		Lethal 12 to 99 h
Crustaceae		6.5	1400		3-week LC <sub>50</sub>
<i>Daphnia</i>	Fresh water	7.5	680		50% reprod. impair

Source: Alfred and Christian (1970)

### 5.3 Toxic effect on Human Being

Aluminum leaching by acid rain increases amount of free aluminum in environment and in surface waters. This can raise possibility on human exposure and neurological diseases (Harris, Berthon et al. 1996; Van Landeghem and D'Haese et al. 1997; Altmann and Cunningham et al. 1999).

Aluminum toxicity has been reported in impairment of the formation and release of parathyroid hormone and associated with the development of bone disorders, including fractures, osteopenia, and osteomalacia (Van Landeghem, D'Haese et al. 1997; Suarez-Fernandez et al. 1999; Kausz et al. 1999).

Damaging of brain cell by aluminum toxicity is discussed as a possible factor in the development of neurodegenerative disorders in humans. Aluminum toxicity also effect to mental status changes, learning disabilities, speech disturbances, coarse tremors, and produces an abnormal EEG, causes a microcytic hypoproliferative anemia which is a toxic agent in the etiology of Alzheimer's disease (Gilbert-Barness et al., 1998; Campbell and Bondy, 2000; Flaten, 2001; Forbes and Hill, 1998; Hachinski, 1998).

Aluminum toxicity can also interfere with the metabolism of calcium. Extreme nervousness, headaches, decreased liver and kidney function, memory loss, speech problems, softening of the bones, and aching muscles can all be caused by aluminum toxicity. Aluminum can accumulate in brain causing seizures and reduced mental alertness (Malluche et al., 1984).

#### **6. Accumulation of Aluminum from Water Treatment Sludge in the Soil**

In sludge form, even if there is an excessive amount of aluminum content, when utilized in agriculture sludge transformed into soil, aluminum will be fixed in the soil under rather high pH levels (Brandy and Weil, 1999; Boul et al., 1989). Thus sludge disposal by landfilling could cause aluminum accumulation in the soil.

#### **7. Leaching of Aluminum from Water Treatment Sludge Amended Soils**

When sludge transformed into soil, there was aluminum accumulating in the soil. And if there is any lowering in soil pH, the soil acidity will promote aluminum solubility and mobilize the aluminum from the soil into the surroundings.

The principle metal forms found in sludge are soluble, precipitated, coprecipitated in metal oxides, adsorbates, and are associated with biological residues (Lester et al., 1983). The distribution of metals between the specific forms varies widely according to the chemical properties of the individual metal and the characteristics of the sludge, which are a function of the physical and chemical properties imposed by the particular sludge treatment process. These parameters include pH, temperature, oxidation-reduction potential, the presence of complex agents, and the concentration of precipitant ligands (Gould and Genetelli, 1978). Solubility of aluminum in soil increases rapidly as soil pH decreases from 5.5 to 4.0. The species of aluminum ( $\text{Al}^{3+}$ ,  $\text{AlOH}^{2+}$ ) also change. (World Health Organization, 1997)

In addition, acid rain is one of various factors to cause aluminum leaching from sludge amended soil into the environment.

## **7.1 Factors Effect on Aluminum Leaching**

### **7.1.1 Acid Soil**

Soils are considered as acidic when pH is less than 7.0 and strongly acid when pH less than 5 (pH measured in water) (Table 16). Acidity is due to hydrogen ion ( $\text{H}^+$ ) concentration in soil. Soil acidity has two components: active acidity and exchangeable (reserve) acidity. Active acidity is the concentration of  $\text{H}^+$  ion in the solution phase of the soil and is measured by pH but is not a measure of the total soil acidity. The exchangeable acidity refers to the amount of  $\text{H}^+$  ions on cation exchange sites of negatively charged clay and organic matter fractions of the soil. In exchangeable acidity, soil acidification reflects an increase of acid cations ( $\text{H}^+$  and  $\text{Al}^{3+}$ ) at the expense of base cations on exchange sites (Brandtberg and Simonsson, 2003).

When soil pH drops below 6.0, the availability of nutrients such as phosphorus, potassium, calcium, and magnesium decreases while the availability of the metallic micronutrients such as zinc, manganese, copper, and iron increases. As the pH decrease below 5.5, the availability of aluminum and manganese increases and may reach a point of toxicity to

plant. Excessive  $\text{Al}^{3+}$  in the soil solution interferes with root growth and function, as well as restricting plant uptake of certain nutrients such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  (Jackson, 1963 ; McBride, 1994; Delhaize, 2004; McLean and Gilbert, 1927).

Table 16 Soil reaction (pH) (Soil : Water = 1:1)

Rating	Range
Ultra Acid	< 3.5
Extremely acid	3.5-4.5
Very strongly acid	4.5-5.0
Strongly acid	5.1-5.5
Moderately acid	5.6-6.0
Slightly acid	6.1-6.5
Neutral	6.6-7.3
Mildly alkaline	7.4-7.8
Moderately alkaline	7.9-8.4
Strongly alkaline	8.5-9.0
Extremely alkaline	> 9.0

Source: Soil Science Dictionary Committee (1998)

Thailand is located in tropical climate zone which hot and humidity atmosphere ease soil decomposition and promote soil acidity. Acid soil in Thailand is approximately 44% of over all area and 18.6% of agricultural area is considered as strongly to extremely acid (Yuwanियom and Charoenjamrascheep, 1994 ; Im-aim, 1993 ; Charoenjamrascheep et al., 1997). In Thailand, 51% of agricultural area has pH value lower than 5.5, 32.4% is considered as strongly to very strongly acid (pH 4.5-5.4)18.6% extremely acid (pH < 4.5), and 19.7% is considered as moderately to slightly acid (Apiradee, 1993).

Percentage of Thailand's agricultural area can be divided according to regions using different soil pH value as shown in Table 17, effects of soil pH on agricultures and crops related to aluminum toxicity are shown in Table 18, and effect of soil pH value on soil nutrient availability is shown in Figure 8.

**Table 17** Approximate percent of soil pH of Thailand's agricultural area (result from 9940 samples from all over the country)

Region	Soil pH (Range)					
	< 4.5	4.5 – 5.4	5.5 – 6.4	6.5 -6.9	7.0	> 7.0
Northern (%)	17.52	44.83	27.19	7.16	0.76	2.53
Central (%)	13.49	20.26	17.89	22.39	1.43	24.53
North-eastern (%)	23.58	46.51	14.02	10.21	0	5.67
Eastern (%)	47.98	32.88	12.13	5.39	0	1.62
Southern (%)	35.12	25.78	7.44	21.62	0	10.03
Overall (%)	18.60	32.40	19.70	15.20	0.90	13.20

Source: Im-aim (1993)

Table 18 Effects of soil pH on agricultures and corps

	Soil pH	Effect
Extremely acid	below 4.5	Few crops survive; aluminum/manganese toxicity.
Very acid	4.5-5.0	Only acid-tolerant plants such as azaleas, carpetgrass and blueberries do well.
Very acid	5.0-5.5	Some aluminum and manganese toxicity; nutrient deficiencies. Mid 5 is good pH for Irish potatoes, because scab bacteria don't survive well at this pH. Most crop yields slightly reduced, especially legumes.
Moderately acid	5.5-6.0	No visible problems with most crops; yields of crops requiring high calcium and magnesium may be reduced (for example, tomatoes and peppers). Good for centipede and carpet grasses.
Slightly acid	6.0-7.0	Ideal for most crops; best for soil bacteria/nitrogen fixation. Optimum nutrient availability. St. Augustine, bermuda & zoysia.
Slightly alkaline	7.0-8.0	Micronutrient deficiencies of iron, zinc and manganese may occur; too high for acid plants.
Alkaline	8.0+	Severe micronutrient deficiencies. Few garden crops do well. Acidulate your soil.
Extremely acid	below 4.5	Few crops survive; aluminum/manganese toxicity.

Source: LSUAgCenter.com (2005)

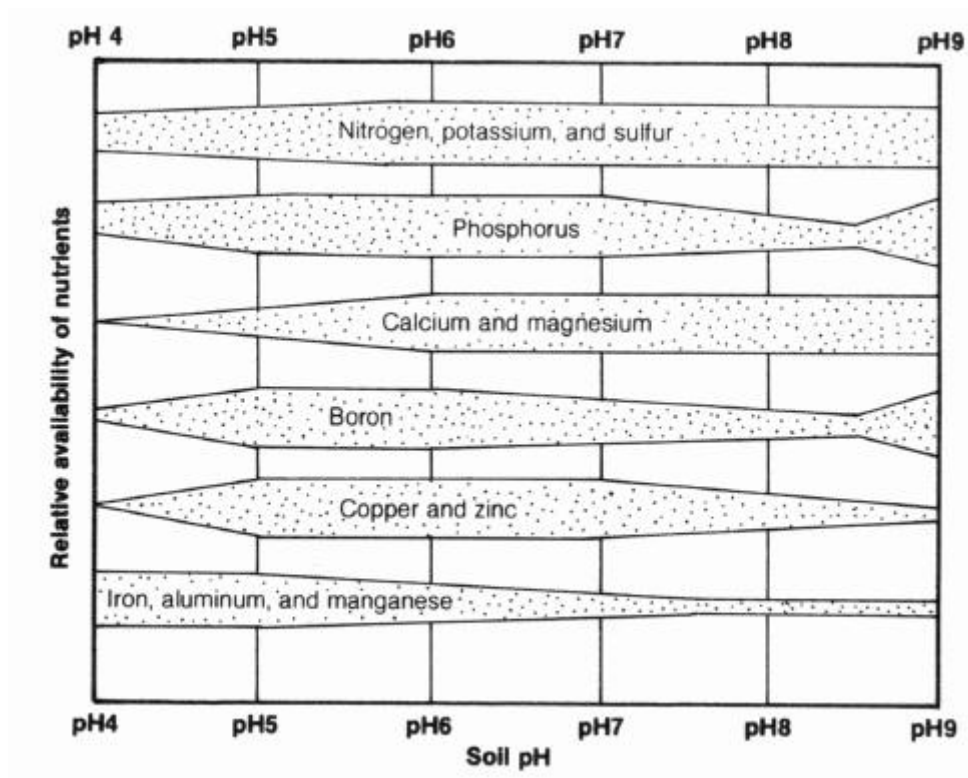


Figure 8 Effect of soil pH on nutrients availability

Source: Spies and Harms (2005)

### 7.1.2 Acid Rain

Acid rain was first reported in Manchester, England during the Industrial Revolution. In 1852, Robert Angus Smith found relationship between acid rain and atmospheric pollution and he started to use the term "acid rain" in 1872.

In 1970s, scientists began to observe and study about the phenomenon of acid rain. In 1990s, public awareness of acid rain in the U.S increased after the New York Times promulgated reports from the Hubbard Brook Experimental Forest in New Hampshire of the myriad deleterious environmental effects demonstrated to result from it (Wikipedia, 2005).

Acid rain or acid precipitation is a scientific term used to describe acidic pollutants deposited from atmosphere to earth's surface in wet and dry form. Wet deposition refers to acidic rain, fog, and snow while dry deposition refers to acidic gases and particles (Pidwirny, 2004; USEPA, 2005).

Acid rain is formed when oxides of nitrogen and sulfite combine with moisture in the atmosphere and make nitric and sulfuric acid which can be carried away far from its origin (Phamornsuwana, 1999).

Natural rain is slightly acidic because of carbon dioxide in the atmosphere dissolved with rain water to form carbonic acid. Natural rain has pH value of 5.6-5.7 while acid rain has pH lower than 5.6 (average pH value of rain water). The extra acidity in rain comes from reaction between air pollutants and gaseous water in clouds to form strong acids which falls to earth's surface as acid rain (Watson, 1997; Postel, 1984; Bourodimos, 1974).

Acid rain or acid deposition adds hydrogen ions in soil. The hydrogen ions displace important nutrients ( $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ , etc.) in soil and mobilize aluminum ions which are normally present in an insoluble nontoxic form of aluminum hydroxide to dissolve into the water, this process called leaching. In the leaching process, ions are washed out of top soil or into subsoil (Ophardt, 2003; Pidwirny, 2004).

Acid rain can mobilize cations in soil in two ways:

1. The  $\text{H}^+$  ion displaces the other positive ions from their binding sites and increases the concentration of these ions in the soil solution.
2. The sulfate and nitrate ions from acid rain (negatively charged) act as counter ions and result in leaching of soil positive ions (Cowley, 2004).

The two primary causes of acid rain are sulfur dioxide ( $\text{SO}_2$ ) and nitrogen oxides ( $\text{NO}_x$ ).

### **Sulfur Dioxide**

Sulfur dioxide (SO<sub>2</sub>) is a colorless gas, soluble in water. Natural sources of sulfur dioxide are volcanic eruption, biological decay and forest fires. Man-made sources are fossil fuel combustion, smelting, coal combustion, industrial processes. Main source of sulfur dioxide emission is from industrial combustion (around 69.4 percent of the emission), around 10 percent of the emission comes from volcanoes, sea spray, plankton, and rotting vegetation and only around 3.7 percent is from transportation (Environment Canada, 2005).

Sulfur dioxide emission is responsible for 60-70 percent of global acid rain precipitation and more than 90 percent of sulfur in the atmosphere is originated from human activity (Environment Canada, 2005).

### **Nitrogen Oxides**

“Nitrogen oxides or oxides of nitrogen (NO<sub>x</sub>)” is a term used to describe compounds of nitrogen with oxygen atoms. Nitrogen oxides are nitrogen monoxide (NO) and nitrogen dioxide (NO<sub>2</sub>). These oxides are by-product gases from extremely high temperature processes such as in automobile plants and chemical industries. Nitrogen monoxide is a colorless, flammable gas with a slight odor. Nitrogen dioxide is a reddish brown, nonflammable gas with detectable smell. Nitrogen dioxide is a strong oxidizing agent that reacts in the air to form corrosive nitric acid, as well as toxic organic nitrates.

Main source of NO<sub>x</sub> emission is the fuel combustion from vehicles, residential and commercial furnaces, industrial and electrical-utility boilers and engines (around 43 percent of all emission). Around 32 percent of the emission is from industrial combustions and only 5 percent is from natural processes such as bacterial action in soil, forest fires, and volcanic eruptions (Environment Canada, 2005).

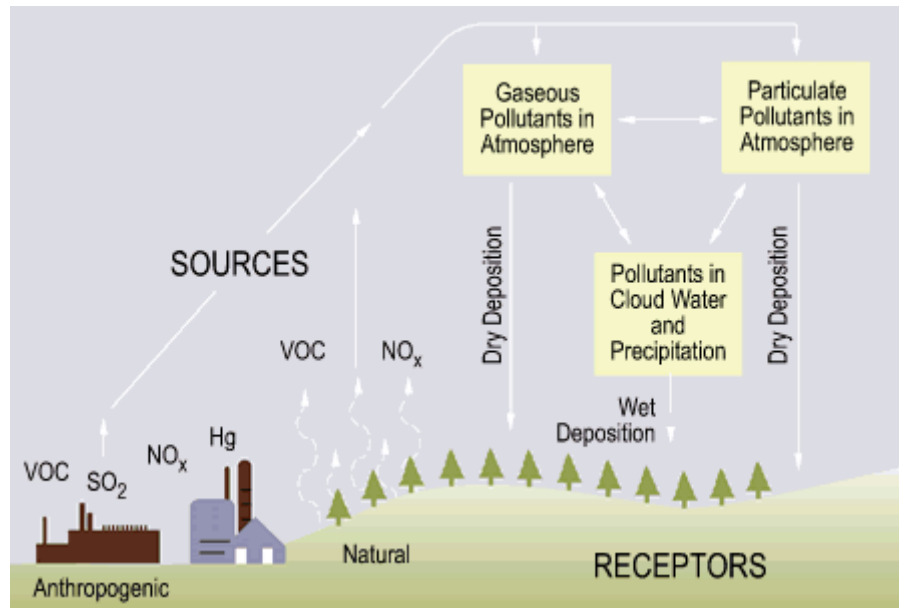


Figure 9 Main sources of SO<sub>2</sub> and NO<sub>x</sub> emission

Source: USEPA (2005)

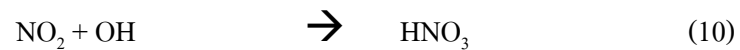
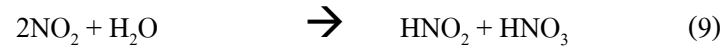
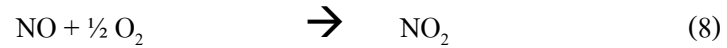
### **Acid Rain Formation:**

Acid rain primarily results from the transformation of sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides into dry or moist secondary pollutants such as sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) and nitric acid (HNO<sub>3</sub>). The transformation of SO<sub>2</sub> and NO<sub>x</sub> to acidic particles and vapors occurs as these pollutants are transported in the atmosphere over distances of hundreds to thousands of kilometers.

Sulfuric acid formation:

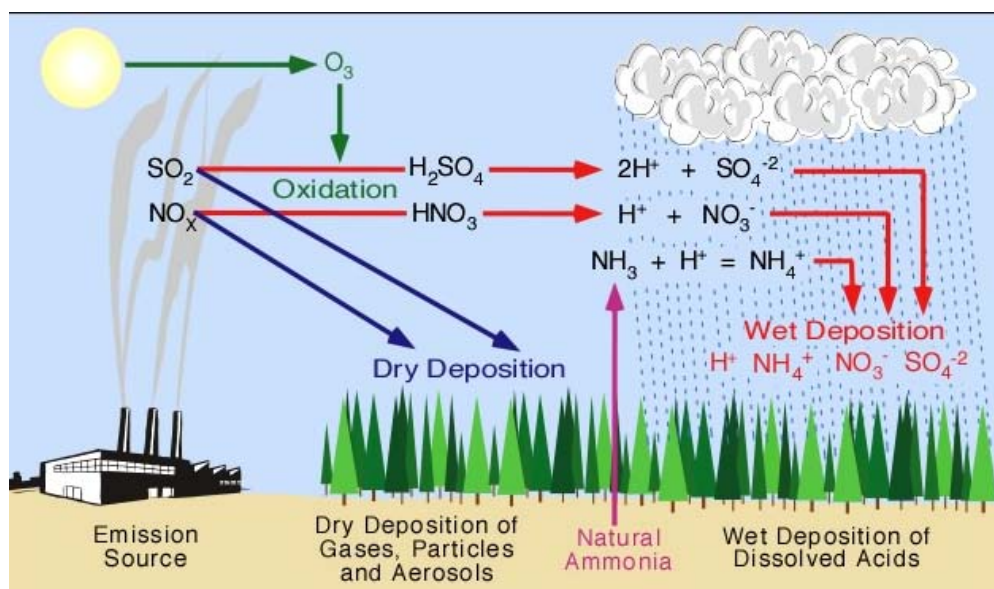


Nitric acid formation:



Source: Pidwirny (2004)

Acidic particles and vapors are deposited by two processes - wet and dry deposition (Figure 10). Wet deposition is acid rain, the process by which acids with a pH normally below 5.6 are removed from the atmosphere in rain, snow, sleet or hail. Dry deposition takes place when particles such as fly ash, sulphates, nitrates, and gases (such as  $\text{SO}_2$  and  $\text{NO}_x$ ), are deposited on, or absorbed onto, surfaces. The gases can then be converted into acids when they contact water (Environment Canada, 2005).



**Figure 10** Acidic particles and vapors deposition

Source: Pidwirny (2004)

## **8. Measuring Aluminum in Soil**

There are many different methods of measuring aluminum in soils available and have been used. Displacement of the soil solution, the use of acid solutions, and buffered and unbuffered salt solutions are methods used to extract aluminum from soil.

### **8.1 Total Aluminum**

Total aluminum provides useful information on the characterization of soils with respect to the origin of parent materials and weathering. It also serves as a basis for calculating the mineralogical composition of the sample (Bertsch and Bloom, 1996). However, total soil aluminum does not indicate aluminum toxicity. It is not possible to correlate the soil solution concentration of aluminum to the total soil aluminum measurement (van Breemen et al., 1989).

### **8.2 Exchangeable and Extractable Aluminum**

Exchangeable and extractable aluminum, displaced most commonly with an unbuffered salt solution such as 1M KCl, 0.5M CaCl<sub>2</sub>, or 0.5M BaCl<sub>2</sub>. The two primary uses of this are to calculate lime requirement for acid soil treatment (Reeve and Summer, 1970) and to establish the effective cation exchange capacity (ECEC) value (Mulder et al., 1987). To measure ECEC value, it is important to do reproducible measurement of exchangeable Al<sup>3+</sup> that reflects as accurate aluminum exchange equilibria as possible (Bertsch and Bloom, 1996).

In general, the exchangeable aluminum in soil solution is firmly fixed with clay anions between layer silicate or oxide-coated layer silicate (Abruna et al., 1975).

There is no clear explanation between aluminum exchanged from a permanently charged surface in equilibrium with unbuffered salt solution and that which concurrently released through solubilization from other nonexchangeable sources. Therefore, because of the nature of the exchangeable aluminum determination operation, exchangeable aluminum is commonly

referred to as extractable aluminum (Bertsch and Bloom, 1996). Examples of exchangeable aluminum in different soils leached with neutral unbuffered salt, potassium chloride, are shown in Table 19.

**Table 19** Cations replaced from B horizons of several southeastern Piedmont soils on leaching with 1N Potassium chloride

Soil	Exchangeable cations (meq. Per 100 g.)						Percent saturation	
	pH	Al	$\sum$ Ca, Mg, K	$\sum$ M+	H	CEC	I	II
Granville	4.8	12.1	1.3	13.4	5.7	19.1	9.7	6.8
Mayodan	4.9	8.9	1.8	10.7	4.3	16.0	15.4	11.2
White Store	4.6	17.9	4.5	22.4	7.3	29.7	20.0	15.1
Iredell	6.3	0.4	15.8	16.2	7.5	23.7	97.5	68.3
Mecklenburg	5.8	0.8	9.0	9.8	8.3	18.1	92.0	49.7
Davidson	5.9	0.2	4.5	4.7	7.6	12.3	95.8	36.6
Cecil	5.6	0.8	1.7	2.5	5.0	7.5	68.0	22.6
Georgeville	5.3	2.0	1.5	3.5	6.7	10.2	42.9	14.7
Appling	5.2	1.6	1.1	2.7	4.2	6.9	40.7	16.0
H-Peat	4.1	5.0	0	-	89.0	94.0	-	-
Ca-H-Peat	5.4	1.2	46.3	-	47.7	94.0	-	-

Source: Tisdale and Werner (1974)

### 8.3 Soluble Aluminum

Soluble aluminum in soil solutions is an important parameter to study the impact of soil acidification and to assess aluminum toxicity to plants in acid soils and aquatic organisms.

Most techniques used to obtain soil solutions for of the typical predominant soil cations,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ , and  $\text{K}^+$ , and aluminum (Eco-SSL, 2003).

### **9. Assessment of Aluminum Leachability**

Leaching methods used for investing leachability of sludge can be classified into 3 types:

- Regulatory methods: methods mandated by a regulatory agency to be used in a legal context.
- Standard methods: methods adopted and approved by a standards organization such as ASTM, ISO, etc. to establish the particular specification under specific conditions and/or materials.
- Research methods: methods developed for certain applications and particular purposes.

The methods can be categorized in a number of ways, by means of leaching solution application, such as static extraction or dynamic tests or categorized as batch leaching, flow through (column leaching), and flow around systems.

Batch leaching methods are the methods accomplished by placing samples in leaching solution for a certain period of time. Agitation may be needed to ensure the constant contact between the sample and the leachant. At the end of testing period, the liquid is removed and analyzed (Kim, 2002).

Column leaching methods are the methods simulating the percolation of the water through the waste or granular material either up-flow or down-flow and the leaching solution can be direct and continuous or intermittent flow through the material.

Leaching tests are performed on sludge from water treatment plant highlight the impact of treatment processes used in the plant on the leachability of a main metal ion in the sludge, aluminum. The test is modified from the column leaching method base on the Dutch standard

column test (NEN 7343) to study leaching of inorganic impurities from solid materials (Nordtest, 1995).

NEN 7343, Determination of the Leaching of Inorganic Components from Granular Materials with the Column Test. NEN 7343 is the up-flow column leaching test used to study leaching characteristics of building and waste materials and to determine the leaching of inorganic components from powdered and granular building materials and waste matters. In the test, the material size is reduced into a grain with diameter less than 4 mm. Leaching liquids are demineralized and acidified water of pH 4 ( $\text{HNO}_3$   $1 \pm 0.1$  mol/l). The test is performed in a vertical column with a  $5 \pm 0.5$  cm. internal diameter. The column is filled with a sample up to the minimum height of 4 times the internal diameter. The leaching solution passes through the sample from the bottom to the top with maximum percolation rate of 2 cm. per hour and 7 eluate fractions using liquid/solid substance ratio (L/S) within the range of 0.1-1.0 l/kg., and are collected to analyzed for the concentration of the leached components in the eluate (TitroWiCo, 1995; van Eijk and Brouwers et al., 2004)

Nordest Method, Leaching Procedure for Granular Waste Materials. The method is mostly identical to the NEN 7343. In the Nordest Method, the column diameter are optional (limiting on minimum diameter (5 cm.), minimum waste height (20 cm.), and minimum height/diameter (4 cm.). Flow rate of the Nordest Method is slower than the NEN 7343 and only 4-5 eluate fractions are collected with the L/S ratio of 0.1-2 l/kg (Poonpolwatanaporn, 2000).

Combined Column and Batch Leaching Test, the Danish Combined Column and Batch Leaching Test developed by VKI for granular waste or industrial sludge. In Combined Column and Batch Leaching Test, the waste is packed in a relatively large column (column diameter = 15 cm., waste height = 60 cm.) and leached in upflow direction with acidified de-mineral water of pH 4, the other leachants may be used depending to the scenario. Four eluate fractions are collected for L/S ratio 0-1 l/kg. Optionally, one more eluate may be collected at L/S ratio of 1-2 l/kg. Then, all the wastes are removed and mixed thoroughly before sending to a serial batch leaching test. This method is useful for granular wastes or industrial sludge with high L/S ratio.

The particle size of the waste and sludge should be less than 4 mm., the column flow rates are relatively comparable to the Nordest (Poonpolwatanaporn, 2000).

## MATERIALS AND METHODS

### General Information

The study was divided into three steps as follows:

Step I: Characterization of sludge, soils, and rain water

Physical and chemical analysis of water treatment sludge, soil, and rain water samples were performed in this part to determine characteristics of the samples.

Step II: Batch leaching experiments

Batch leaching tests were conducted to investigate effects of soil texture (indirectly relating to soil pH), temperature, and soil moisture content on aluminum leaching from the sludge and the optimum soil-sludge mixing ratio that give highest amount of aluminum leaching.

Step III: Column leaching experiments

Column leaching tests were conducted to determine the leachability of aluminum from water treatment sludge. The leachants were natural rain water (pH 6.7) and artificial rain water (pH 7, pH 5.5, and pH 3).

### Materials

1. Sludge from Bangkhen water treatment plant (Sampling on September 2004)
2. Natural soils (Sampling on September 2004);
  - Sandy soil

(from Pak-Chong District, Nakorn Ratchasima Province)

- Clay soil

(from Rangsit District, Pratumthani Province)

- Silt soil

(from Bang Gruay District, Nonthaburi Province)

3. Rain water (Sampling on September 2004);

- Natural rain water from Bangkhen District, Bangkok (pH 6.7)

- Artificial rain water with pH 7.0

- Artificial rain water with pH 5.5

- Artificial rain water with pH 3

Artificial rain water were prepared according to the ‘Synthetics acid rain extraction fluid’, ministry of industry announcement copy No. 6 (1997).

4. Special equipments for parameters analysis;

- Atomic Absorption Spectroscopy, nitrous, acetelene, and Al lamp

- Energy Dispersive X-Ray, gold sputtering

- Gas Chromatography (Aligent 6890), oven temperature 35 °C, carrier gas: N<sub>2</sub>, flow rate: 30 ml/min.

- Particle Size Analyzer

- Poresizer

- Spectrophotometry (HACH DR/4000), wave length: 395 nm.

## **Methods**

### **1. Experimental Preparation**

Prior to the experiment, the sludges, soils and rain water were sampled and stored in 5 °C room to maintain their original properties. The equipments were checked and calibrated to

assure the accuracy of the result of the testing/analysis to be conducted. The properties of samples being tested/analysis were:

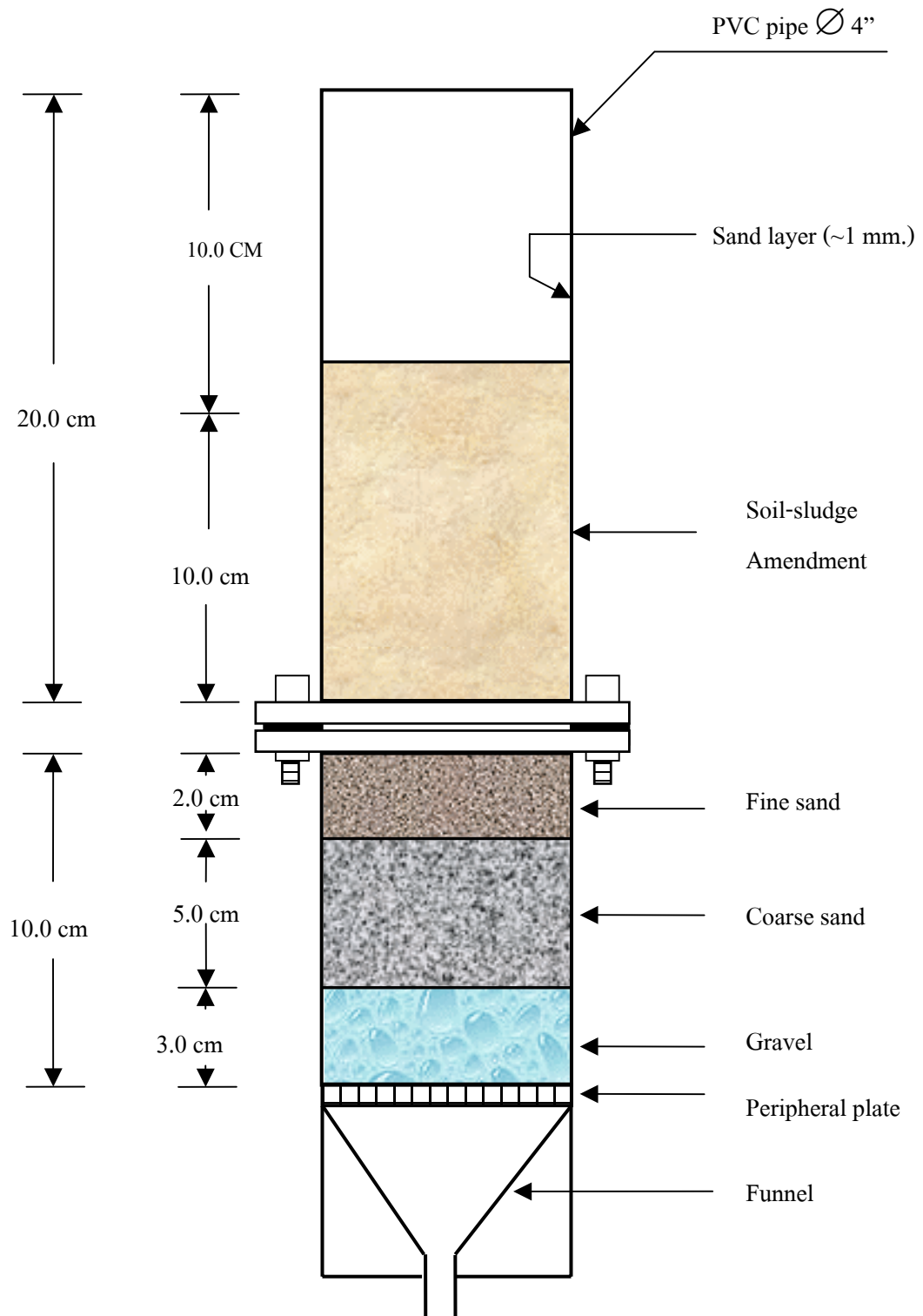
- Sludges were tested for physical, chemical and biological properties, i.e. pH, moisture content, organic matters, total solids, particle density, particle size distribution, element composition, and respiration rate.
- Soils were tested for pH, moisture content, organic matters, total solids, and element composition.
- Rain water was tested for only pH value.

## **2. Column Design**

The columns were made of Polyvinyl Chloride (PVC) pipe with diameter 10.16 cm and height 30 cm. To prevent metal leaching from the pipe itself, the inside surface of the column was layered with sand with ~1mm thickness. The peripheral acrylic plate was placed in the bottom of the column as base to prevent the gravel from coming off from the column.

The column was filled with gravel, coarse sand, fine sand and the sample tested with depth of 3, 5, 2 and 10 cm, respectively. Details of column design and dimensions are shown in Figure 11.

The columns were installed in the laboratory under ambient temperature.



**Figure 11** Column dimension and design

### **3. Sampling and Preparation of Samples**

Sludge sampling: sludge samples were collected from Bangkok water treatment plant. During sludge sampling, the samples were handled with great care under 5 °C to keep their properties as close as in the actual situation before the experiment started. Location of the sludge sampling points is shown in Figure 12.

Soil sampling: the soils were collected from three different areas to obtain three different soil textures.

- Sandy soil was taken from Pak-Chong District, Nakhon Ratchasima Province
- Clay soil was taken from Rangsit District, Pratumthani Province
- Silt soil was taken from Bang Kruey District, Nonthaburi Province.

The soil samples were kept with great care under 5 °C to maintain their properties as close as in actual situation prior to the experiment start.

Rain water: the natural rain water were collected at Bangkok District, Bangkok and kept under temperature of 5 °C before using in the experiment.

Artificial rain water were prepared by additional of chemical reagents and distilled water to adjust pH to desired value before using in the experiment.

In this experiment, preparation of artificial rain water used 80% by weight of sulfuric acid well mixed with 20% by weight of nitric acid and then, added distilled water to adjust pH value.

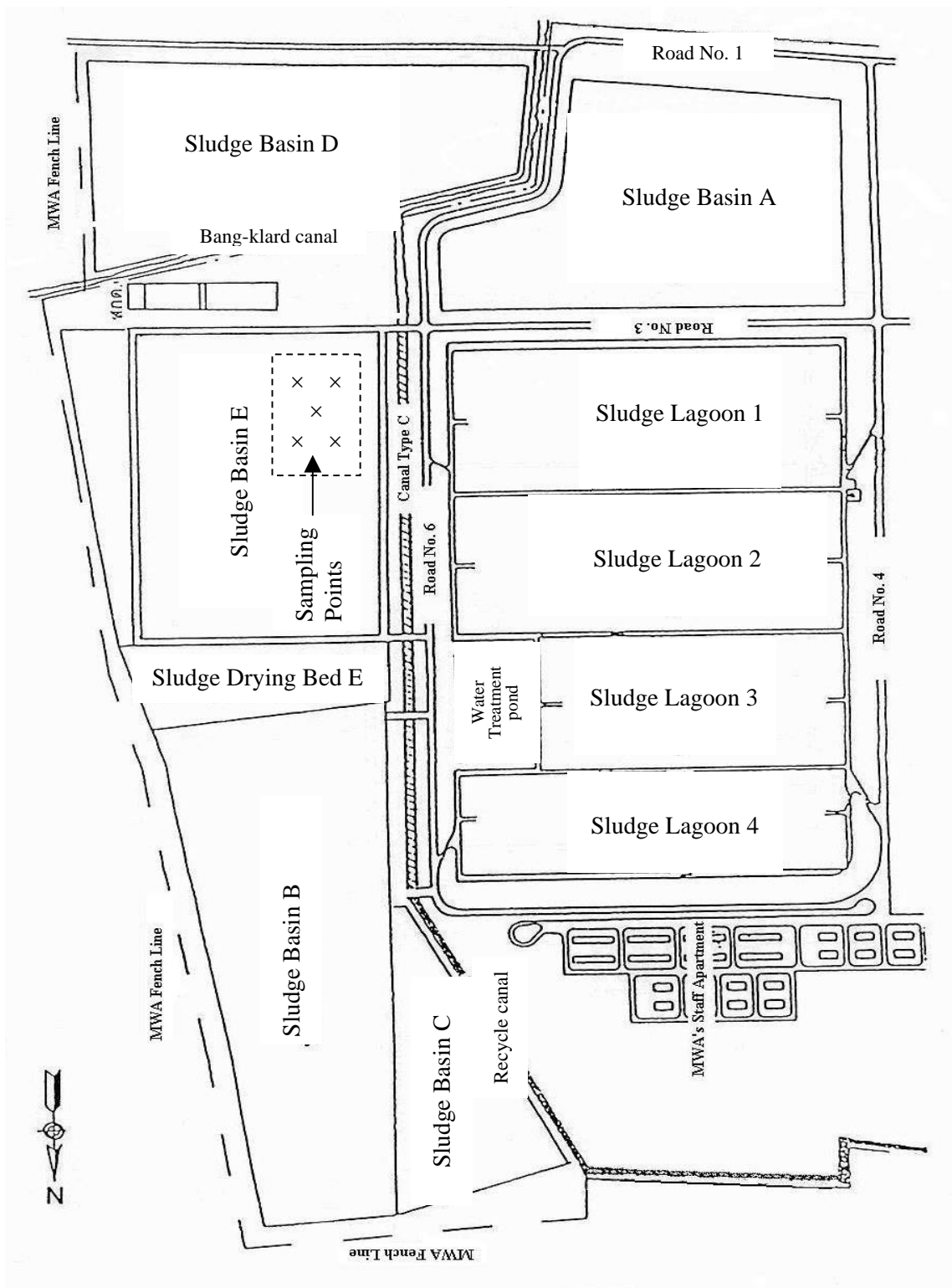


Figure 12 Sludge sampling locations

#### 4. Sample Analysis

The determined parameters of the samples were analyzed in Laboratory of Environmental Engineering Department at Kasetsart University, Bangkok, Thailand. The analytical parameters are shown in Table 20.

Table 20 Analytical parameters for the samples

Sample	Parameter
Rain water	- pH
Soil	- pH - Moisture content - Organic matter content - Total solids - Element composition
Water treatment sludge	Physical properties: - Particle density - Particle size distribution - Moisture content - Total solids Chemical properties: - pH - Organic matters Biological property: - Respiration rate

## 5. Analytical Methods

The analytical methods for all parameters were followed the standard methods. They are shown in Table 21.

Table 21 Parameters and analytical methods of soil and sludge samples

Parameter	Method/Reference
pH	Electrometric method (pH meter) 1: 2 (sludge : distilled water)
Element composition (%)	E1508-98 standard guide for quantitative analysis by energy-Dispersive spectroscopy E1621-94
Moisture content (%)	Incubated at 105 °C, 20 hrs.
Organic matters (%)	Gravimetric method (Dried at 550 °C, 1.5 hrs.)
Particle density (%)	Poresizer
Particle size distribution (%)	Sieving and gravimetric method (Dry weight)
Soil respiration rate ( $\text{g}_{\text{O}_2}/\text{g}_{\text{soil}}/\text{day}$ )	Oxygen depletion with time by Gas chromatography technique
Total solids (%)	Standard methods, methods 2540 B./APHA, 1995
Extractable aluminum (mg/l) (Appendix B)	KCl Extractable Al, atomic absorption spectrometry 3111/APHA, 1995 (for soil samples) and 8-Hydroxyquinoline-Butyl Acetate, spectrophotometric methods for aluminum analysis methods of soil analysis, 1996 (for leachate samples)

## **6. Batch Leaching Experiments**

The experiments were carried out to study effects of soil acidity, soil moisture content and soil temperature on aluminum leaching under different soil textures and soil-sludge mixing ratios. The pH of soil/sludge experiments were in their natural values. The soil and sludge were mixed manually by hands in closed plastic bags to prevent moisture lost. To prepare the well-mixed mixture, the sludge was applied into the soil then, they were kneaded homogenously inside the plastic bag.

Description of each experiment is explained as follows:

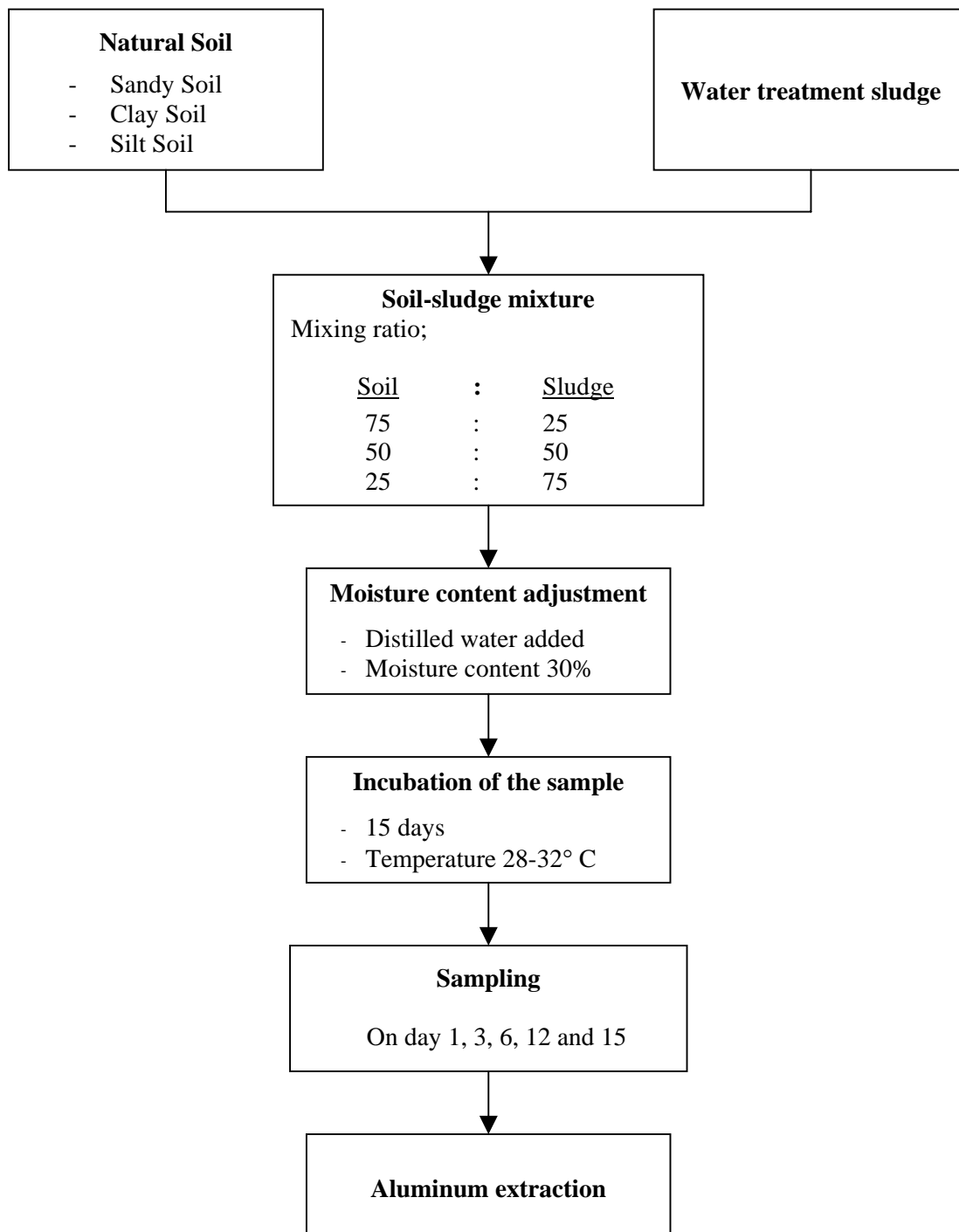
### 6.1 Experiment on effect of soil texture on aluminum leaching

Each soil type (clays, sands and silts) was mixed with the sludge in 3 ratios of 25:75, 50:50, and 75:25 based on dry weight (equivalent to sludge disposal of 444.82, 148.27, and 49.42 kg/m<sup>2</sup> respectively for Korat soil, 497.36, 165.79, and 55.26 kg/m<sup>2</sup> respectively for Rangsit soil, and 505.28, 168.43, and 56.14 kg/m<sup>2</sup> respectively for Nonthaburi soil). Then, water was added to obtain 30% moisture content of the mixture. Each ratio was duplicated. There were 10 mixture samples for each ratio. The mixtures were kept in plastic cup closed with plastic lid to prevent moisture lost and incubated under room temperature for 15 days. During the incubation period, two cups of the each mixing ratio were taken out on day 1, 3, 6, 12 and 15 to examine amount of leaching aluminum. The optimum mixing ratio which produced the highest amount of aluminum in extraction would be applied into the next batch and the column leaching tests (Figure 13).

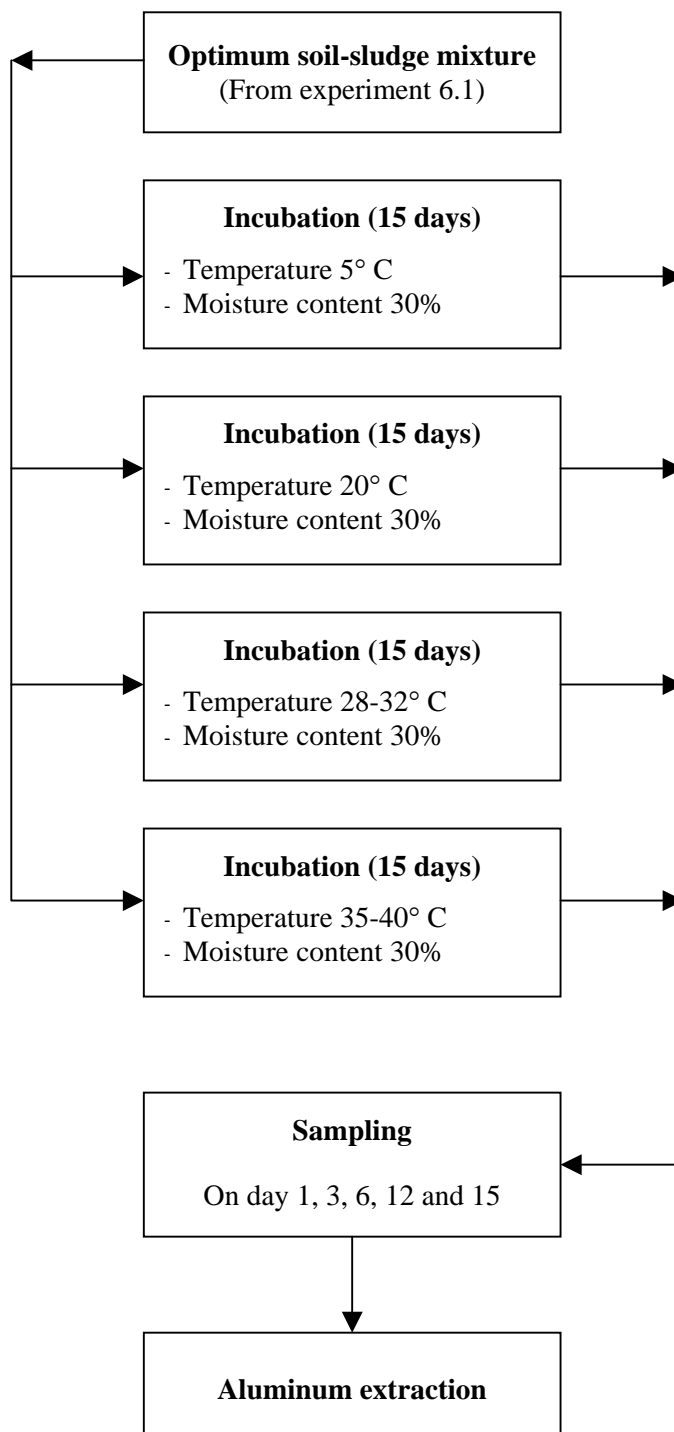
### 6.2 Experiment on effect of soil temperature on aluminum leaching

Four sets of the optimum ratio of soil-sludge mixture from experiment 6.1 were prepared, each set was triplicated with 15 mixture samples and each sample was put into plastic cup with cover lid. Each set of the sample was incubated under 5<sup>o</sup>, 20<sup>o</sup>, 28-32<sup>o</sup> and 35-40

°C respectively for 15 days. During incubation period, 3 cups of each set were taken out on day 1, 3, 6, 12 and 15 to determine the amount of aluminum leaching (Figure 14).



**Figure 13** Batch leaching experiment procedures to study effect of soil texture on aluminum leaching



**Figure 14** Batch leaching experiment procedures to study effect of temperature on aluminum leaching

### 6.3 Experiment on effect of soil moisture content on aluminum leaching

Two groups of the optimum ratio of soil-sludge mixture from experiment 6.1 were prepared. Each group had 4 sets of sample with 4 different initial moisture contents (5, 10, 20, and 30% respectively). Each set was triplicated with 45 mixture samples and put into plastic cup with cover lid. One group of the samples were incubated under room temperature (28-32 °C), another were incubated under 35-40 °C for 15 days. During incubation period, 3 cups of each set were taken out on day 1, 3, 6, 12 and 15 to determine the amount of aluminum leaching (Figure 15).

## 7. Column Leaching Experiment

The column leaching experiment was a modified method with the reference to NEN 7343 of which column scale was reduced to 10.16 cm diameter and 30 cm. height with downflow of water through the column (Poonpolwatanaporn, 2000).

This experiment was conducted to investigate effects of rain water pH on leachability of aluminum from the sludge. There were 4 sets of leaching columns filled with the optimum soil-sludge mixing ratio (from the 6.1 experiment) to determine leachability of aluminum from soil-sludge mixture. Each set was triplicated with 3 columns. There were 4 different leachants for each set of the mixture sample, natural rain water with pH 6.7, artificial rain water with pH 7, artificial rain water with pH 5.5, and artificial rain water with pH 3.

The mixture was packed into the column and incubated under room temperature for fifteen days to reach stabilization. During incubation period, the mixture was rinsed by the leachant on day 1, 3, 6, 12 and 15 and the leachate was sampling in the same day to determine the amount of aluminum leaching. The amount of sludge sample in column can be calculated step by step as follow:

$$\text{Moisture Content (MC)} = \frac{\text{Wet weight (kg)} - \text{Dry weight (kg)}}{\text{Wet weight (kg)}} \times 100\%$$

$$\text{Dry weight of sludge} = \text{Volume of column (L)} \times \text{Bulk density (kg dry weight/l)}$$

$$\text{Wet weight of sludge (kg)} = \frac{\text{Dry weight of sludge (kg)}}{(1 - \text{Moisture content \%})}$$

The procedure for column leaching experiments is shown in Figure 16.

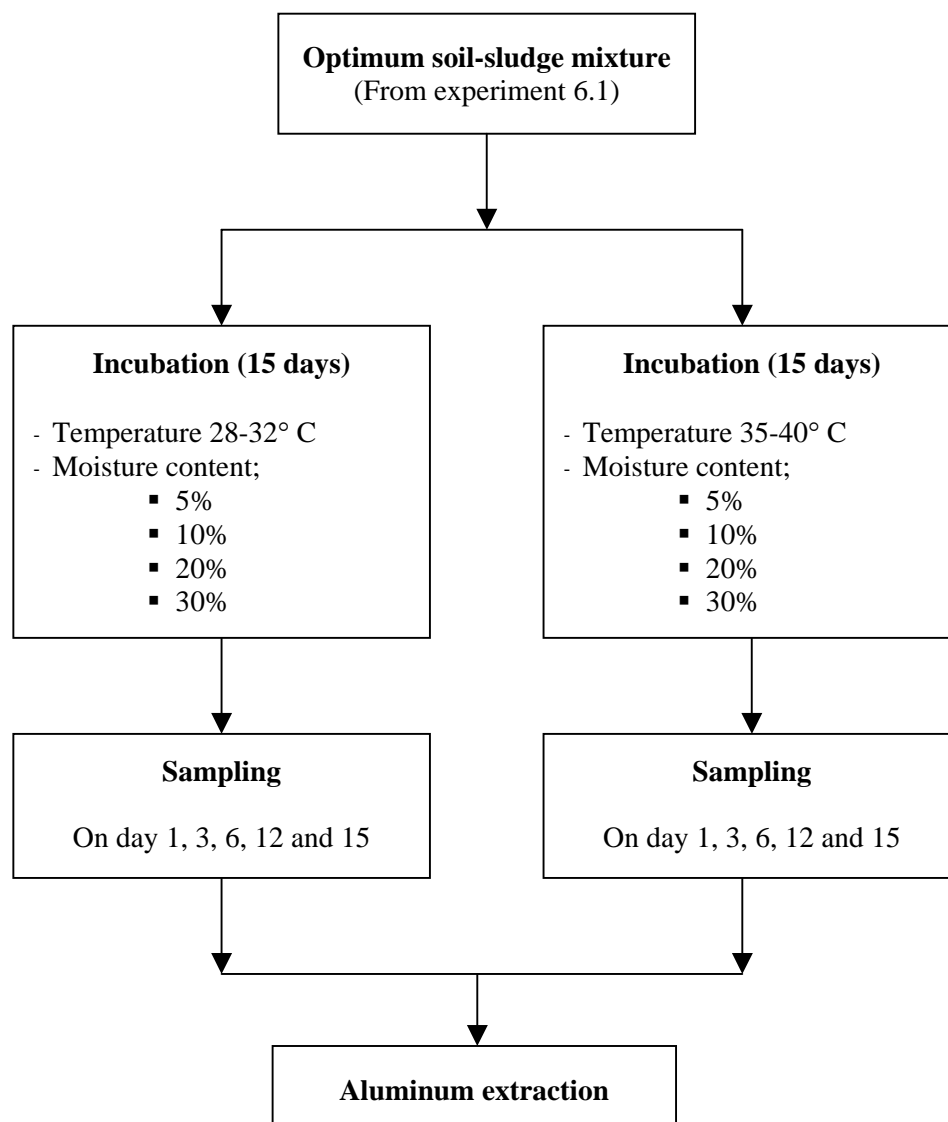
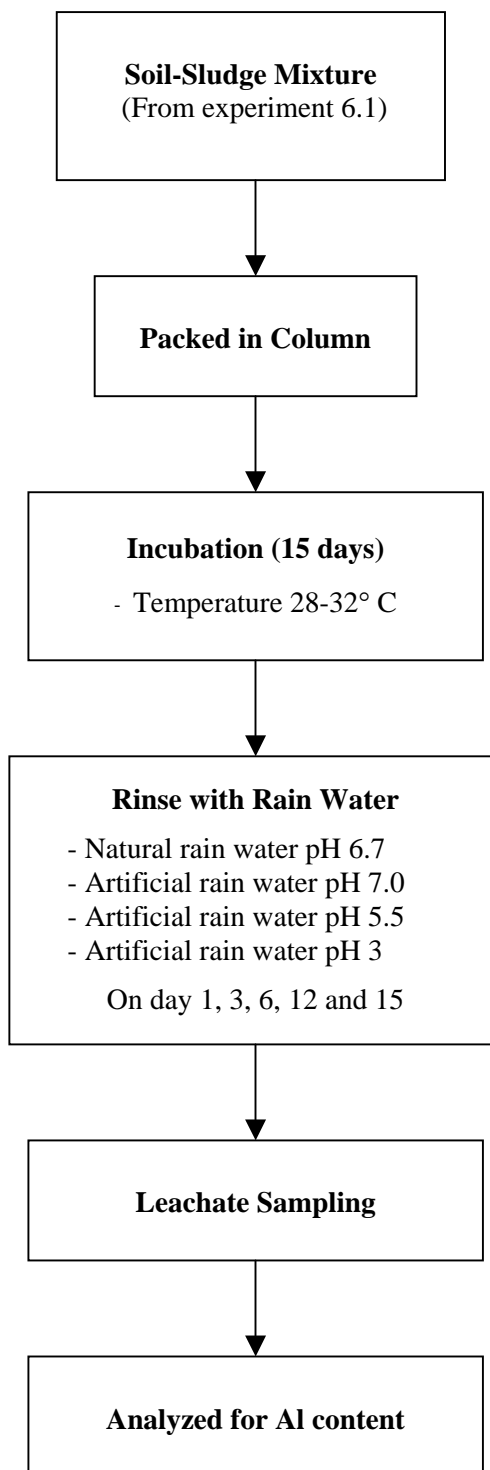


Figure 15 Batch leaching experiment procedures to study effect of soil moisture on aluminum leaching



**Figure 16** Procedures for column leaching experiments

## RESULTS AND DISCUSSIONS

### 1. Characterization of Sludge, Soil, and Rain Water

#### **1.1 Characterization of Bangkhen Water Treatment Sludge**

The characteristics of sludges from Bangkhen water treatment plant were analyzed as shown in Table 22. The analysis results show that the sludges were slightly acidic (pH 6.52-6.60) with rather slightly high moisture content (50.78-54.03%), very high organic matter content (8.50-8.71%), and medium quantity of total solid content (46.27-47.57%).

Sludge texture was homogeneous. The result from particle size distribution analysis shows that the sludge had low quantity of sand particles (less than 40 g/kg; 4%) and consisted mainly of silt particles (505.2-740.5 g/kg; 50.52-74.05%) which is almost double of clay particle (246.2-458.7 g/kg; 24.62-45.87%). In general, the sludge was silty clay sediment texture (Table 22).

The sludge has low particle density. The particle density of the sludge samples was 2.57-2.58 g/cm<sup>3</sup> (Table 22).

Microbial activities of the sludge were determined as respiration rate measurement. The result shows that there were microbial activities, the average respiration rate of microbial in sludge was 162.00 g<sub>(O<sub>2</sub>)</sub>/g<sub>(Soil)</sub>/day. This indicates that the sludge contained active, aerobic microorganisms involving in organic/inorganic decomposition. These activities resulted in oxygen depletion concurrent with production of carbon dioxide, as shown in the Figure 17.

In general, decomposition of organic matters or inorganic substances via microbial metabolism releases CO<sub>2</sub> which reacts with water and finally results in HCO<sub>3</sub><sup>-</sup> and H<sup>+</sup> releasing. Normally, the free H<sup>+</sup> can replace aluminum in sludge subsequently in Al<sup>3+</sup> leaching (McBride, 1994).

For element composition, it was found that the sludge had total aluminum content of 133.56 g/kg, equivalent to 13.35% of total element (other element composition of the sludge is shown in Table 22). Comparing with soils used in the experiment, sludge had highest total aluminum content (13.35%) then, Nonthaburi, Korat soil and Rangsit soil (10.30, 9.88 and 9.87%) respectively. The sludge had extractable aluminum value of 1.63 mg/l.

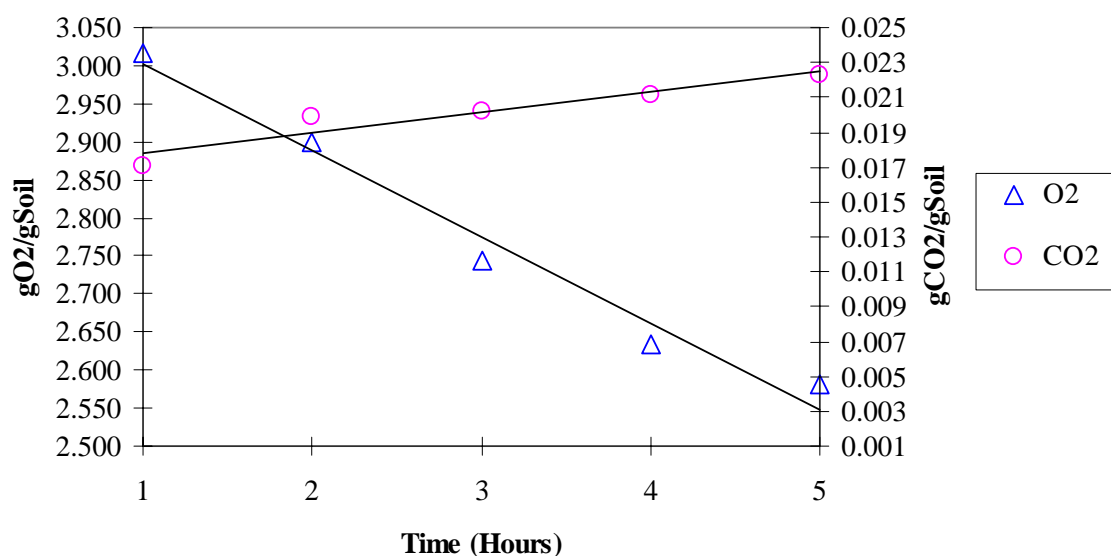
Table 22 Characteristics of Bangkhen water treatment sludge

Parameter	Range	Average	
pH	6.52-6.60	6.57	
Moisture content (%)	50.78-54.03	53.09	
Organic matter (%)	8.64-8.86	8.74	
Total solid (%)	46.27-47.57	46.95	
Extractable aluminum (mg/l)	0.61-2.18	1.63	
Particle size distribution (g/kg) *			%
- Sand (2-0.05 mm)	13.34-36.05	26.32	2.63
- Silt (0.05-0.002 mm)	505.21-740.46	630.48	63.05
- Clay (< 0.002 mm)	246.20-458.74	343.21	34.32
Particle density (g/cm <sup>3</sup> )	2.57-2.58	2.58	-
Element composition(g/kg);			%
- C	0.00-0.00	0.00	0.00
- O	547.01-556.46	551.3	55.12
- Na	3.47-3.84	3.67	0.37
- Mg	7.80-8.56	8.16	0.82
- Al	126.63-139.11	133.56	13.35
- Si	233.71-241.47	238.29	23.82
- K	14.03-15.46	14.65	1.46

Table 22 (Continued)

Parameter	Range	Average
- Ca	5.10-5.44	5.26
- Ti	4.13-4.29	4.23
- Fe	38.92-44.01	40.86

Note: \* Particle size distribution classification according to United States Department of Agriculture (USDA);

Figure 17 O<sub>2</sub> depletion and CO<sub>2</sub> production of Bangkok water treatment sludge

## 1.2 Characteristics of Natural Soil Samples

### 1.2.1 Nonthaburi Soil

Nonthaburi soil used in this study was strongly acid soil with pH range of 5.16-5.27. The soil had moisture content in range of 20.92-23.16%, organic matter content of 6.06-6.30% and total solids of 77.55-79.37%. For Al content, it had Al content in range of 99.90-

105.44 g/kg which was equivalent to 9.99-10.54% of total element. Other element compositions are shown in Table 23. Nonthaburi soil had extractable aluminum value in range of 5.39-7.68 mg/l.

**Table 23** Characteristics of Nonthaburi soil

Parameter	Range	Average	
pH	5.16-5.17	5.21	
Moisture content (%)	20.92-23.16	22.07	
Organic matter (%)	6.06-6.30	6.18	
Total solid (%)	77.55-79.37	78.29	
Extractable aluminum (mg/l)	5.39-7.68	6.07	
Element composition(g/kg);			%
- C	0.00-0.00	0.00	0.00
- O	586.70-612.18	598.79	59.88
- Na	4.30-4.94	4.61	0.46
- Mg	6.43-7.13	6.85	0.68
- Al	99.90-105.44	102.98	10.30
- Si	186.44-220.03	201.59	20.16
- K	11.83-14.27	12.93	1.29
Element composition(g/kg);			%
- Ca	3.33-3.85	3.58	0.36
- Ti	5.86-7.25	6.50	0.65
- Fe	59.21-65.36	62.17	6.22

### 1.2.2 Rangsit Soil

Rangsit soil was extremely acid soil with pH range of 4.29-4.43. The soil had 32.97-39.14% moisture content, 10.03-10.15% organic matter, and 60.97-66.39% total solids (Table 24).

Rangsit soil had Al content in range of 92.90-104.10 g/kg which is equivalent to 9.29-10.41% of total element. Other element compositions are shown in Table 24. Rangsit soil had extractable aluminum value in range of 5.66-7.55 mg/l.

**Table 24** Characteristics of Rangsit soil

Parameter	Range	Average	
pH	4.29-4.43	4.35	
Moisture content (%)	32.97-39.14	35.58	
Organic matter (%)	10.03-10.15	10.10	
Total solid (%)	60.79-66.39	64.09	
Extractable aluminum (mg/l)	5.66-7.55	6.42	
Element composition(g/kg);			%
- C	0.00-0.00	0.00	0.00
- O	558.78-602.55	580.98	58.10
- Na	2.93-6.46	4.18	0.42
- Mg	6.31-8.35	7.29	0.73
- Al	92.90-104.10	98.72	9.87
- Si	231.80-261.86	247.94	24.79
- S	14.20-16.53	15.07	1.51
- K	11.24-13.27	12.12	1.21
- Ca	3.38-4.19	3.75	0.37

Table 24 (Continued)

Parameter	Range	Average	
- Ti	5.12-6.90	5.77	0.58
- Fe	22.54-25.84	24.18	2.24

### 1.2.3 Korat Soil

Korat soil is moderately alkaline soil which may result from over-liming. The soil pH was 8.22-8.37. The soil has low moisture content of 13.46-14.95%, low organic matter (2.76-3.03%), and high total solids content (83.86-86.54%).

Korat soil had Al content in range of 91.54-100.47 g/kg which is equivalent to 9.62-10.03% of total element. Other element compositions are shown in Table 25. Korat soil had extractable aluminum value in range of 1.33-1.70 mg/l.

Table 25 Characteristics of Korat soil

Parameter	Range	Average	
pH	8.22-8.37	8.29	
Moisture content (%)	13.46-14.95	14.25	
Organic matter (%)	2.76-3.03	2.91	
Total solid (%)	83.86-86.54	85.74	
Extractable aluminum (mg/l)	1.33-1.70	1.47	
Element composition(g/kg);			%
- C	0.00-0.00	0.00	0.00
- O	494.92-518.00	504.53	50.73
- Na	4.22-5.79	4.95	0.50
- Mg	6.38-7.45	7.35	0.73
- Al	91.54-100.47	96.79	9.88

Table 25 (Continued)

Parameter	Range	Average	
- Si	300.43-316.10	307.48	30.92
- K	14.58-15.27	14.88	1.50
- Ca	6.66-7.29	6.93	0.39
- Ti	5.08-5.93	5.52	0.56
- Fe	48.61-56.42	51.56	4.80

Note: Korat Soil is excessive liming treatment soil from Pak-Chong Distric, Nakorn Ratchasima Province

Among the three soils type, Rangsit soil had the lowest soil pH while Korat soil had the highest soil pH. Results of extractable aluminum from soils and sludge indicated that Rangsit soil gave the highest amount of extractable aluminum (average 6.42 mg/l) and Korat soil gave lowest amount (1.47 mg/l). Information of soils and sludge properties are shown in Table 26.

**Table 26** Comparison of soils and sludge properties

Parameter	Sludge	Nonthaburi soil	Rangsit soil	Korat soil
pH value	6.52-6.60 (6.57)	5.16-5.27 (5.21)	4.29-4.43 (4.35)	8.22-8.37 (8.29)
Al content (g/kg)	126.63-139.11 (133.56)	99.90-105.44 (102.98)	92.90-104.10 (98.72)	91.54-100.47 (96.79)
Extractable Al (mg/l)	0.61-2.18 (1.63)	5.39-7.68 (6.07)	5.66-7.55 (6.42)	1.33-1.70 (1.47)
Moisture content (%)	50.78-54.03 (53.09)	20.92-23.16 (22.07)	32.97-39.14 (35.58)	13.46-14.95 (14.25)
Organic matter (%)	8.50-8.71 (8.74)	6.06-6.30 (6.18)	10.03-10.15 (10.10)	2.76-3.03 (2.91)

Note: Number in the blanket is average value.

### 1.3. Characteristics of Natural Rain Water

The natural rain water samples were collected in Bangkhen area, the rain was slightly acidic with pH value in range of 6.63-6.74.

## 2. Batch Leaching Experiment

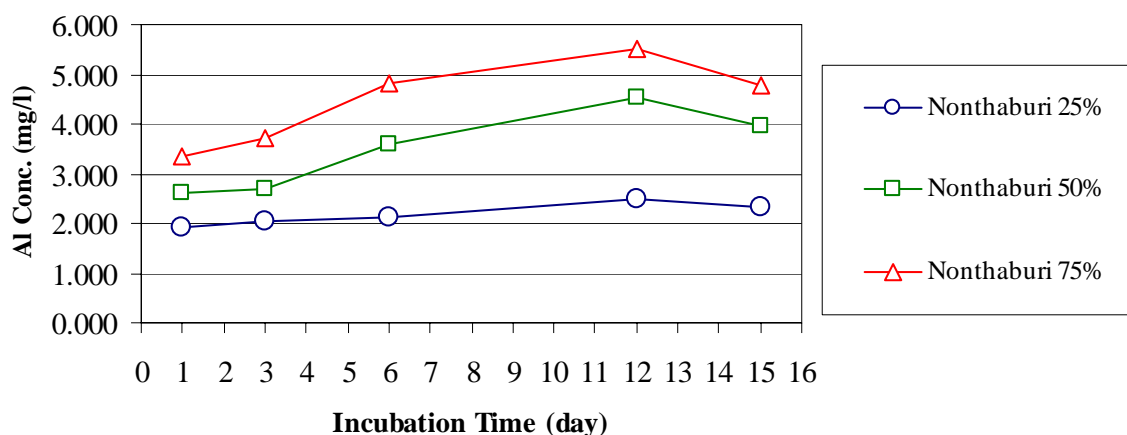
### 2.1. Effect of soil texture on Al leaching from soil-sludge amendment

There were 3 different types of soil used in this experiment, sandy soil (Korat soil), clay soil (Rangsit soil), and silt soil (Nonthaburi soil) of which the initial pH values were 8.29, 4.35, and 5.21, respectively.

The reason of using different soil type was that each soil type had different pH value which had different effect on amount of aluminum leachability. Amount of aluminum leaching increases when soil pH decreases (McBride, 1994).

In the experiment, each soil was amended with sludge using 3 different soil-sludge mixing ratios, 25:75, 50:50, and 75:25 percent dry weight.

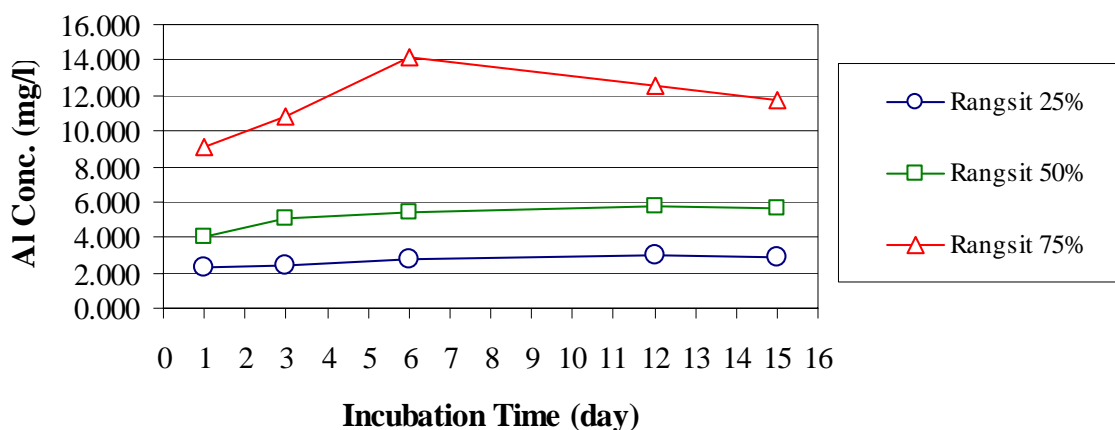
The results from Nonthaburi soil-sludge amendments shows that aluminum leaching of every soil-sludge mixing ratio was rather constant in the first 3 days of incubation and increasing in the latter incubation period (Figure 18). Among the 3 mixing ratios, the amendment with 75% Nonthaburi soil and 25% sludge gave the highest amount of aluminum leaching. However, all ratios gave peaks of Al leaching at day 12 of incubation.



**Figure 18** Average values of aluminum leaching amount from Nonthaburi soil-sludge amendment

The ratio of 75% Nonthaburi soil and 25% sludge gave highest amount of aluminum leaching may result from soil pH value. The higher the ratio of soil being used, the lower pH of soil-sludge amendment appeared and this may result to high amount of Al leaching according to effect of soil pH value.

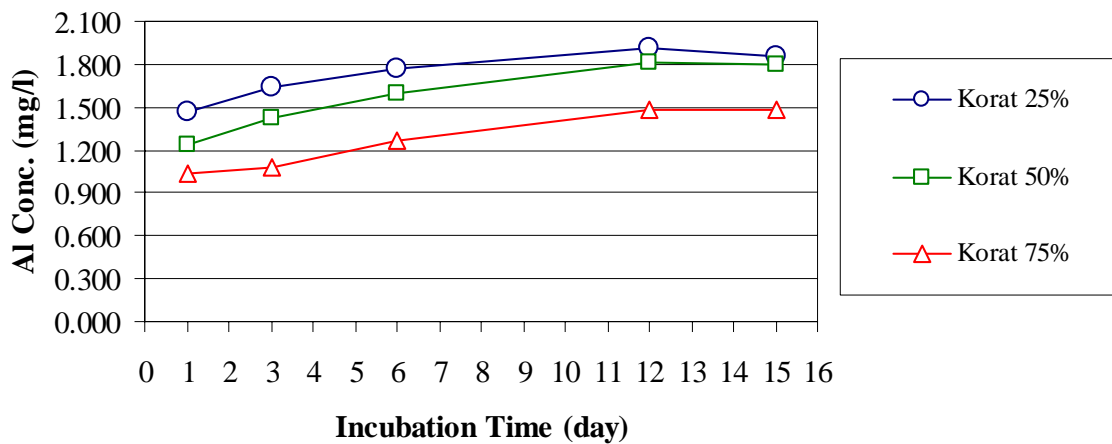
In case of Rangsit soil-sludge amendments it was found that trends of aluminum leaching were increasing with an increase in the amount of soil ratio: the soil-sludge mixing ratio with Rangsit soil 75% and sludge 25% gave the highest amount of aluminum leaching as compared to the other two mixing ratios. This may be because of the soil itself had low pH value and the high amount of soil used in the soil-sludge amendment reduced the soil-sludge amendment pH into acidic level which result in high Al leaching. It also gave highest amount of aluminum leaching at day 6 of incubation period then, slightly decreased after. While the other two mixing ratios had slightly increase (0.7-1.8 mg/l) (Figure 19). The slightly increasing of amount of Al leaching from the last two mixing ratios may result from the effect of sludge pH over the soil-sludge amendment as the sludge as rather high pH value (6.57) compared with soil pH value (4.35). Therefore, for the mixing ratio with high fraction of sludge mixed result in rather high pH value of the soil-sludge amendment and this may result to low amount of Al leaching.



**Figure 19** Average values of aluminum leaching amount from Rangsit soil-sludge amendment

For Korat soil-sludge experiment, it appeared that amounts of aluminum leaching had opposite trends from cases of Nonthaburi and Rangsit soil experiments. The mixing ratio of 75:25 % soil-sludge gave the lowest amount of aluminum leaching while the mixing ratio at 25:75 % soil-sludge gave highest amount of aluminum leaching through experiment period (Figure 20).

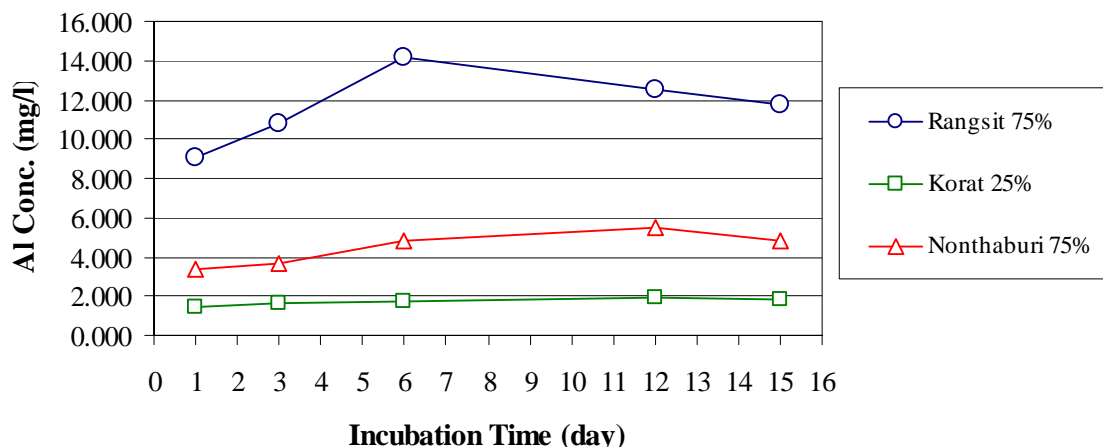
All experiments had trends to increase Al leaching content during 12 days of incubation period and kept the levels until the last day of experiment.



**Figure 20** Average values of aluminum leaching amount from Korat soil-sludge amendment

Comparison of all soil textures, it could be summarized that Rangsit soil had the strongest effect on aluminum leaching as shown in Figure 21. This may result from the soil pH value of Rangsit soil having the lowest pH among the 3 soil types. The low pH value of soil could increase the solubility of aluminum (Matsumoto et al., 1980).

Although Al ions could be immobilized by organic matter in the soil, however organic matter in Rangsit soil, which has the highest content of organic matter than the others, did not help much in trapping the free Al ions in the experiment. The effect of pH had strongly influenced Al leaching as supported by the amount of aluminum leaching from Korat soil-sludge amendments. The soil pH value of Korat soil was basic. Under high soil pH value, most of aluminum will be fixed (Brandy and Weil, 1999; Buol et al., 1989).



**Figure 21** Comparison of maximum aluminum leaching from 3 different soil textures

It can summarize that soil acidity, normally clay texture, effected on aluminum leaching. Especially in case of soil with pH value less than 5.5, obviously high amount of aluminum leaching from soil-sludge amendment was observed. For example, Rangsit soil (classified as extremely acid soil with pH 4.35) mixed water treatment sludge about 25% dry weigh, this gave maximum 14.2 mg/l aluminum leaching.

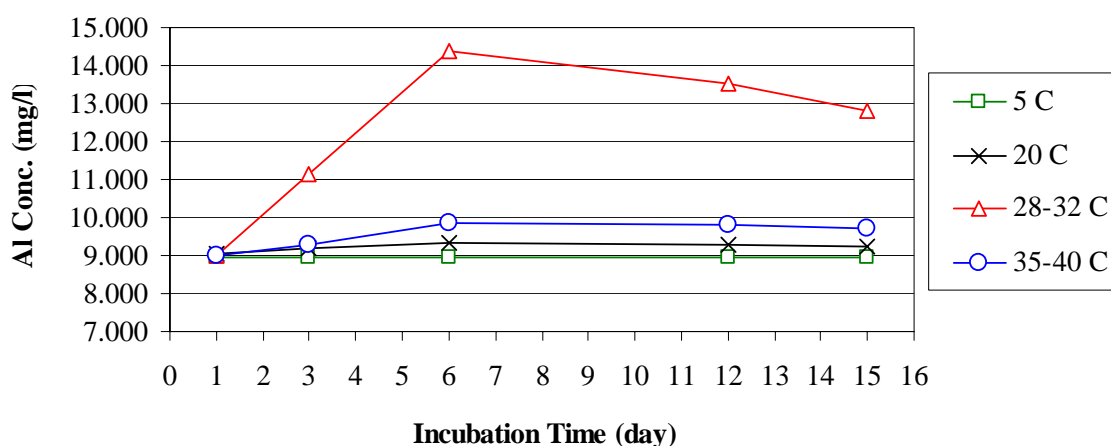
This amount of aluminum leaching (14.2 mg/l (14.2 ppm)) could inhibit plant root growth and may interfere with the acquisition and metabolism of mineral nutrients (Matsumoto et al., 1976, McLean and Gilbert, 1927). From Table 13, 12.22 ppm of aluminum in soil solution drastically reduced maize and millet root weight while the 50% cotton root length reduction resulted from only 0.5 ppm of aluminum concentration (Table 14). Therefore, the 14.2 ppm concentration would be significant on inhibit plant root growth. And if this amount of aluminum present in water, it will cause lethal death to gold fish within 1 to 10 hours (Table 15).

## 2.2. Effect of temperature on Al leaching from soil-sludge amendment

The optimum soil-sludge mixing ratio from previous experiment had been used (Rangsit soil-sludge amendment, 75:25) in this experiment. The moisture content of the

soil-sludge amendment samples were controlled at 30%. Temperature was varied from 5, 20, 28-32 and 35-40 °C.

Under 5, 20, and 35-40 °C incubation, it appeared that amounts of the aluminum leaching were quite stable along the incubation time. However, under 28-32 °C incubation amount of aluminum leaching increased sharply from initial day and reached the maximum level on the 6 day of incubation and then started to decrease gradually (Figure 22). From the results, amount of aluminum leaching from samples kept under 5, 20, and 35-40 °C were much lower than amount of aluminum leaching from samples kept under 28-32 °C. This resulted from effect of temperature which could increase microbiological activity (Department of Soil Science, 1998). It suggests that in tropical area like in Thailand temperature at approximately 30 °C is suitable for the growth of soil microbes which normally their activities producing  $H^+$  which increases soil acidity and subsequently the increasing of solubility of aluminum. For temperature of 5, 20, and 35-40 °C; which commonly suitable for the psychrophilic and thermophilic microorganisms respectively which are not the majority in Thailand climate. This may result to low activity of the microorganisms subsequently to low aluminum solubility (Department of Soil Science, 1998).



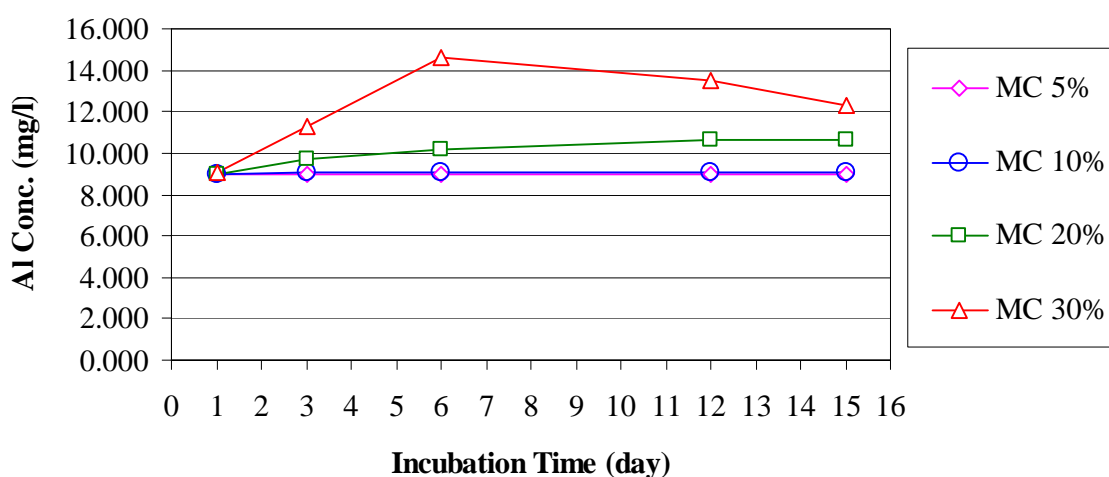
**Figure 22** Average values of effect of temperature on aluminum leaching

It can summarize that temperature was indirect effect on aluminum leaching especially under 28-32 °C, or in tropical area such as Thailand.

### 2.3. Effect of moisture content on Al leaching from soil-sludge amendment

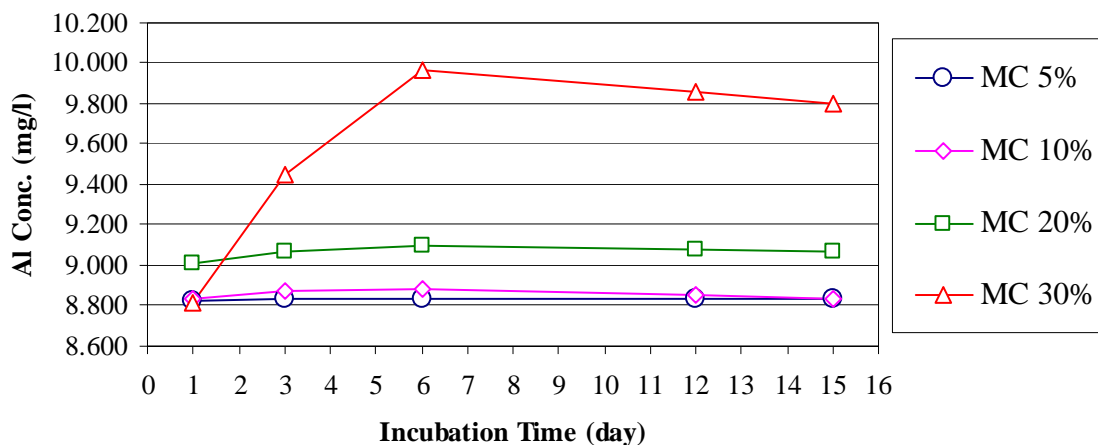
In this experiment, the 75:25% Rangsit soil-sludge mixing ratio was used. The soil-sludge amendment samples were kept under controlled two temperatures at 28-32 and 35-40 °C with varying moisture contents from 5, 10, 20, and 30% (initial moisture content). It was noted that the soil-sludge mixtures were left throughout incubation period without any additional water to maintain the initial moisture value.

Under 28-32 °C incubation, amounts of aluminum leaching concentration of the samples with moisture content of 5 and 10% were almost constant during the incubation period. At moisture content of 20%, the Al leaching gradually increased along with incubation period while at 30% moisture content the amount of aluminum leaching sharply increased and gradually decreased after it reached the maximum leaching level on the 6<sup>th</sup> incubation day (Figure 23).



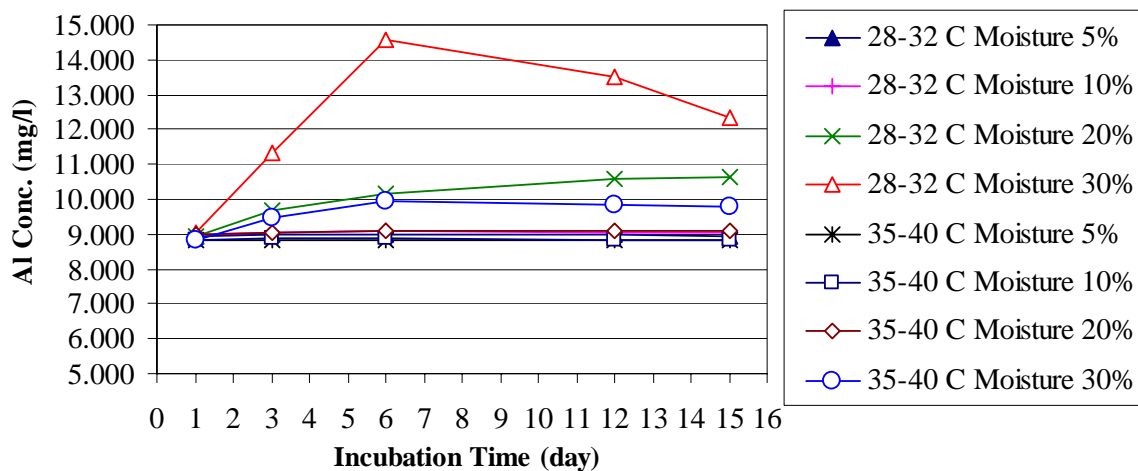
**Figure 23** Average values of effect of moisture content on aluminum leaching, incubated under 28-32 °C

Same trends of Al leaching of the 35-40 °C incubation, amount of aluminum leaching of the samples with moisture content 5, 10, and 20% were almost constant through the incubation period. In case of the samples with 30% moisture content, it was shown that the peak was seen on the 6 day of incubation and slightly decreasing thereafter (Figure 24).



**Figure 24** Average values of effect of moisture content on aluminum leaching, incubated under 35-40 °C

When comparing the results of both temperature incubation, the result shown that samples with moisture content 30% incubated under 28-32 °C gave highest amount of aluminum leaching while the same moisture content of 30% under 35-40 °C had lower aluminum leaching. This indicates that the temperature had more influence on Al leaching than the moisture content (Figure 25).



**Figure 25** Average values of effect of soil moisture content on aluminum leaching

From results of batch leaching experiments, Rangsit soil with soil-sludge mixing ratio of 75:25 with 30% moisture content gave highest amount of Al leaching. This may result from the soil pH which is acidic. Thus, when adding 75% soil mixed with 25% sludge, soil pH reduced the overall pH value of the amendment which also resulted in high amount of Al leaching compared with the others soil textures and the other soil-sludge mixing ratios.

In the opposite way, Korat soil gave the lowest amount of Al leaching. As Korat soil itself had high pH value which was resulted from over-liming thus, aluminum in the soil-sludge amendment may be fixed rather than leaching out. The results from batch experiment that used Korat soil supported this reason which showing that soil-sludge mixing ratio with 75% Korat soil and 25% sludge gave the lowest amount of Al leaching. This may because of the higher ratio of sludge used could reduce the mixture pH value.

For the 30% moisture content which also gave the highest amount of Al leaching, this may result from the 30% moisture content is the most suitable moisture for soil microbes.

The result indicates that soil moisture content affected on aluminum leaching too, low soil moisture content gave low amount of Al leaching. Soil moisture acted as media of chemical reaction in soil and also biochemical reaction in microbes. From the experiment results, 75:25%

Rangsit soil-sludge amendment with 30% moisture content under 28-32 °C temperature resulted in maximum 14.5 mg/l aluminum leaching.

However, as the moisture content of the soil-sludge mixtures were controlled only on the initial day of incubation to desired value and without additional adjustment throughout the incubation period. Therefore, this might result in losing moisture content of the mixture under high incubation temperature (28-32 and 35-40 °C). Water in the mixtures could evaporate into atmosphere resulting in the relative humidity in the incubation cup increasing while the mixture moisture content decreasing. This might result in slowing down the rate of chemical reaction and microbial activities subsequently in lowering amount of aluminum leaching.

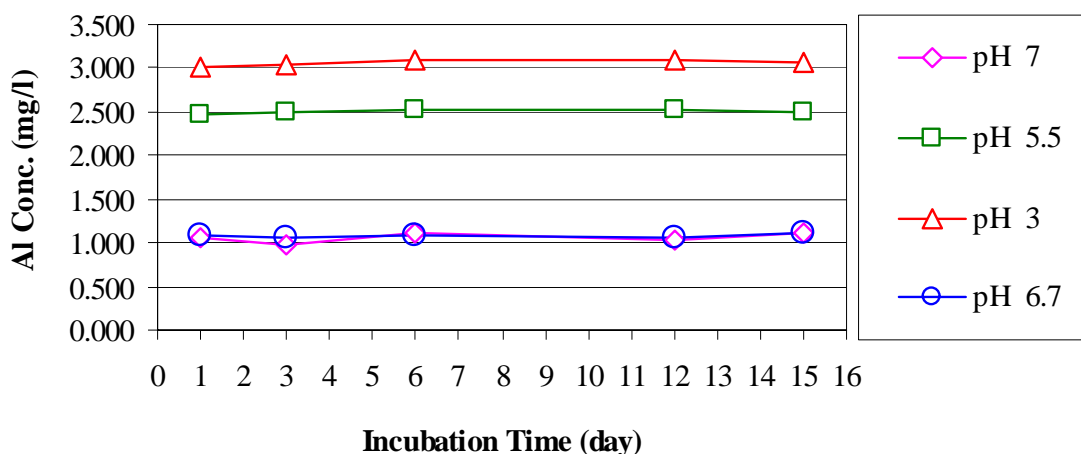
Source of extractable aluminum from Rangsit soil-sludge mixtures might be mainly from the sludge under effect of soil acidity, temperature, and moisture content. Although amount of extractable aluminum from Rangsit soil was rather high (5.66-7.55 mg/l) as compared to amount of extractable aluminum from the sludge (0.61-2.18 mg/l) however, the amount of extractable aluminum from Rangsit soil-sludge mixtures were even higher (~14 mg/l) while the overall pH value of the mixture would have been higher than pH of soil alone. This indicated that the excessive amount of aluminum leached from Rangsit soil-sludge mixture should be from the sludge.

### **3. Column Leaching Experiment**

For column leaching experiment, Rangsit soil-sludge amendment with the mixing ratio of 75:25 was used. The soil-sludge mixture were packed into the column (12 columns) and incubated under room temperature (28-32 °C) for 15 days. During the incubation period, triplicate columns were rinsed with rain water with varies of pH values (7, 6.7, 5.5, and 3) every 1, 3, 6, 12, and 15 day of incubation period and the leachates were collected and analyzed for aluminum leaching in the same sampling day.

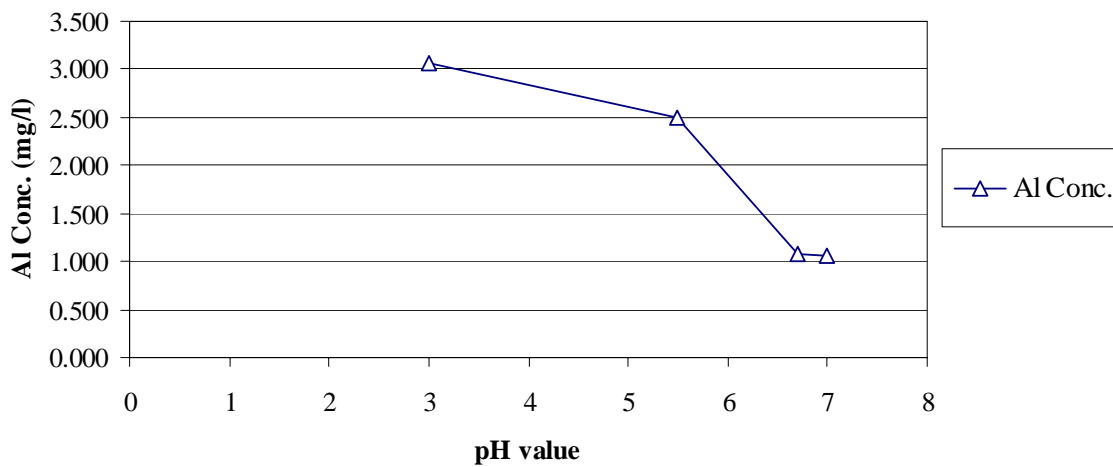
The results showed that all columns had been giving the stability of Al leaching during 15 days of experiment period. Amount of aluminum leaching of natural rain water (pH 6.7) and artificial rain water (pH 7) had small amount of Al leaching (~ 1 mg/l). For pH 5.5, amount of aluminum leaching was higher than pH 7 but, also in low level (~ 2.5 mg/l). In this experiment, amount of aluminum leaching from artificial rain water pH 3 was the highest (~3.5 mg/l) as shown in Fig. 26.

From the results of this experiment, it indicates that acid rain could cause aluminum leaching significantly from soil-sludge amendment. The increase in acidity of rainwater could increase in the Al leaching as same as soil acidity.



**Figure 26** Average values of effect of rain pH on aluminum leaching

From the result of column leaching experiments, it is obvious that pH value has great effect on Al leaching. The lower the pH value, the higher the concentration of soluble aluminum appeared (Figure 27) (McBride, 1994).



**Figure 27** Effect of rainwater pH on concentration of aluminum leaching

From the experiment results, it can be summarized that soil acidity and rain acidity effected in aluminum leaching. However, the amount of aluminum leached out from soil-sludge mixtures under acid soil condition was higher than those from strong acid rain. This may be assumed as follow;

1. Amount of  $Al^{3+}$  from batch leaching experiment may be gained from;

- Available of  $Al^{3+}$  in soil solution from soil acidity. As the soil acidity results mainly from exchangeable aluminum in soil (dissociation of aluminum compound in soil, one aluminum ion gives three hydrogen ions). This results in availability of free  $Al^{3+}$  in soil solution.

- The chemical reaction on replacement of  $K^+$  from KCl extractant solution with aluminum in soil-sludge mixture which resulted in solubilization of aluminum from permanently charged surface and other nonexchangeable sources.

2. Amount of  $\text{Al}^{3+}$  from column leaching experiment may be gained from;

- Exchangeable aluminum in soil solution and the permanently charged surface. As under leaching condition, rain water will wash out cations in soil solution ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^{2+}$ ,  $\text{Al}^{3+}$ , etc.) from soil surface and sub-soil and displace these cations with hydrogen ions.

Therefore, the amount of aluminum leaching from column leaching experiment was lower than in batch leaching experiment.

## CONCLUSIONS

1. Soil texture effects on aluminum leaching as indirectly resulted from the soil pH value. From the batch experiment, Rangsit soil, which had lowest pH (4.29-4.43), gave highest amount of aluminum leaching as compared with the other 2 soil types. And among the 3 different soil-sludge mixing ratios of Rangsit soil-sludge mixing, the mixing ratio of 75:25 gave the outstanding amount of aluminum leaching. This may result from soil pH value induced the lower pH value of the soil-sludge mixture which result in leaching of aluminum from the mixture.

Therefore, to dispose the sludge by landfilling on such soil type should be carefully considered about possibility on aluminum leaching in future land application on agriculture and possibility on aluminum accumulation in plant.

2. Effect of temperature on aluminum leaching, even if the amount of aluminum leached out from the soil-sludge mixture was not high however, at 28-32 °C the amount of aluminum leaching was higher than the experiment under the other temperature.

The temperature may not be a direct factor on aluminum leaching. But, temperature effects on microorganisms growth and this growing activity of the microorganisms resulted in releasing of H<sup>+</sup> ions. Therefore, the releasing of H<sup>+</sup> ions lowering soil pH and finally resulted in increasing of amount of aluminum leaching.

3. Soil moisture content effects on aluminum leaching. The experiment under low soil moisture content (5%, 10%, and 20%) gave low amount of aluminum leaching (~9.0-10.6 mg/l) while 30% moisture content gave highest amount of aluminum leaching.

Soil moisture content in association with temperature had strong effect on amount of aluminum leaching, as the temperature (28-32 °C) is suitable for microorganism growth and moisture content (30%) is enough for soil chemical reaction media. Thus, these 2 factors promotes aluminum solubility and results in aluminum leaching.

4. From column leaching experiment, pH value of rain water effects on aluminum leaching especially low pH value. Rain water pH of 6.7-7 had small effect on aluminum leaching while acid rain (pH 5.5 and lower) gave higher amount of aluminum leaching (~3.5 mg/l at pH 3).

Consideration on individual factor, each of them may have not such extreme effect on aluminum leaching but, in reality environment where the sludge may be disposed there are factors associated together. On any area there are temperature, rain pH, soil texture, soil pH, soil moisture content, soil microbial activities, etc. These factors are all affecting on each other and each and all of them may have strong effect on chemical and biological reactions resulting in aluminum leaching into environment.

### **RECOMMENDATION**

Sludge disposal by land application or landfilling or even soil amendment using sludge as fertilizer should be carefully considered. Even if the sludge itself has rather high organic matter and some other element composition which may be useful for agriculture as plant nutrient but, there is aluminum which could be toxic to plant and aquatic life if it leaches into environment in some certain amount. And this aluminum could be leached out under acidic condition. Therefore, sludge disposal by landfilling may better be done on alkaline soil area than acidic soil area such as the Northeastern of Thailand to minimized amount of aluminum leaching into environment.

Thus, sludge disposal or soil-sludge amendment should be controlled especially; environment's acidic factor.

### **FUTURE STUDY**

1. Study on effect of sludge application on aluminum accumulation/aluminum toxicity in plants and possibility of sludge usage.
2. Study on effect of soil microorganisms on aluminum leaching.
3. Study on long-term pH changing of soil and amount of aluminum leaching in landfilling area.
4. Study on long-term effect on sludge disposal in alkaline soil/environment to reduce possibility of aluminum leaching into environment.
5. Field study on effect of acid rain and environment such as sunlight, ambient air, organic matter, etc. on aluminum leaching.
6. Study on species/forms of aluminum and its compound in sludge and in soil-sludge mixture.

### LITERATURE CITED

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## **APPENDIX**

## APPENDIX A

### Calculations

## APPENDIX A

1. Aluminum extraction reagent, 1 M KCl;

M = Molar solution  
 = Number of equivalent weight / Litre

Number of equivalent weight = Reagent weight / equivalent weight

Equivalent weight = Form weight / cation charge

KCl =  $K^+ + Cl^-$  (K = 39, Cl = 35.5)

Thus, equivalent weight =  $74.5 / 1$   
 = 74.5

KCl 74.5 g has number of equivalent weight;  
 =  $74.5 / 74.5$   
 = 1

Therefore, 1M KCl is from weighing 74.5 g KCl then, added water to 1 L.

2. Calculation for aluminum using aluminum standard curve;

Using 1 ml of Al standard solution (1 ml standard solution = 1 mg Al)

From  $M_1V_1 = M_2V_2$   
 Then,  $M_1 = 200 \text{ mg Al/l}$   
 $V_1 = 1 \text{ ml.}$

$$V_2 = 40 \text{ ml. (volume used for Spectrophotometry analysis)}$$

$$\begin{aligned} \text{Therefore, } M_2 &= \frac{(1 \text{ mg Al/l}) \times (1 \text{ ml})}{40 \text{ ml}} \\ &= 0.025 \text{ mg Al/l} \end{aligned}$$

In the extraction, extracted volume of Al was 10 ml.

$$\begin{aligned} \text{From } X_1 V_1 &= X_2 V_2 \\ \text{So, } X_1 &= 0.025 \text{ mg Al/l} \\ V_1 &= 40 \text{ ml. (final prepared volume)} \\ V_2 &= 10 \text{ ml.} \end{aligned}$$

$$\begin{aligned} \text{Hence, } X_2 &= \frac{(0.025 \text{ mg Al/l}) \times (40 \text{ ml})}{10 \text{ ml}} \\ &= 0.1 \text{ mg Al/l} \end{aligned}$$

Volume of KCl used in the extraction was 15 ml.;

$$\begin{aligned} \text{From } M_1 V_1 &= M_2 V_2 \\ \text{Then, } M_1 &= 0.1 \text{ mg Al/l} \\ V_1 &= 10 \text{ ml (prepared volume)} \\ V_2 &= 15 \text{ ml} \end{aligned}$$

$$\begin{aligned} \text{Therefore, } M_2 &= \frac{(0.1 \text{ mg Al/l}) \times (10 \text{ ml})}{15 \text{ ml}} \\ &= 0.067 \text{ mg Al/l} \end{aligned}$$

Amount of aluminum extracted from soil sample is 0.067 mg Al/l.

3. Calculation for amount of sludge disposal per m<sup>2</sup>;

Soil : Sludge Mixing Ratio (Base on 100 g. dry weight of the mixture)

Soil	Ratio	25 : 75		50 : 50		75 : 25	
		Soil (g)	Sludge (g)	Soil (g)	Sludge (g)	Soil (g)	Sludge (g)
Korat		29.16	162.11	58.31	108.08	87.47	54.04
Rangsit		39.11	162.11	78.23	108.08	117.34	54.04
Nonthaburi		32.08	162.11	64.17	108.08	96.25	54.04

Estimated soil density;

1. Korat soil (sandy soil)  $\approx$  0.8 kg/l
2. Rangsit soil (clay soil)  $\approx$  1.2 kg/l
3. Nonthaburi soil (Silt soil)  $\approx$  1.0 kg/l

$$\text{From Volume} = \text{Mass/Density}$$

For Korat soil : sludge mixing ratio 25 : 75;

$$\begin{aligned} \text{Then, Soil volume (Liter)} &= (29.16\text{g}/1000\text{g}) \text{ kg}/0.8 \text{ kg/l} \\ &= 3.645 \times 10^{-2} \text{ Liter} \\ 1 \text{ Liter} &= 10^{-3} \text{ m}^3 \end{aligned}$$

$$\text{Therefore, } 3.645 \times 10^{-2} \text{ Liter} = 3.645 \times 10^{-5} \text{ m}^3$$

$$\text{From Area} = \text{Volume/Depth}$$

When depth of soil = 10 cm (0.1 m);

$$\begin{aligned} \text{Disposed area (m}^2\text{)} &= 3.645 \times 10^{-5} \text{ m}^3 / 0.1 \text{ m} \\ &= 36.45 \times 10^{-5} \text{ m}^2 \end{aligned}$$

Area  $36.45 \times 10^{-5} \text{ m}^2$  disposed sludge (from table above) 162.11 g (0.162 kg)

$$\begin{aligned} \text{Therefore, area } 1 \text{ m}^2 \text{ disposed sludge} &= 0.162 / (36.45 \times 10^{-5}) \\ &= 444.44 \text{ kg} \end{aligned}$$

Thus, for Korat soil with soil : sludge mixing ratio 25 : 75 is equivalent to sludge disposal on Korat soil =  $444.44 \text{ kg/m}^2$

#### 4. Unit conversion from cmol/kg and meq/100g into ppm;

$$\begin{aligned} \text{From} \quad \text{cmol/kg} &= \text{meq/100 g} \\ \text{Then,} \quad 0.03 \text{ cmol Al/mg} &= 0.03 \text{ meq Al/100 g} \\ \\ \text{Equivalent weight of Al} &= 27/3 \\ \text{Therefore,} \quad 0.03 \text{ meq Al/100 g} &= \frac{0.03 \text{ g Al/100 g}}{(27/3) \times 1,000} \\ &= 3.333 \times 10^{-6} \text{ g Al/100 g} \end{aligned}$$

In 100 g soil there is  $3.333 \times 10^{-6} \text{ g Al}$

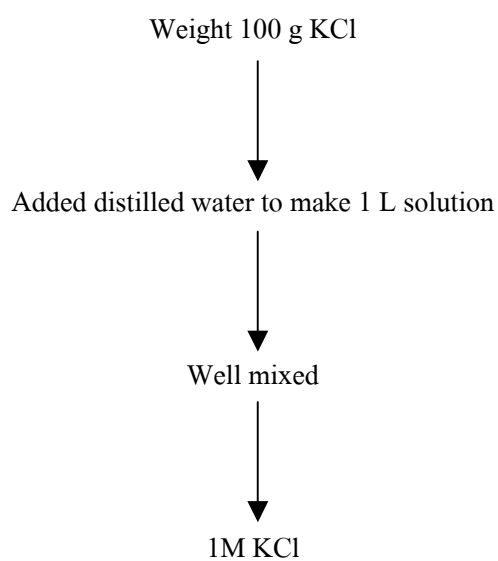
$$\begin{aligned} \text{So, in } 1,000,000 \text{ g soil there will be} & \frac{3.333 \times 10^{-6} \text{ g Al} \times 1,000,000 \text{ g soil}}{100 \text{ g soil}} \\ &= 0.033 \text{ g Al} \\ &= 0.033 \text{ ppm} \end{aligned}$$

## APPENDIX B

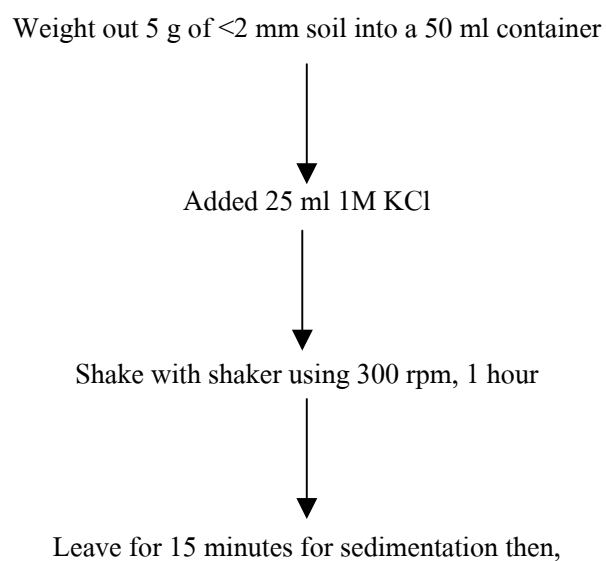
### Analysis methods

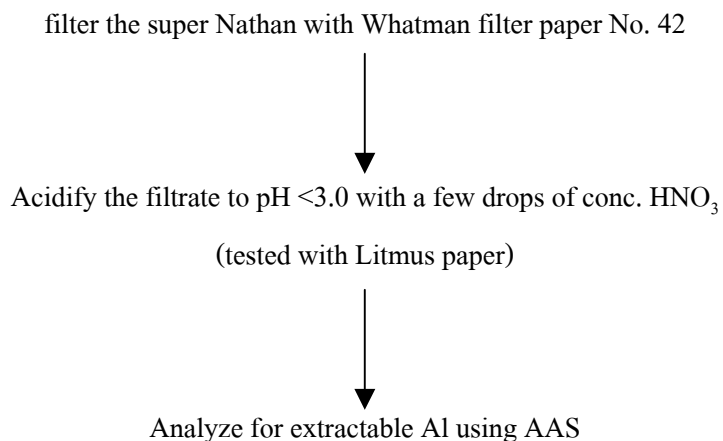
**APPENDIX B****Analysis methods**

## 1. Preparation of 1M KCl

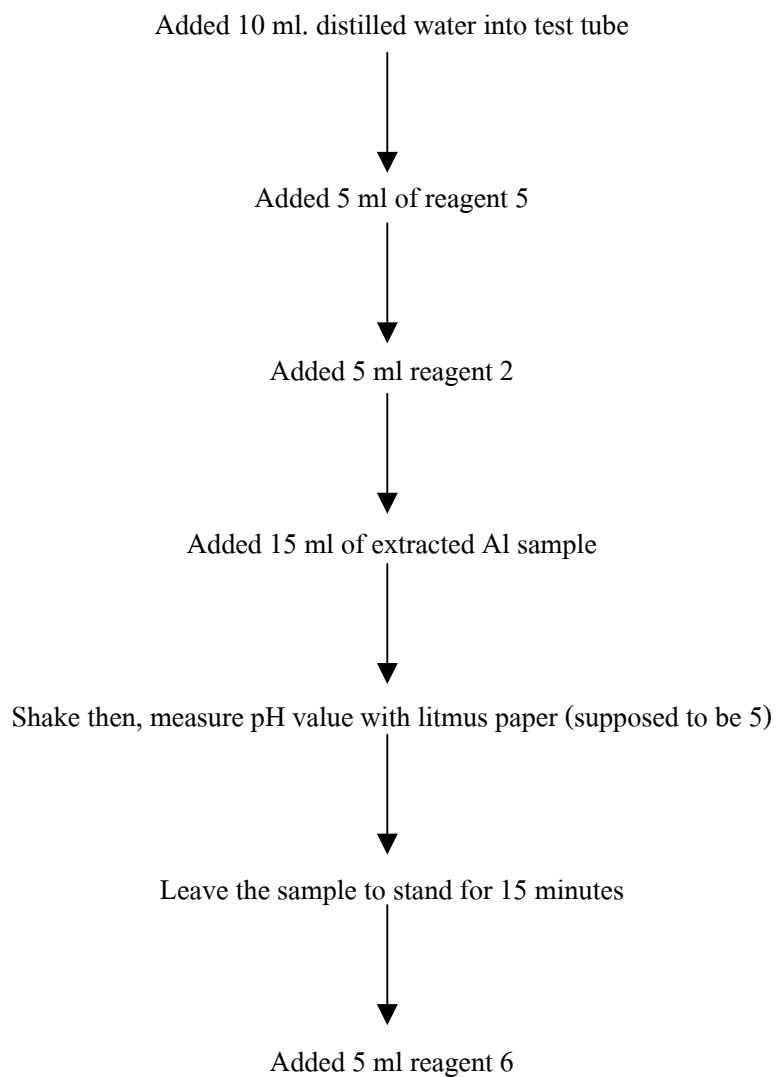


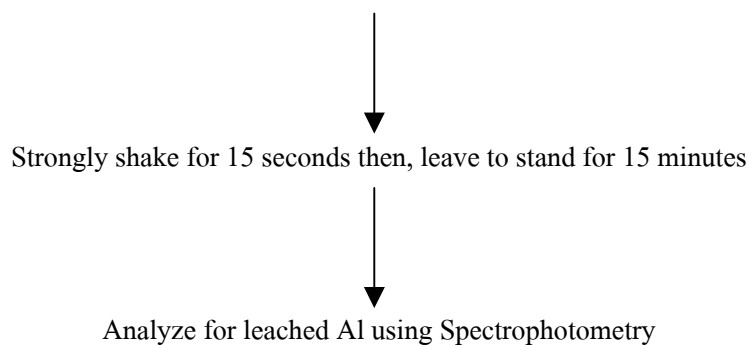
## 2. Aluminum extraction from soil-sludge amendment samples (Bertsch and Bloom, 1996)



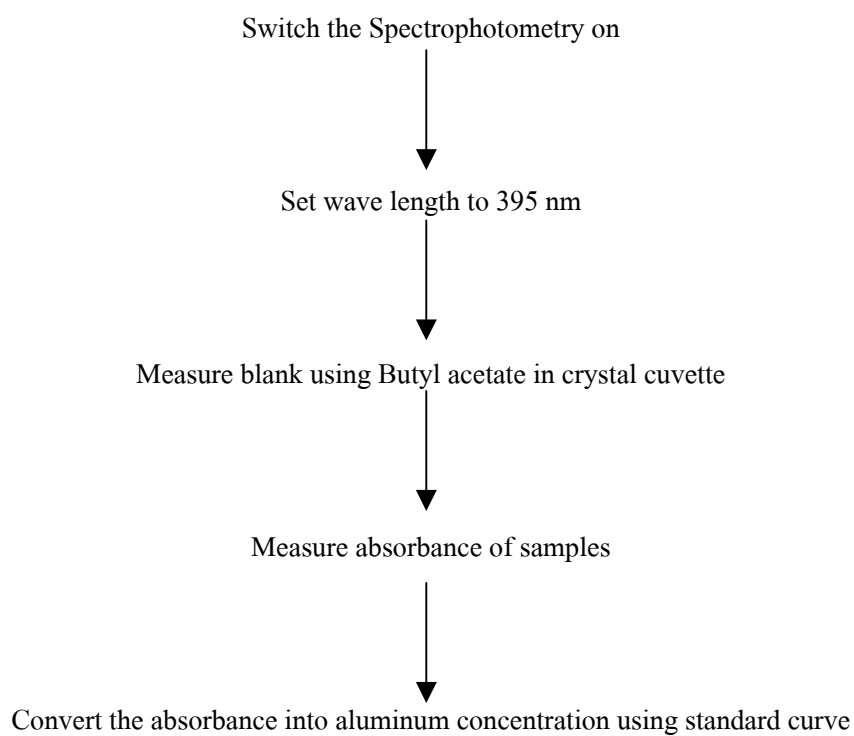


### 3. Sample preparation for Al analysis using Spectrophotometry (Bertsch and Bloom, 1996)

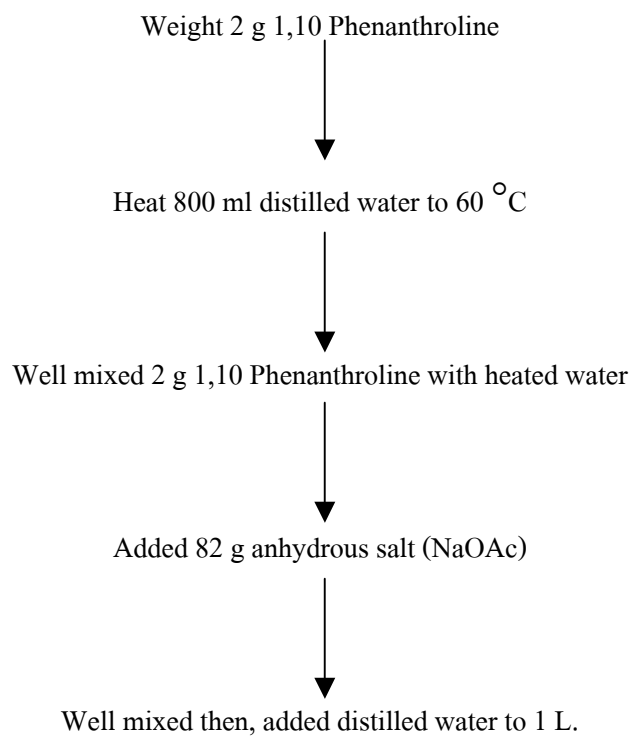




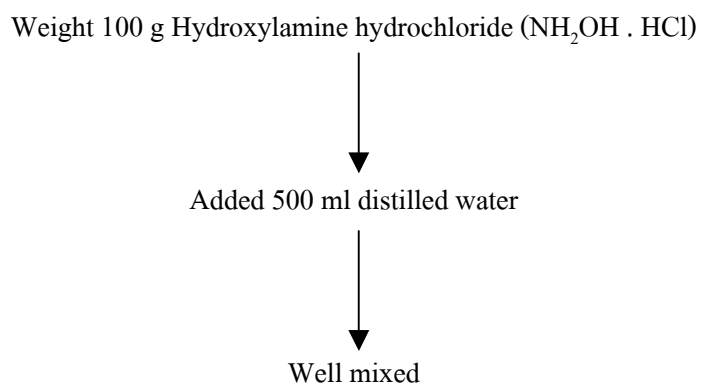
#### 4. Aluminum analysis using Spectrophotometry



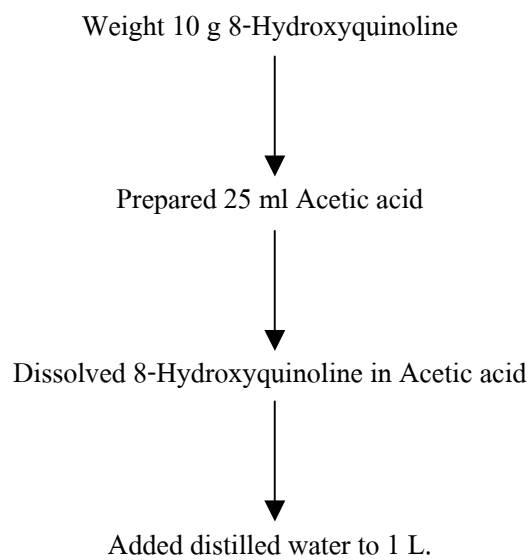
## 5. Preparation of reagent 2



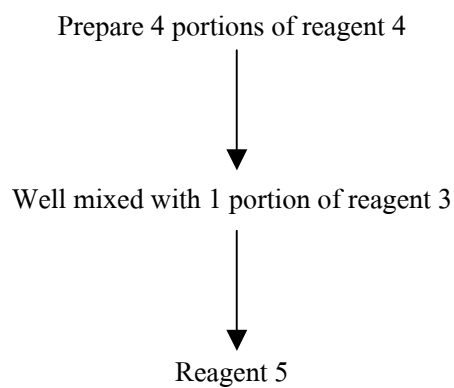
## 6. Preparation of reagent 3



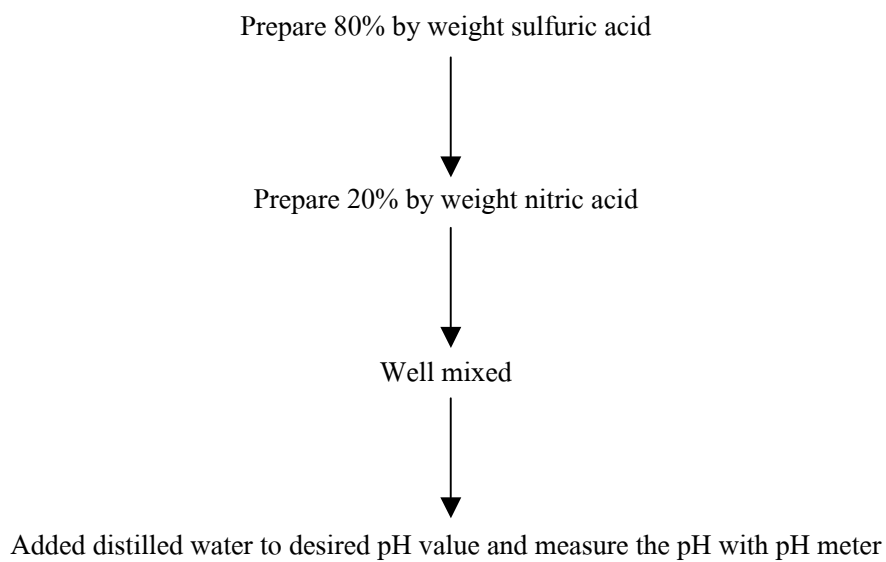
## 7. Preparation of reagent 4



## 8. Preparation of reagent 5



9. Preparation of artificial acid rain (Synthetic acid rain extraction fluid', ministry of industry announcement copy No. 6 (1997)



## APPENDIX C

Characteristics and properties of water treatment sludge, soils, and rain water

## APPENDIX C

### 1. Properties and characteristics of Bangkhen water treatment sludge

Appendix TableC1 Characteristics of Bangkhen watertreatment sludge

Test	pH Value	Moisture Content (%)	Organic Matter (%)	Total Solid (%)
1	6.52	53.3631	8.7069	47.4290
2	6.58	53.2686	8.6386	47.1403
3	6.60	50.7797	8.8502	46.3471
4	6.58	54.0161	8.6516	46.2739
5	6.56	54.0290	8.8625	47.5683
Average	6.57	53.0913	8.7420	46.9517

Appendix TableC 2 Particle size distribution of Bangkhen watertreatment sludge

Sample No.	Particle Size Distribution, g/kg (%)		
	Sand (2-0.05 mm.)	Silt (0.05-0.002 mm.)	Clay (< 0.002 mm.)
1	13.34 (1.34)	740.46 (74.05)	246.20 (24.62)
2	28.23 (2.82)	651.16 (65.12)	320.61 (32.06)
3	36.05 (3.61)	505.21 (50.52)	458.74 (45.87)
4	24.32 (2.43)	593.16 (59.32)	382.52 (38.25)
5	29.71 (2.97)	662.29 (66.23)	308.00 (30.80)
Average	26.32 (2.63)	630.48 (63.05)	343.21 (34.32)

Remark: Particle size distribution criteria according to United State Department of Agriculture, USDA) in g/kg

- Particle size
- Sand: 2-0.002 mm.
  - Silt: 0.05-0.002 mm.
  - Clay: <0.002 mm.

Appendix Table C3 Particle density

Sample No.	Particle Density (g/cm <sup>3</sup> )
1	2.5816
2	2.5738
3	2.5723
4	2.5736
5	2.5780
Average	2.5759

Appendix Table C4 Element composition of Bangkhen water treatment sludge

Sludge : Composition (g/kg)

Element	C	O	Na	Mg	Al	Si	K	Ca	Ti	Fe
Test 1	0.00	556.46	3.71	8.12	134.94	233.71	14.03	5.10	4.29	39.65
Test 2	0.00	550.51	3.84	8.56	126.63	241.47	15.46	5.23	4.28	44.01
Test 3	0.00	547.01	3.47	7.80	139.11	239.68	14.45	5.44	4.13	38.92
Average	0.00	551.33	3.67	8.16	133.56	238.29	14.65	5.26	4.23	40.86

## **2. Properties and characteristics of soils**

### 2.1 Nonthaburi soil;

Appendix Table C5 Characteristics of Nonthaburi soil

Test	pH Value	Moisture Content (%)	Organic Matter (%)	Total Solid (%)
1	5.20	22.4527	6.1403	77.5473
2	5.19	21.7648	6.2359	78.2352
3	5.24	22.0487	6.1834	77.9781
4	5.16	23.1641	6.2986	78.2915
5	5.27	20.9165	6.0634	79.3729
Average	5.21	22.0694	6.1843	78.2850

Appendix Table C6 Element composition of Nonthaburi soil

Nonthaburi Soil : Composition (g/kg)

Element	C	O	Na	Mg	Al	Si	K	Ca	Ti	Fe
Test 1	0.00	612.18	4.94	6.43	103.62	186.44	11.83	3.33	5.86	65.36
Test 2	0.00	586.70	4.57	6.97	99.90	220.03	12.69	3.54	6.40	59.21
Test 3	0.00	597.51	4.30	7.13	105.44	198.30	14.27	3.85	7.25	61.95
Average	0.00	598.79	4.61	6.85	102.98	201.59	12.93	3.58	6.50	62.17

## 2.2 Rangsit Soil;

Appendix Table C7 Characteristics of Rangsit soil

Test	pH Value	Moisture Content (%)	Organic Matter (%)	Total Solid (%)
1	4.33	33.6139	10.0955	66.3861
2	4.40	34.3056	10.1493	64.5763
3	4.30	39.1428	10.1156	60.7854
4	4.43	32.9743	10.1279	63.0312
5	4.29	37.8641	10.0343	65.6925
Average	4.35	35.5801	10.1045	64.0943

Appendix Table C8 Element composition of Rangsit soil

## Rangsit Soil : Composition (g/kg)

Element	C	O	Na	Mg	Al	Si	S	K	Ca	Ti	Fe
Test 1	0.00	581.61	3.14	7.22	92.90	250.17	16.53	13.27	4.19	5.12	25.84
Test 2	0.00	558.78	6.46	8.35	104.10	261.86	14.47	11.24	3.67	6.90	24.16
Test 3	0.00	602.55	2.93	6.31	99.17	231.80	14.20	11.83	3.38	5.30	22.54
Average	0.00	580.98	4.18	7.29	98.72	247.94	15.07	12.12	3.75	5.77	24.18

## 2.3 Korat soil;

Appendix Table C9 Characteristics of Korat soil

Test	pH Value	Moisture Content (%)	Organic Matter (%)	Total Solid (%)
1	8.37	14.9506	2.9021	85.1023
2	8.22	14.4162	2.9235	85.5860
3	8.35	13.4622	2.9370	86.5374
4	8.28	14.2704	2.7638	83.8635
5	8.23	14.1684	3.0251	85.8539
Average	8.29	14.2535	2.9103	85.7419

Appendix Table C10 Element composition of Korat soil

Korat Soil : Composition (g/kg)

Element	C	O	Na	Mg	Al	Si	K	Ca	Ti	Fe
Test 1	0.00	494.92	4.85	7.45	100.47	316.10	14.79	6.66	5.08	49.67
Test 2	0.00	518.00	5.79	8.22	91.54	300.43	14.58	7.29	5.55	48.61
Test 3	0.00	500.68	4.22	6.38	98.36	305.90	15.27	6.84	5.93	56.42
Average	0.00	504.53	4.95	7.35	96.79	307.48	14.88	6.93	5.52	51.56

Remark: Korat Soil is excessive liming treatment soil from Pak-Chong Distric, Nakorn

Ratchasima Province

### **3. Characteristic of natural rain water**

Appendix Table C11 pH values of natural rain water

Test	pH Value
1	6.73
2	6.74
3	6.63
4	6.72
5	6.67
Average	6.70

### **4. Extractable aluminum from soils and sludge samples**

Appendix Table C12 Extractable aluminum from soils and sludge samples

Extractable Al (mg/l)	Sludge	Nonthaburi soil	Rangsit soil	Korat soil
Test I	0.610	5.390	5.655	1.700
Test II	2.180	5.145	6.040	1.390
Test III	2.105	7.675	7.550	1.330
Average	1.632	6.070	6.415	1.473

## APPENDIX D

Experiments result

## APPENDIX D

### 1. Batch experiments

#### 1.1 Effect of soil texture on aluminum leaching;

Appendix Table D1 Nonthaburi soil

Sampling day	Al Conc. (mg/l)	Test I			Test II		
		25:75	50:50	75:25	25:75	50:50	75:25
Initial	1.880	2.500	3.215	1.960	2.730	3.510	
Day 3	2.015	2.570	3.640	2.105	2.840	3.770	
Day 6	2.055	3.665	4.875	2.185	3.510	4.740	
Day 12	2.490	4.460	5.920	2.530	4.580	5.085	
Day 15	2.360	3.845	4.990	2.330	4.085	4.560	

Appendix Table D2 Rangsit soil

Sampling day	Al Conc. (mg/l)	Test I			Test II		
		25:75	50:50	75:25	25:75	50:50	75:25
Initial	1.930	3.690	8.605	2.585	4.450	9.565	
Day 3	2.025	4.610	10.190	2.725	5.515	11.480	
Day 6	2.500	4.990	13.315	3.105	5.830	15.100	
Day 12	2.685	5.540	11.660	3.190	5.965	13.395	
Day 15	2.605	5.115	11.355	3.100	6.105	12.175	

Appendix Table D3 Korat soil

Sampling day	Al Conc. (mg/l)	Test I			Test II		
		25:75	50:50	75:25	25:75	50:50	75:25
Initial	1.620	1.290	1.090	1.320	1.170	0.995	
Day 3	1.865	1.470	1.125	1.410	1.380	1.040	
Day 6	1.950	1.705	1.365	1.595	1.495	1.165	
Day 12	2.105	1.995	1.620	1.735	1.625	1.335	
Day 15	2.065	2.025	1.550	1.650	1.560	1.420	

## 1.2 Effect of Temperature on aluminum leaching;

Appendix Table D4 Temperature 5 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.945	8.980	8.870
Day 3	8.940	8.975	8.895
Day 6	8.945	8.990	8.905
Day 12	8.955	8.995	8.900
Day 15	8.950	9.000	8.890

Appendix Table D5 Temperature 20 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	9.025	9.110	9.075
Day 3	9.110	9.230	9.205
Day 6	9.305	9.350	9.390
Day 12	9.260	9.325	9.280
Day 15	9.180	9.245	9.235

Appendix Table D6 Temperature 28-32 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.975	9.005	9.055
Day 3	11.540	11.070	10.875
Day 6	13.930	14.415	14.845
Day 12	13.485	13.445	13.655
Day 15	12.820	12.885	12.785

Appendix Table D7 Temperature 35-40 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	9.025	9.010	8.995
Day 3	9.170	9.560	9.090
Day 6	9.990	9.925	9.630
Day 12	9.805	9.865	9.745
Day 15	9.865	9.750	9.580

## 1.3 Effect of moisture content on aluminum leaching;

Appendix Table D8 Moisture content 5%, temperature 28-32 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.975	8.965	8.940
Day 3	9.000	8.950	8.955
Day 6	9.015	8.980	8.950
Day 12	8.980	8.960	8.960
Day 15	8.965	8.950	8.930

Appendix Table D9 Moisture content 10%, temperature 28-32 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.965	8.985	9.025
Day 3	9.050	9.065	9.070
Day 6	9.065	9.050	9.100
Day 12	9.050	9.045	9.085
Day 15	9.030	9.055	9.095

Appendix Table D10 Moisture content 20%, temperature 28-32 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	9.025	8.880	9.040
Day 3	9.915	9.455	9.330
Day 6	10.135	10.160	9.965
Day 12	10.695	10.500	10.375
Day 15	10.840	10.395	10.510

Appendix Table D11 Moisture content 30%, temperature 28-32 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	9.115	9.040	8.990
Day 3	11.530	11.600	10.780
Day 6	14.765	14.735	14.200
Day 12	13.570	13.940	13.065
Day 15	12.965	12.560	11.505

Appendix Table D12 Moisture content 5%, temperature 35-40 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.820	8.815	8.835
Day 3	8.825	8.825	8.84
Day 6	8.825	8.835	8.84
Day 12	8.830	8.830	8.835
Day 15	8.825	8.835	8.830

Appendix Table D13 Moisture content 10%, temperature 35-40 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.845	8.850	8.815
Day 3	8.875	8.905	8.845
Day 6	8.865	8.920	8.860
Day 12	8.835	8.895	8.825
Day 15	8.790	8.880	8.840

Appendix Table D14 Moisture content 20%, temperature 35-40 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	9.035	9.005	8.975
Day 3	9.100	9.075	9.030
Day 6	9.120	9.090	9.070
Day 12	9.105	9.095	9.045
Day 15	9.110	9.100	9.005

Appendix Table D15 Moisture content 30%, temperature 35-40 °C

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	8.825	8.850	8.770
Day 3	9.500	9.475	9.360
Day 6	9.965	10.000	9.930
Day 12	9.945	9.880	9.740
Day 15	9.735	9.890	9.785

## 2. Column leaching experiment

Appendix Table D16 pH 7

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	0.969	1.155	1.068
Day 3	0.861	1.113	0.942
Day 6	0.930	1.236	1.146
Day 12	0.978	1.083	1.071
Day 15	0.903	1.185	1.209

Appendix Table D17 pH 6.7

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	1.083	1.047	1.122
Day 3	1.074	0.990	1.134
Day 6	1.107	1.014	1.143
Day 12	1.089	0.996	1.128
Day 15	1.110	1.047	1.167

Appendix Table D18 pH 5.5

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	2.466	2.493	2.484
Day 3	2.478	2.499	2.499
Day 6	2.499	2.538	2.532
Day 12	2.490	2.517	2.535
Day 15	2.505	2.502	2.493

Appendix Table D19 pH 3

Sampling day	Al Conc. (mg/l)		
	Test I	Test II	Test III
Initial	3.009	2.994	3.000
Day 3	3.024	3.039	3.036
Day 6	3.069	3.126	3.090
Day 12	3.084	3.114	3.072
Day 15	3.057	3.087	3.039

Appendix Table D20 Average value of aluminum concentration VS pH value

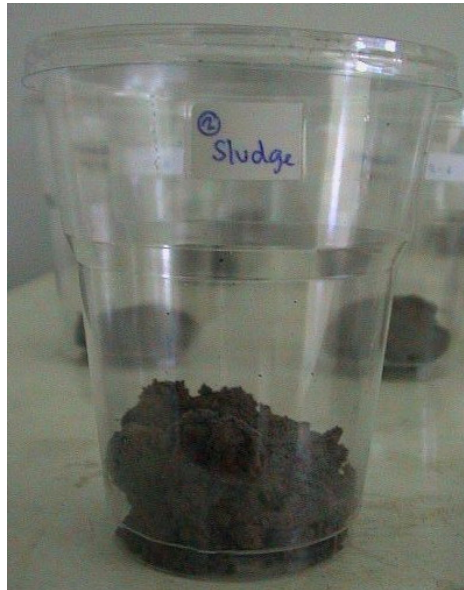
Sampling day	Average Al Conc. (mg/l)			
	pH 7	pH 6.7	pH 5.5	pH 3
Initial	1.064	1.084	2.481	3.001
Day 3	0.972	1.066	2.492	3.033
Day 6	1.104	1.088	2.523	3.095
Day 12	1.044	1.071	2.514	3.090
Day 15	1.099	1.108	2.500	3.061

## APPEDIX E

### Figures

**APPENDIX E****Pictures**

1. Sludge;

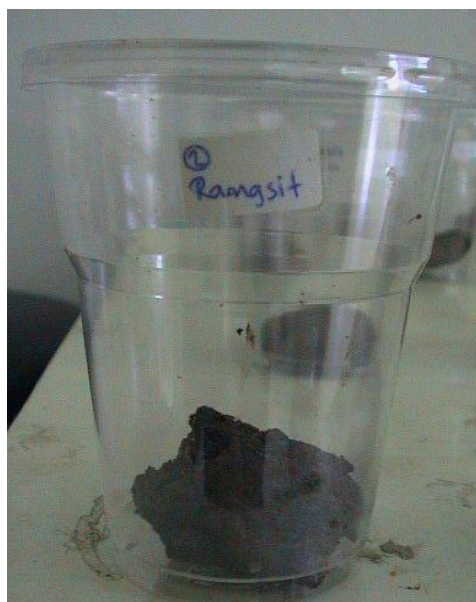


Appendix Figure E1 Bangkhen water treatment sludge

## 2. Soil;



Appendix Figure E2 Nonthaburi soil



Appendix Figure E3 Rangsit soil



Appendix Figure E4 Korat soil



Appendix Figure E5 Soil-sludge amendment incubation



Appendix Figure E6 Air-drying of soil-sludge amendment

3. Column;



Appendix Figure E7 Columns



Appendix Figure E8 Column separator to ease column cleaning



Appendix Figure E9 Peripheral plate



Appendix Figure E10 Sand layer on inside column wall



Appendix Figure E11 Funnel

