Original Research Article

Study of the effects of soil acidity and salinity on aluminium mobility in selected soil samples in Sri Lanka

Abstract

Aluminium is the most abundant metal in the earth's crust. Aluminium in soil is found to be incorporated into aluminosilicate soil minerals and this aluminium is in stable inactive form. Very minute quantities appear in soluble forms capable of influencing biological systems. Aluminium mobility and as a consequence aluminium toxicity, is mainly restricted to acid environments. Aluminium has low mobility under most environmental conditions. However, below a pH of 4.0 its solubility increases and aluminium is released from silicate rocks under such acidic conditions. The levels of dissolved aluminium in natural groundwater samples are generally low, probably due to its low solubility at neutral pH values. Release of acids by anthropogenic activities influence the soil acidity levels. Therefore, elevated levels of aluminium have been found in acidified soil solutions and surface waters causing harmful effects to living organisms.

The present study was aimed at proving the above theoretical hypothesis and existence of a possible relationship between soil salinity and soil acidity on the concentration of mobile aluminium ions in samples obtained during the location surveys. Samples were collected from selected locations in Ratnapura, Rathupaswala, Marawila, Mabima and Muthurajawela to get different soil types and the survey results were used to test the hypothetical relationship between the presence of the stated factors, and the existence of a high concentration of mobile aluminium in the soil water samples. The analysis covered basic parameters such as soil pH, soil cation exchange capacity (CEC), soil organic matter, soil electrical conductivity and the influence of the concentration of mobile aluminium at different pH and Na⁺ concentration levels. The total aluminium concentration in the soil was assayed by digesting samples with strong acid. Concentration of mobile aluminium in soil samples were analyzed using the flame atomic absorption spectrophotometer (analytikjena nova 400p). The results indicated that there is no clear relationship between mobile aluminium and total aluminium in the soil. It was also found that the concentration of mobile aluminium released increased with decrease in soil pH and that the increase was marked when the pH of soil water was less than 4.0. Highest mobile aluminium release to the soil solution was found from Mabima sites [pH 5.00 =0.54 (± 0.06) mg kg⁻¹ of dry soil, pH 0.00= 90.12 (± 7.01) mg kg⁻¹] and least in samples from Marawila sites [pH 5.00 =0.48 (± 0.03)mg kg⁻¹, pH 0.00= 4.52 (± 0.36)mg kg⁻¹]. This result confirmed that there is a relationship between soil acidity and concentration of mobile aluminium in the soil. Results also showed that the concentration of mobile aluminium released increased with increasing soil salinity and that the increase was rapid when the Na⁺ ion concentration was higher than 2.0 %. Highest mobile aluminium release to the soil solution was found from Muthurajawela sites [Na⁺ 1.0% = lower than detection limit, Na⁺ 5.0% = 9.87 (± 0.67) mg kg⁻¹] and least found from Marawila sites [Na⁺1.0% = lower than detection limit, Na⁺ 5.0% = 2.24 (± 0.23) mg kg⁻¹] confirming the relationship between soil salinity and the concentration of mobile aluminium in the soil. The study also points towards the future opportunities for research to confirm these findings using wider samples and employing more vigorous research methodologies.

Key words: Mobile Al, total Al, soil acidity, soil salinity, CEC.

1. Introduction

Aluminium is an element commonly occurring in nature, the third most abundant in the earth's crust after oxygen and silicon. It forms numerous mineral and organic complexes, characterized by different degrees of hydration. Its easy transition from solid to liquid phase and high solubility in acid environment are decisive factors for its important function in the environment¹. Until recently, aluminium was considered harmless for the human organism as it is readily excreted through urine. However, studies of environmental toxicology conducted in recent years indicated that aluminium could be a cause of many diseases in humans, animals and plants. Acid rains and increasing acidification of the environment induced water and soil pH changes which resulted in the mobilization of toxic aluminium ions which in turn evoked many unfavorable alterations, such as plant poisoning and numerous disturbances in the function of human and animal organism.

The maximum Aluminium level in ground water recommended by the World Health Organization is less than 0.1 mg L⁻¹. The concentration of Aluminium in natural waters can vary significantly depending on various physicochemical and mineralogical factors. Dissolved Aluminium concentrations in waters with near-neutral pH values usually range from 0.001 to 0.05 mg/L but rise to 0.5 - 1 mg/L in more acidic waters or water rich in organic matter. At the extreme acidity of water affected by acid mine drainage, dissolved Aluminium concentrations of up to 90 mg/L been measured².The total Aluminium concentration in the human body is approximately 9 mg/kg. In some organs, specifically the spleen, kidneys and lung, concentrations up to 100 mg/kg may be present. Daily Aluminium intake is approximately 5 mg, of which only a small fraction is absorbed. This leads to relatively low acute toxicity. Absorption is about 10 µg per day. These amounts are considered harmless to humans. However, once the element is taken up in the body it is not easily removed. Large Aluminium intake may negatively influence health. This was connected with nerve damage. Particularly people with kidney damage are susceptible to Aluminium toxicity. There is a risk of allergies. Aluminum is probably mutagenic and carcinogenic. However, this is uncertain because Aluminium concentrations always increase with age. Increased Aluminium intake may also cause osteomalacia (vitamin D and calcium deficits). Aluminum intake mainly occurs through food and drinking water. The most recent standards were between 50 and 200 µg/L. Aluminum particles may cause functional lung disorder. Aluminum chloride may corrode the skin; irritate the mucous membranes in the eyes, and cause perspiration, shortness of breath and coughing. Alum increases blood clotting³,

Aluminum toxicity is considered the most important growth-limiting factor for plants in acid soils⁵. In Poland, the most extensive research in this field until now was carried out in the 1990s and covered the determination of different Aluminium fractions in river and lake water^{6, 7} and the soil solution itself⁸. This work is aimed at assessing the quantity and forms of Aluminium occurring in the soils. Among other authors, the effect of organic matter on exchangeable Aluminium concentrations in soil has been emphasized by many researches^{9, 10, 11}. However Aluminium concentrations are still very low in natural waters due to the extreme low solubility of Al-bearing minerals. Concentrations in groundwater

are strongly pH dependent¹². Few studies have been carried out in Sri Lanka to monitor Aluminium releases from soil matrix to water under acidic conditions and Aluminium concentrations in acidic well water but this research only restricted to soils and wells near Ranala, Kaduwela in Sri Lanka. In this research the concentrations of Aluminium was measured using spectrophotometric methods based on the formation of the complex between Aluminium and pyrocatechol violet^{13, 14}.

Al toxicity can be a matter of public health concern but this has not received much research attention in Sri Lanka. Therefore this research was carried out to find the relationship between soil pH and available Al concentration in soil and to find the relationship between soil salinity and available Al concentration in soil that can effect to the public hearth. In the current work, soil from five different areas was analyzed using different parameters and model study was carried out using different pH values and different Sodium concentrations. Atomic absorption spectrophotometer was used to determine the Al concentrations in the soil.

2. Materials and Methods

2.1 Materials and Reagents

Five different soil types, Calcium chloride (97.0%, M.W=147.02 g mol-1), H_2SO_4 acid (96.5% w/v, M.W=96.08 g mol-1), HCI (35% w/v, M.W=36.46 g mol-1, density=1.18 mL-1), NaOH (97%, M.W=40.0 g mol-1), Sodium acetate (99%, M.W= 82.03 g mol-1), Ethanol (95% w/v), Ammonium acetate (97%, M.W=147.02 g mol-1), Sodium chloride (99%, M.W=58.44 gmol-1), Potassium chloride (99% w/v, M.W= 74.55 g mol-1)

2.2 methods

2.2.1 Sample collection

Soil samples were collected from five sites around Ratnapura, Mabima, Rathupaswala, Marawila and Muthurajawela in Sri Lanka. Several sub samples were collected from each site using the random sampling technique. The samples were collected from 30-35 cm depth from the soil surface and stored in clean polythene bags.

2.2.2 Soil characterization

Physical characterization methods were carried out in the laboratory soon after sample collection. All soil and ground water characterization was done using Food and Agriculture Organization (FAO) Procedures.

2.2.2.1 Moisture content

Soil sample (10.00 g) was transferred into a clean, dry crucible and initial weight of the sample and crucible was recorded. The sample was dried in an oven at 105 °C until constant weight was obtained. It was then allowed to cool to room temperature in a desiccator for 30 minutes and the final weight was recorded.

2.2.2.2 Organic matter content

Oven dried soil sample (10.00 g) was measured in to crucible with a lid and the initial weight was recorded. The sample was then placed in a muffle furnace and ashed at 450 °C for 4 hours. The crucible was then removed and allowed to cool to room temperature in a desiccator and the final weight was recorded.

2.2.2.3 Soil pH

Water soluble soil pH

Wet soil sample (20.00 g) was measured into a beaker and deionized water (40.00 mL) was added. The soil suspension was allowed to stand in water for 1 hour and then stirred thoroughly for 10 seconds using a clean glass rod. The soil suspension was allowed to settle for 30 minutes with occasional stirring. The suspension was then centrifuged and the supernatant was taken. The pH of the supernatant was determined using a calibrated pH meter. Above procedure was carried out for air dried soil samples.

Exchangeable soil pH

Wet soil sample (20.00 g) was measured into a beaker and CaCl2 solution (0.01 M, 40.00 mL) was added. The soil suspension was allowed to stand in CaCl2 solution for 1 hour and then stirred thoroughly for 10 seconds using a clean glass rod. The soil suspension was allowed to settle for 30 minutes with occasional stirring. The suspension was then centrifuged and the supernatant was taken. The pH of the supernatant was determined using a calibrated pH meter. Above procedure was carried out for air dried soil samples.

2.2.2.4 Soil Cation exchange capacity

Air dried soil (1.00 g) was transferred into a centrifuge tube and Sodium acetate (1.0 M, 5.00 mL) was added. Then it was shaken for 5 minutes and it was centrifuged until the supernatant is clear. The supernatant was removed completely and the extraction was repeated another three more times.

Then ethanol (95%, 5.00 mL) was added and above procedure was repeated until the electrical conductivity of the decant reads less than 40 mS/cm. Then ammonium acetate (1.0 M, 5.00 mL) was added and above procedure was repeated for three times. Supernatant was collected at each time and it was top up to 20.00 mL using ammonium acetate solution.

A standard series of Sodium ion (0.20, 0.40, 0.70 ppm) was prepared using analytical grade chemicals and deionized water. The solutions were stored in polypropylene bottles at 4 °C ready for FAAS analysis.

2.2.2.5 Aluminium in the soil and model study

Total Aluminium in the soil

Air dried soil (0.50 g) was transferred into a beaker and HCl (conc, 5.00 mL) was added. Then it was stirred and the suspension was gravity filtered. Then the solution was top up to 100 mL using deionized water.

Exchangeable Aluminium in the soil

Air dried soil (10.00 g) was transferred into a dry filter paper in a funnel placed in a volumetric flask (100 mL). Then KCl (1.0 M, 10.00 mL) ten portions were added with 15 minutes time intervals. After that the funnel was removed and the volumetric flak was filled to the mark with KCl.

Model study

Exchangeable Aluminium in the soil at pH=5.00

Air dried soil (10.00 g) was transferred into a dry filter paper in a funnel placed in a volumetric flask (100.0 mL). Then deionized water with pH=5.00 (10.00 mL) ten portions were added with 15 minutes time intervals. After that the funnel was removed and the volumetric flak was filled to the mark with deionized water.

The same procedure was followed for pH=4.00, 3.00, 2.00, 1.00, 0.00 solutions.

Exchangeable Aluminium in the soil at Na= 1.0%

Air dried soil (10.00 g) was transferred into a dry filter paper in a funnel placed in a volumetric flask (100.0 mL). Then deionized water with Na (1.0%, 10.00 mL) ten portions were added with 15 minutes time intervals. After that the funnel was removed and the volumetric flak was filled to the mark with deionized water.

The same procedure was followed for Na=2.0%, 3.0%, 4.0%, 5.0% solutions.

3. Results and Discussion

3.1Soil pH

The soil pH of the soil samples used for this study is represented in **Table 1**. The results (Exchangeable H⁺) indicated that the soil samples were found in, Rathupaswala area has moderately acidic soil, Ratnapura area has strongly acidic soil, Marawila area has slightly acidic soil, Mabima area has slightly acidic and Muthurajawela area has extremely acidic soil.

Soil pH is one of the most influential parameters controlling the conversion of Aluminium from immobile solid phase form to more mobile or bioavailable solution phase forms. Exchangeable Aluminium concentrations in soil are primarily affected by its acidity ^{15, 16, 17, 18}, which was confirmed by research results (**Table 3** and **Fig 1**). An increase in soil acidity brought an exponential increase in mobile Aluminium concentration (**Table 3**).

3.2 Soil organic matter content

The soil organic matter content of the soil samples used for this study is represented in Table 1. The results indicated that the soil samples were found in Muthurajawela area has higher organic matter content than other areas. The increasing concentration of organic matter in the soil brought about an increase in Aluminium content in the organic fraction, the negative effect of acidity was reduced by the sorptive properties of organic matter or the formation of Aluminium ion complexes by organic matter. This experiment was carried using different concentration of organic matter and constant pH, since it was impossible to select soil samples of different pH and different humus content to show the effect of organic matter on Aluminium mobility 19. According to the results obtained in this research study which cannot illustrate the effect of organic matter content on Aluminium mobility because in this study pH values are different in different selected areas.

3.3 Soil Cation Exchange Capacity (CEC)

Soils have a CEC primarily because of the clay particles and organic matter in the soil tends to be negatively charged, which was confirmed by research results (**Table 1**). According to the results obtained, sandy soils low in organic matter has a very low CEC while heavier clay soils or soils high in organic matter generally have a much higher CEC. This usually means that high CEC (clay and organic) soils have a greater water holding capacity than low CEC (sandy)

soils, which was confirmed by experimental results (moisture content on **Table 1**).

3.4 Total Aluminium in soil

Total Aluminium concentration of the soil samples used for this study is represented in **Table 2**. The Aluminium, being one of the basic components of soil, is bound primarily in sparingly soluble aluminosilicates. All total Aluminium in soil are not in mobile phase and there is no strong relationship between total Aluminium concentration and mobile Aluminium concentration, which was confirmed by research results (**Table 2**).

3.5 Exchangeable Aluminium in soil

Exchangeable Aluminium concentration of the soil samples used for this study is represented in **Table 2**. Exchangeable Aluminium concentrations in soil are primarily affected by its acidity, which was confirmed by research results (soil pH on **Table 1** and Exchangeable Aluminium on **Table 2**). Increasing pH decreased the content of exchangeable Aluminium in the soil. The results indicated that the Muthurajawela area soils high in soil acidity have a very high exchangeable Aluminium concentration while Exchangeable Aluminium concentrations in Mabima area is very low (lower than the instrument detection limit).

3.6 Model study 1- Effect of soil acidity on Aluminium mobility in selected soil samples

The exchangeable Aluminium concentration of the soil samples used for this study with different pH values are represented in **Table 3** and **Fig 1**. The results indicated that the concentration of Aluminium released increased with decrease in pH, and the increase is rapid when solution pH was less than 3.00. According to the results obtained, Mabima area soil samples were contained highest mobile Aluminium

content (about 90 mg kg⁻¹) and Marawila area soil samples were contained lowest mobile Aluminium content (about 9 mg kg⁻¹) when decreasing soil pH up to zero. There is a relationship between soil acidity and Aluminium mobility, which was confirmed by research results (**Table 3** and **Fig 1**).

3.7 Model study 2- Effect of soil salinity on Aluminium mobility in selected soil samples

The exchangeable Aluminium concentration of the soil samples used for this study with different Na⁺ concentrations are represented in Table 4 and Fig 2. The results indicated that the concentration of Aluminium released increased with increase in Na⁺ concentration. Cation exchange capacity play a major role for this part because when the cations are present in equivalent amounts, the order of strength of adsorption is $Al^{3+} > Ca^{2+} > Mg^{2+} > K^+ = NH^{4+} > Na^+$. This shows Na⁺ has least affinity with cation exchange sites in soil and Al3+ has the highest affinity with cation exchange sites in soil due to its +3 charge. But the relative concentrations of the cations in soil solution help to change above phenomena, when increasing Na concentration in soil solutions it increases the mobile Aluminium concentration in soil. Soil particles are negatively charged and this Na⁺ ion has a poor ability to neutralize the soil particles so the soil particles repel each other and break the soil structure. This also can increase the mobile Aluminium concentration in the soil. The increase is rapid when Na⁺ concentration was higher than 3.0%. According to the results obtained, Muthurajawela area soil samples were contained highest mobile Aluminium content (about 10 mg kg⁻¹) and Marawila area soil samples were contained lowest mobile Aluminium content (about 2 mg kg⁻¹) when increasing soil salinity up to 5.0%. There is a relationship between soil salinity and Aluminium mobility, which was confirmed by research results (Table 4 and Fig 2).

Table 1 Soil characterization

| Soil type | pH (Soluble) | pH (Exchangeable) | Moisture content (%) | Organic matter content (%)-dry weight | Cation exchange capacity (meq/100 g)- dry weight |
|------------------------------------|--------------|----------------------|-------------------------|---------------------------------------|--------------------------------------------------|
| Rathupaswala (Red- clay loam Soil) | 6.38(±0.41) | 5.47 (±0.80) | 13.31(±1.95) | 5.52(±0.91) | 15.61(±1.12) |
| Rathnapura (Clay loam Soil) | 5.49(±0.40) | 4.77(±0.77) | 14.35(±2.07) | 6.69(±1.45) | 12.13(±1.88) |
| Marawila (Sandy Soil) | 7.57(±0.63) | 6.86(±0.55) | 2.88(±0.59) | 1.37(±0.15) | 8.86(±3.12) |

| Mabima (Clay Soil) | 6.85(±0.73) | 6.49(±0.58) | 23.48(±5.72) | 6.70(±1.85) | 36.45(±9.33) |
|--------------------------------|-------------|-------------|--------------|---------------|--------------|
| Muturajawala (Organic Soil) | 3.82(±0.42) | 3.63(±0.36) | 56.63(±4.18) | 58.42(±11.64) | 28.90(±9.59) |

Table 2 Aluminium in the soil samples

| Soil type | Total Aluminium concentration (mg kg ⁻¹) | Exchangeable Aluminium concentration (mg kg ⁻¹) |
|-----------------------------|------------------------------------------------------|-------------------------------------------------------------|
| Rathupaswala (Red Soil) | 136.60(±0.14) | 0.22 (±0.09) |
| Rathnapura (Clay loam Soil) | 290.95(±14.62) | 0.25 (±0.11) |
| Marawila (Sandy Soil) | 14.64(±5.19) | 0.53 (±0.14) |
| Mabima (Clay Soil) | 308.20(±13.85) | ND |
| Muturajawala (Organic Soil) | 139.80(±10.18) | 34.06 (±3.84) |

Table 3Effect of soil acidity on Aluminium mobility in selected soilsamples

| | | | | | _ | _ |
|---------------|-----------------------|-----------------------|-----------------------|-----------------------|------------------------|-----------------------|
| Soil | pH = 0.00 | pH = 1.00 | pH = 2.00 | pH = 3.00 | pH = 4.00 | pH = 5.00 |
| | Mahila Al | Makila Al | Makila Al | Mahila Al | Mahila Al | Malaila Al |
| | Mobile Al | Mobile Al |
| | concentration | concentration | concentration | concentration | concentration | concentration |
| | (mg kg^{-1}) | (mg kg^{-1}) | (mg kg^{-1}) | (mg kg^{-1}) | (mg kg ⁻¹) | (mg kg^{-1}) |
| Rathupaswala | 24.21(±1.24) | 4.76(±0.28) | 3.32(±0.01) | 1.93(±0.07) | 1.37(±0.02) | $0.66(\pm 0.07)$ |
| | 54.55(±3.33) | 10.31(±2.01) | 3.52(±0.27) | 2.03(±0.12) | 1.41(±0.13) | 1.03(±0.14) |
| Ratnapura | 21.89(±2.09) | 13.71(±0.38) | 3.96 (±0.17) | 1.81 (±0.06) | 1.36(±0.12) | 1.14(±0.12) |
| | 19.05(±2.01) | 8.97(±0.88) | 4.47(±0.36) | 1.93(±0.17) | 0.62(±0.03) | 0.49(±0.03) |
| Marawila | 8.43(±0.52) | 6.63 (±0.22) | 3.55(±0.12) | 2.07(±0.19) | 0.67(±0.05) | 0.35(±0.03) |
| | 4.52(±0.36) | 4.97 (±0.27) | 1.52(±0.16) | 1.79(±0.11) | 0.35(±0.04) | 0.48(±0.03) |
| Mabima | 52.48(±3.27) | 22.57(±0.23) | 2.10(±0.12) | 2.08(±0.19) | 0.83(±0.06) | 0.54(±0.06) |
| 1 | 90.12(±7.01) | 36.61(±2.55) | 2.95(±0.17) | 2.18(±0.20) | 0.14(±0.02) | 0.04(±0.01) |
| Muthurajawela | 62.75(±4.17) | 22.04(±0.10) | 3.01(±0.30) | 2.18(±0.22) | 0.45(±0.05) | 0.32(±0.04) |
| | 25.66(±1.62) | 6.95 (±0.42) | 1.84(±0.16) | 2.14 (±0.11) | 0.34(±0.04) | 0.23(±0.03) |

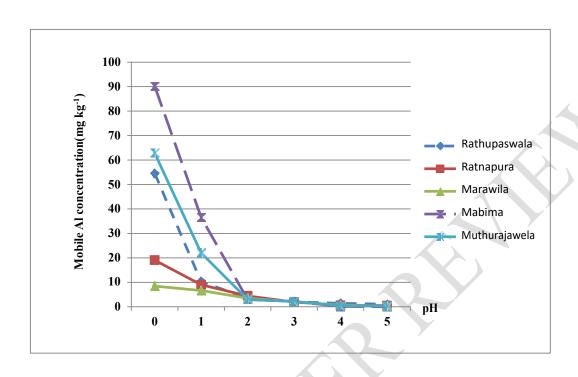


Figure 1Effect of soil acidity on Aluminium mobility in selected soil samples

Table 4Effect of soil salinity on Aluminium mobility in selected soil samples

| Soiltype | Na =1.0% | Na =2.0% | Na =3.0% | Na =4.0% | Na =5.0% |
|---------------|------------------------------------------------------|------------------------------------------------------|------------------------------------------------------|------------------------------------------------------|------------------------------------------------------|
| | Mobile Al concentration (mg kg ⁻¹) |
| Rathupaswala | ND | 0.37(±0.04) | 1.37(±0.04) | 1.40 (±0.05) | 2.46(±0.13) |
| | ND | ND | 0.95(±0.07) | 1.88(±0.19) | 3.00 (±0.22) |
| Ratnapura | 0.01 (±0.00) | 0.45(±0.04) | 1.46 (±0.05) | 3.45(±0.13) | 3.76(±0.15) |
| | ND | 0.92(±0.10) | 1.66(±0.12) | 4.89(±0.36) | 4.95 (±0.27) |
| Marawila | ND | 0.16(±0.02) | 1.51(±0.14) | 1.76(±0.18) | 2.24(±0.23) |
| | ND | ND | 1.71(±0.16) | 1.30(±0.11) | 2.35(±0.21) |
| Mabima | ND | 1.54(±0.16) | 2.83 (±0.16) | 4.46(±0.22) | 5.35 (±0.32) |
| | ND | ND | 1.08(±0.12) | 2.85(±0.19) | 3.09(±0.02) |
| Muthurajawela | ND | ND | 1.05(±0.11) | 3.36(±0.22) | 6.87 (±0.47) |
| | 0.53(±0.06) | 2.35 (±0.14) | 9.23 (±0.73) | 9.75(±0.58) | 9.87 (±0.67) |

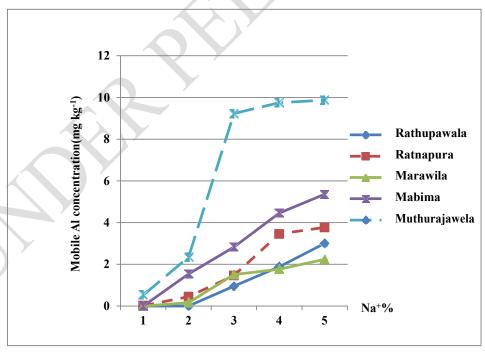


Figure 2 Effect of soil salinity on Aluminium mobility in selected soil samples

4. Conclusion

Soil and ground water from Rathupaswala, Ratnapura, Marawila, Mabima and Muthurajawela sites were used for this research and soil texture of five sites were different. Highest total Aluminium was found from Mabima sites and least found from Marawila sites. Highest exchangeable Aluminium was found from Muthurajawela sites and least found from Mabima sites. This experiment results reveal that there is no relationship between total Aluminium concentration in soil and exchangeable Aluminium concentration in soil. The mobile Aluminium concentration in soil increased with the increasing soil acidity. With a gradually increasing amount of added

H⁺ to the soil, much higher increases of mobile Aluminium to the soil solution. The increase is rapid when the pH of the solution was less than 3.00. Highest mobile Aluminium release to the soil solution was found from Mabima sites and least found from Marawila sites. Results of this study showed that there is a strong relationship between soil acidity and Aluminium mobility.

The mobile Aluminium concentration in soil increased with the increasing soil salinity. With a gradually increasing amount of added Na to the soil, much higher increases of mobile Aluminium to the soil solution. The increase is rapid when the Na⁺ of the solution was more than 3%. Highest mobile Aluminium release to the soil solution was found from Muthurajawela sites and least found from Marawila sites. Results of this study showed that there is a strong relationship between soil salinity and Aluminiummobility. Mobile Aluminium concentration in ground water samples of all sites were very low values. Aluminium affinity to cation exchange capacity is very high due to this factor Aluminium ions are strongly bind with soil and usually not seen in the ground water. Release of the Aluminium from soil due to soil acidity or salinity can affect to the mobile Aluminium in ground water and this can cause health problems to humans, plants andanimals.

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