Alleviating aluminum toxicity in an acid sulfate soil from Peninsular Malaysia by calcium silicate application

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Abstract

In response to human population increase, the utilization of acid sulfate soils for rice cultivation is one option for increasing production. The main problems associated with such soils are their low pH values and their associated high content of exchangeable Al, which could be detrimental to crop growth. The application of soil amendments is one approach for mitigating this problem, and calcium silicate is an alternative soil amendment that could be used. Therefore, the main objective of this study was to ameliorate soil acidity in rice-cropped soil. The secondary objective was to study the effects of calcium silicate amendment on soil acidity, exchangeable Al, exchangeable Ca, and Si content. The soil was treated with 0, 1, 2, and 3 Mg ha^{-1} of calcium silicate under submerged conditions and the soil treatments were sampled every 30 days throughout an incubation period of 120 days. Application of calcium silicate induced a positive effect on soil pH and exchangeable Al; soil pH increased from 2.9 (initial) to 3.5, while exchangeable Al was reduced from 4.26 (initial) to 0.82 cmol$_c$ kg^{-1}. Furthermore, the exchangeable Ca and Si contents increased from 1.68 cmol$_c$ kg^{-1} (initial) to 4.94 cmol$_c$ kg^{-1} and from 21.21 mg kg^{-1} (initial) to 81.71 mg kg^{-1}, respectively. Therefore, it was noted that calcium silicate was effective at alleviating Al toxicity in acid sulfate, rice-cropped soil, yielding values below the critical level of 2 cmol$_c$ kg^{-1}. In addition, application of calcium silicate showed an ameliorative effect as it increased soil pH and supplied substantial amounts of Ca and Si.

1 Introduction

Acid sulfate soils are widespread in Malaysia, occurring almost exclusively along its coastal plain (Shamshuddin and Auxtero, 1991; Shamshuddin et al., 1995; Muhrizal et al., 2006; Enio et al., 2011). In these areas, the alluvial sediments are intermittently inundated by seawater during low and high tides. These soils are dominated by pyrite with high acidity (soil pH < 3.5) (Shamshuddin, 2006) and are produced when the pyrite-laden soils in the coastal plains are opened up for crop production and/or development. This scenario leads to the release of large amounts of Al into the soil environment (Shamshuddin et al., 2004), which affects crop growth. For example, it affects oil palm growth (Auxtero and Shamshuddin, 1991) and cocoa production (Shamshuddin et al., 2004). In Peninsular Malaysia, acid sulfate soils are used for rice cultivation with mixed success. At times, rice cultivation in these soils is successful; but most often, the rice yield each season is very low (< 2 t ha^{-1}). Amelioration practices with ground magnesium lime (GML) and/or basalt have shown improvements of up to 3.5 t ha^{-1} in rice yield (average per season).

The application of soil amendments to acid sulfate soil is a common approach for improving fertility. Suswanto et al. (2007), Shazana et al. (2013), Elisa et al. (2014), and Rosilawati et al. (2014) reported that the infertility of acid sulfate soils can be ameliorated by application of lime, basalt, organic fertilizer, and/or their combination at an appropriate rate. Application of these ameliorants increased soil pH and reduced Al toxicity, resulting in improved rice growth. In addition to these improvements, these ameliorants also supply calcium (Ca) and magnesium (Mg), which are needed for crop growth and development.

Besides Ca and Mg, silicon (Si) is also important for rice growth. It has a positive effect on the growth of crops such as tomato (Peaslee and Frink, 1969), barley and soybean (Hodson and Evans, 1995), and many others (Liang et al., 2007). The application of Si may reduce the severity of fungal diseases such as blast and sheath blight of rice

(Farnaz Abed-Ashtiani et al., 2012); powdery mildew of barley, wheat, cucumber, muskmelon, and grape leaves; and vermin damage of rice by plant hopper (Crock and Prentice, 2012; Ma et al., 2001; Menzies et al., 1992; Bowen et al., 1992; Datnoff et al., 2001). In addition, Si can effectively reduce Al toxicity (Barcelo et al., 1993). Calcium silicate application could be a source of Si for soils. This material is available in Peninsular Malaysia. Therefore, this study is relevant because calcium silicate could be used to alleviate Al toxicity of soil from the Merbok granary area located in the northern state of Kedah, Peninsular Malaysia. Certain regions of the rice cultivation area are classified as acid sulfate soils and the average rice yield in these areas is less than 2 t ha^{-1} $season^{-1}$. This is due to high soil acidity, Al toxicity, and/or rice blast disease (*M. grisea*). Therefore, the main objective of this study was to ameliorate soil acidity in the rice-cropped soils of this area. The secondary objective was to study the effects of calcium silicate amendment on soil acidity, exchangeable Al, exchangeable Ca, and Si content.

2 Materials and methods

2.1 Soil type, treatments, and experimental design

The experiment was conducted at the Field 2 Glasshouse at Universiti Putra Malaysia, Serdang, Malaysia. The soil used in this study was obtained from Merbok, Kedah, Peninsular Malaysia. The soil sampling site was a rice-cropped area and the sampling was performed 1 month prior to rice cultivation (dry conditions). A composite soil sample of approximately 2500 g was taken from 0–15 cm depth using an auger. The sample was taken within a 0.5 ha region of the rice-cropped area. Afterward, the soil was crushed, passed through a 2 mm sieve, and mixed thoroughly prior to incubation.

Five hundred grams of soil was used to fill a plastic pot, which was then incubated for 120 days. The treatments included 0 (CS0), 1 (CS1), 2 (CS2), and 3 (CS3) Mg ha^{-1} of calcium silicate, with three replications. These were arranged in a completely randomized design (CRD). The total number of samples was 48 (4 treatments × 3 replications × 4 sampling times). Twelve pots were sampled every 30 days throughout the incubation period, i.e., the sampling times were at 30 days (30D), 60 days (60D), 90 days (90D), and 120 days (120D) of incubation and corresponded to the vegetative, reproductive, flowering, and maturity phases of rice growth, respectively. The calcium silicate ($CaSiO_3$) used in this experiment was obtained from Kaolin (Malaysia) Sdn. Bhd., Malaysia. This calcium silicate had the following composition: SiO_2= 40–55%, calcium (as CaO) = 40–50%, Al_2O_3 = below 1.5%, MgO = below 3%, iron (as Fe_2O_3) = below 1%, and pH = 8.54.

The soils were mixed thoroughly with the added calcium silicate prior to the addition of water. Tap water was added regularly and the water levels were maintained at approximately 5 cm (height) above the soil surface. The composition of the tap water in relation to phosphorus (P), potassium (K), aluminum (Al), calcium (Ca), iron (Fe), magnesium (Mg), and silicon (Si) was 0.74, 10.62, 0.14, 19.78, 0.03, 1.00, and 5.18 mg L^{-1}, respectively. The pH of the tap water used was 7.37.

2.2 Soil analyses

Soil samples were air-dried, ground, and passed through a 2 mm sieve prior to chemical analyses. Soil pH was determined in water at a ratio of 1:2.5 (soil/distilled water) using a glass electrode pH meter. Total C, N, and S were determined using a Leco CNS analyzer. Cation exchange capacity (CEC) was determined using 1M NH_4OA_c at pH 7

(Chapman, 1965). Exchangeable Ca, Mg, K, and Na were determined using 1N NH_4Cl (Ross and Ketterings, 1995; Shamshuddin, 2006). To achieve this, 2 g of air-dried soil was placed in a 50 mL centrifuge tube and 20 mL 1N NH_4Cl was added. The sample was shaken for 2 h on an end-to-end shaker at 150 rpm, followed by centrifugation at 2500 rpm for 15 min. The extract was passed through a filter paper into a 50 mL plastic vial. The exchangeable Ca, Mg, K, and Na in the extract were determined by inductively coupled plasma-optical emission spectroscopy (ICP-OES). Exchangeable Al was determined by extracting 5 g of soil with 50 mL of 1M KCl. The mixture was shaken for 30 min and the extracted Al was analyzed by ICP-OES. Extractable Fe, Cu, Zn, and Mn were extracted using extracting agent (0.05N HCl and 0.025N H_2SO_4). To achieve this, 5 g of air-dried soil was shaken with 25 mL of extracting agent for 15 min. The extract was passed through a filter paper and used to determine Fe, Cu, Zn, and Mn by atomic absorption spectrometry (AAS). Additionally, 0.01M $CaCl_2$ was used to extract plant-available Si from the soil. For this, 2 g of soil was shaken for 16 h with 20 mL $CaCl_2$ extractant in a 50 mL centrifuge tube on an end-to-end shaker. The sample was centrifuged at 2000 rpm for 10 min before the supernatant was filtered and analyzed for Si (Datnoff et al., 2001) using ICP-OES.

2.3 Statistical analysis

Statistical analysis for means comparison was performed using Tukey’s test in SAS version 9.2 (SAS, Institute, Inc., Cary, NC).

3 Results

3.1 Initial soil chemical characteristics

Initial soil pH and exchangeable Al were 2.90 and 4.26 cmol$_c$ kg^{-1}, respectively. Exchangeable Ca, Mg, K, and Na were 1.68, 2.61, 0.55, and 2.61 cmol$_c$ kg^{-1}, respectively. Total C, N, and S were 3%, 0.2%, and 0.13%, respectively. At the site where the soil was sampled, rice is normally grown twice per year and the straw is often left to rot on the paddy field. The decomposition of the rice straw, to some extent, contributed to the increased C content and CEC of the soil. In this study, the CEC of the soil was 18.12 cmol$_c$ kg^{-1}. The values for extractable Fe, Cu, Zn, Mn, and Si prior to incubation were 1118.6, 0.23, 0.96, 1.60, and 21.21 mg kg^{-1}, respectively.

3.2 Effect of calcium silicate on soil pH

Figure 1 shows the effect of calcium silicate application on soil pH under the submerged conditions. It shows that soil pH increased in line with the incremental increases in the calcium silicate application rate. The highest soil pH increase was from 2.90 (initial) to 3.95 due to the application of 3 t ha^{-1} calcium silicate. After 30 days of incubation (D30), soil pH of CS2 was significantly higher than that of CS1, with values of 3.77 and 3.62, respectively. Treatment CS3 was significantly higher in terms of soil pH compared with CS0, CS1, and CS2 at D60; CS0 and CS1 at D90; and CS0, CS1, and CS2 at D120, showing values of 3.90, 3.84, and 3.95, respectively.

3.3 Effect of calcium silicate on exchangeable Al

Figure 2 shows the effect of calcium silicate application on exchangeable Al. It shows that as the calcium silicate rate increased, the exchangeable Al decreased from 4.26 to 0.82 cmolc kg^{-1}. This is a 74% decrease in exchangeable Al in the acid sulfate soil due to the application of calcium silicate. At 30 and 120 days of incubation, exchangeable Al content in the soil treated with 2 and 3 t ha^{-1} of calcium silicate had significantly decreased compared to that in the untreated soil. However, there was no significant effect of calcium silicate on exchangeable Al after 60 and 90 days of incubation.

3.4 Effect of calcium silicate on exchangeable calcium

Figure 3 show that the application of calcium silicate increased exchangeable Ca. There was a significant effect among the treatments after 30 days of incubation. At 60, 90, and 120 days of incubation, soil treated with 2 and 3 t ha^{-1} of calcium silicate had significantly increased soil-exchangeable Ca compared with both untreated soil and soil treated with 1 t ha^{-1} of calcium silicate.

3.5 Effect of calcium silicate on silicon content

Application of calcium silicate increased the Si content of the soil, as shown in Figure 4, from 14% to 74%. At 30 days of incubation, soil treated with 2 and 3 t ha^{-1} of calcium silicate had a significantly increased Si content compared with both untreated soil and soil treated with 1 t ha^{-1} of calcium silicate. At 60 days of incubation, the Si content increased significantly for soil treated with 2 and 3 t ha^{-1} of calcium silicate compared with the soil treated with 1 t ha^{-1} of calcium silicate. The Si content of the soil continued to increase at 90 days of incubation; in the soil treated with 1 t ha^{-1} calcium silicate, it was significantly increased compared to the 2 t ha^{-1} treatment. However, no significant effect was observed among the treatments after 120 days of incubation.

4 Discussion

From this study, it was found that calcium silicate can neutralize H^+ ions in soil, as noted by the pH increase in acid sulfate soils upon calcium silicate application (Figure 1). Similar findings have been found by Smyth and Sanchez (1980) and Fiantis et al. (2002). These authors attributed their results to the OH^- released from colloidal surfaces during the adsorption of the silicate ions. Due to the application of calcium silicate, soil pH increased significantly from 2.90 (initial) to 3.41–3.95.

During the incubation period, there was a strong relationship between calcium silicate and soil pH at D30 ($R^2 = 0.77$), D60 ($R^2 = 0.77$), D90 ($R^2 = 0.84$), and D120 ($R^2 = 0.92$). The increasing correlation coefficient over time was related to the increasing capacity of the soil to adsorb silicate anions.

It was observed that the soil pH was slightly lower for CS0, CS1, and CS2 at D60 and D90 compared to that at D30 and D120. The decrease in soil pH is believed to be due to the release of protons as pyrite in the soil was oxidized during the incubation period. Shamshuddin et al. (2004) reported that after 12 weeks of incubation, soil pH in the Cg horizon of acid sulfate soil was lowered by 1 unit. The results from the current study are consistent with those from other studies on acid sulfate soils (Shamshuddin and Auxtero, 1991; Shamshuddin et al., 1995; Shamshuddin et al., 2014). The oxidation of pyrite, which produces acidity, may have taken place according to the following reactions outlined by van Breemen (1976):

$$2FeS_{2(s)} + 7O_{2\,(aq)} + 2H_2O \rightarrow Fe^{2+}{}_{(aq)} + 4SO_4^{2-} + 4H^+{}_{(aq)}$$

Further oxidation of Fe^{2+} to Fe^{3+} oxide could also promote acidity:

$$2Fe^{2+}{}_{(aq)} + 1/2O_{2\,(aq,g)} + H_2O \rightarrow Fe_2O_{3(s)} + 4H^+{}_{(aq)}$$

As the soil pH increased due to the application of calcium silicate, exchangeable Al decreased to below the critical level for rice growth (2 $cmol_c\ kg^{-1}$). This is consistent with the findings of Hiradate et al. (2007). Figure 2 shows the effect of the treatments on exchangeable Al. It shows that exchangeable Al decreased significantly among the treatments after 30D and 120D. After 30 days of incubation, the exchangeable Al contents of treatments CS2 and CS3 were significantly reduced compared to CS0, which was near the critical level of 2 $cmol_c$ kg^{-1}. It is also shown that exchangeable Al decreased further as the incubation period was further extended. Figure 5 shows the relationship between exchangeable Al and soil pH, where the lines for D60, D90, and D120 are below the line for D30. This implies that a prolonged incubation period would further reduce the exchangeable Al content. The decrease in Al could also be due to the precipitation of Al in the form of inert Al-hydroxides. The exchangeable Al content was reduced to below the critical level of 2 $cmol_c$ kg^{-1} at D90 and D120.

The reduction in exchangeable Al is explained as follows. It is possible that soil Al can be reduced by the reactions of Si-rich compounds. By such reactions, Datnoff et al. (2001) postulated five mechanisms of Al reduction: 1) monosilicic acids increase soil pH (Lindsay, 1979); 2) monosilicic acids are adsorbed on Al hydroxides, reducing their mobility (Panov et al., 1982); 3) soluble monosilicic acid forms slightly soluble substances with Al ions (Lumsdon and Farmer, 1995); 4) mobile Al is strongly adsorbed on silica surfaces (Schulthess and Tokunaga, 1996); and 5) mobile silicon compounds increase plant tolerance to Al (Rahman et al., 1998). All of these mechanisms may work simultaneously, with one perhaps prevailing under certain soil conditions (Dantoff et al., 2001).

The silicate anion can also neutralize H^+ in the soil solution. As the silicate anion captures H^+ ions, it forms monosilicic acid (H_4SiO_4), as shown in the equation below:

$$2CaSiO_3 + 4H^+ + 2H_2O \rightarrow Ca^{2+} + 2H_4SiO_4 \text{ (monosilicic acid)}$$

Monosilicic acid could complex with Al^{3+} in the soil solution to form non-toxic alumino-silicate and hydroxyl-alumino-silicate compounds, which precipitate in the root zone. This reaction would reduce Al toxicity in rice grown on acid sulfate soils treated with calcium silicate (Miranda, 2012).

Furthermore, the application of calcium silicate to the acid sulfate soil showed an immediate ameliorative effect, i.e., the Ca content increased from 1.68 (initial) to above the critical level of 2 $cmol_c\ kg^{-1}$ (Palhares de Melo et al., 2001) at D30. Increasing the rate of calcium silicate increased the Ca content of the soil significantly (Figure 3). For treatment CS3, exchangeable Ca increased significantly compared to CS0 and CS1 throughout the incubation period, with increases of 42.48%, 47.78%, 60.65%, and 38.66% after 30D, 60D 90D and 120D, respectively. However, no significant difference was observed between treatments CS2 and CS3 at D90 and D120.

In the current study, the Si content prior to the incubation was 21.21 mg kg^{-1}; the critical soil Si concentration for crop production is 40 mg kg^{-1} (Dobermann & Fairhurst, 2000). Figure 4 shows the effect of calcium silicate application on Si content. At D30, the Si content in treatments CS2 and CS3 was significantly higher than in treatments CS0 and CS1. At D60, treatment CS3 increased the Si content significantly compared to that of CS0 and CS1, with a value of 40.81 mg kg^{-1} Si. In all treatments at D90 and D120, the Si content of the soil surpassed the deficiency level. At D90, the Si content of treatment CS1 was significantly higher than that of CS2, with a value of 83.53 mg kg^{-1}. The Si content of the soil was affected by the length of incubation, i.e., the Si content of all treatments further increased at 120 days of incubation.

When the soil pH increased, the Si content of the soil also increased (Figure 6). The Si content was positively correlated with soil pH at D30 and D60, likely due to the dissolution of calcium silicate. The ability of the soil to adsorb Si was higher at D30 and D60 than at D90 and D120. There was no correlation observed at D90 and D120, even though the Si content was higher, probably because the soil-exchangeable sites became fully occupied with Si through adsorption processes. This proves that the application of calcium silicate to soil, accompanied by an increase in soil pH, enhances the ability of soil to adsorb Si.

The positive effect of the presence of Si at D30 and D60 corresponds with the early growth stage of rice, i.e., the active tillering stage. This means that a rice plant can actively uptake Si during the tillering stage, hence improving rice growth. Figure 7 shows the relationship between the exchangeable Al and Si contents of the soil throughout the incubation period after the application of calcium silicate. The reduction in exchangeable Al corresponded directly with the availability of Si in the soil. This means that as more Si is available in acid sulfate soil, a reduction in the exchangeable Al content occurs. Exchangeable Al was negatively correlated with Si content in the soil at D30 (R = 0.77) and D60 (R = 0.92), whereas no correlation was observed at D90 and D120. In Figure 7, the D60 line is below the D30 line, indicating that as the incubation period increased, the Al and Si contents showed an antagonistic pattern: Al decreased, while Si increased. This phenomenon indicates that when the Al content of the soil is low, its toxicity may not be the dominant factor inhibiting rice plant growth. On the other hand, Si becomes more readily available for plant uptake. Therefore, the optimal time to plant rice is 30 days after applying calcium silicate because the exchangeable Al is almost reduced to below the critical value of 2 $\text{cmol}_c\ \text{kg}^{-1}$. Because the Si content increased with incubation time, the rice plant could actively uptake Si for growth during active tillering.

Silicon is released from calcium silicate into the bulk soil solution and may become absorbed by plants as $Si(OH)_4$. It may thus be involved in the diverse structural and dynamic aspects of plant life and crop performance. Although not considered an essential element for plant growth and development, Si is considered a beneficial element and is reported as being very useful to plants when they are under abiotic or biotic stress (Datnoff et al., 2001). An alleviating effect of Si on Al toxicity has been reported in many crops including soybean (Baylis et al., 1994), teosinte (Barcelo et al., 1993), sorghum (Hodson and Sangster, 1993), wheat, maize, cotton, and rice (Cocker et al., 1998).

A prolonged incubation of soil not treated with calcium silicate might have also influenced the changes in soil chemical characteristics. As such, CS0 (untreated soil) showed an increase in soil pH from 2.90 (prior to incubation) to 3.63 at D30. A decrease in soil pH values was noted for D60 and D90, likely due to pyrite oxidation in the soil system, and no significant effect was observed among the days of incubation. Meanwhile, exchangeable Al decreased significantly with increasing incubation time. For the first 2 months, exchangeable Al was above the critical level of 2 $cmol_c\ kg^{-1}$ and no significant difference between D30 and D60 was observed. Exchangeable Al was significantly reduced to 1.89 $cmol_c\ kg^{-1}$ at D90, but no significant effect was observed thereafter, i.e., at D120. Application of calcium silicate significantly increased the Si content of the soil. However, no significant effect on Si content was observed between D30 and D60 or between D90 and D120. The Si content of the soil increased significantly, to 59.81 mg kg^{-1}, after 90 days of incubation. The significant increase in Si was due to the hydrolysis of silicate minerals present in the acid sulfate soils. For instance, the hydrolysis of silicate is generalized in the following reaction:

$$\text{Silicate} + H_2O + H_2CO_3 \rightarrow \text{base cation} + HCO^-_3 + H_4SiO_4 + \text{accessory mineral}$$

In this reaction, the base cation would commonly be Mg^{2+} or Ca^{2+}, H_2CO_3 is a proton source, HCO^-_3 is bicarbonate, H_4SiO_4 is silicic acid, and gibbsite [Al $(OH)_3$] is a representative accessory mineral (Essington, 2005).

Farmers in the study area have applied GML to overcome soil fertility problems associated with Al toxicity. As an alternative to GML application, this study suggests that such farmers could benefit from the use calcium silicate as a soil amendment. Therefore, the costs of the input (calcium silicate) and labor should be taken into account to better understand the feasibility of such an approach for farmers in this region. Table 1 shows the costs of applying calcium silicate to 1 ha area for rice production. The costs for calcium silicate and labor were USD 407 t^{-1} and USD 45 t^{-1}, respectively. The total cost (calcium silicate and labor) ranged from USD 452 to USD 1354 ha^{-1}.

5 Conclusions

Application of calcium silicate showed an ameliorative effect on acid sulfate soil, i.e., an increase in soil pH, exchangeable Ca content, and Si content, and a reduction in exchangeable Al. This suggests that calcium silicate amendment is effective in alleviating Al toxicity in acid sulfate, rice-cropped soils. Furthermore, it is an affordable soil amendment, with a cost ranging from USD 452 to USD 1354 ha^{-1}.

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Table 1. Cost of calcium silicate applied to a 1 ha area for rice production

Rate (t/ha)	0	1	2	3
Price (calcium silicate)	0	USD 407 t^{-1} = USD 407	USD 407 t^{-1} = USD 813	USD 407 t^{-1} = USD 1219
Labor	0	USD 45 t^{-1} = USD 45	USD 45 t^{-1} = USD 90	USD 45 t^{-1} = USD 135
Total	**0**	**USD 452**	**USD 903**	**USD 1354**

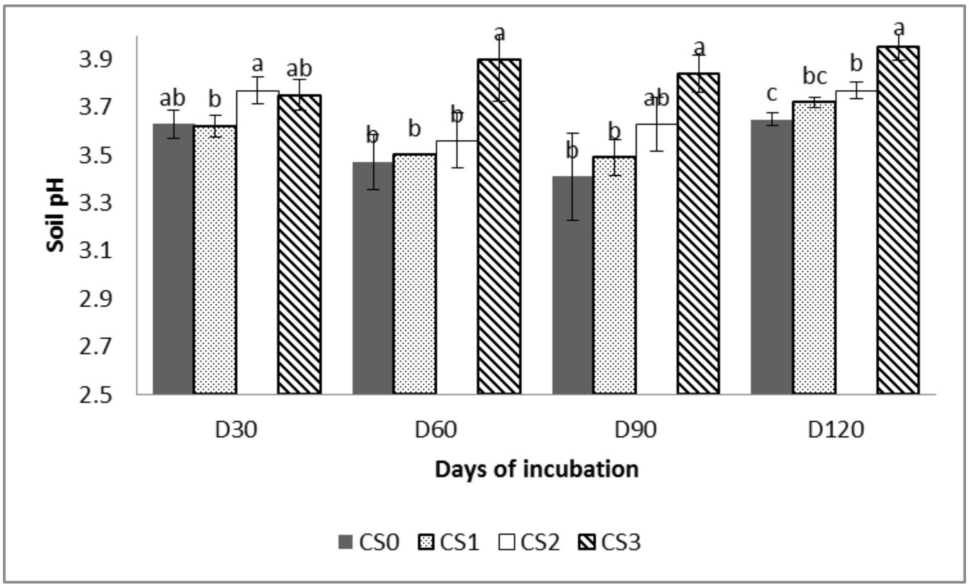

Figure 1. Effects of calcium silicate application on soil pH under submerged conditions. Means marked with the same letter for each incubation day are not significantly different at $p < 0.05$ (Tukey`s Test).

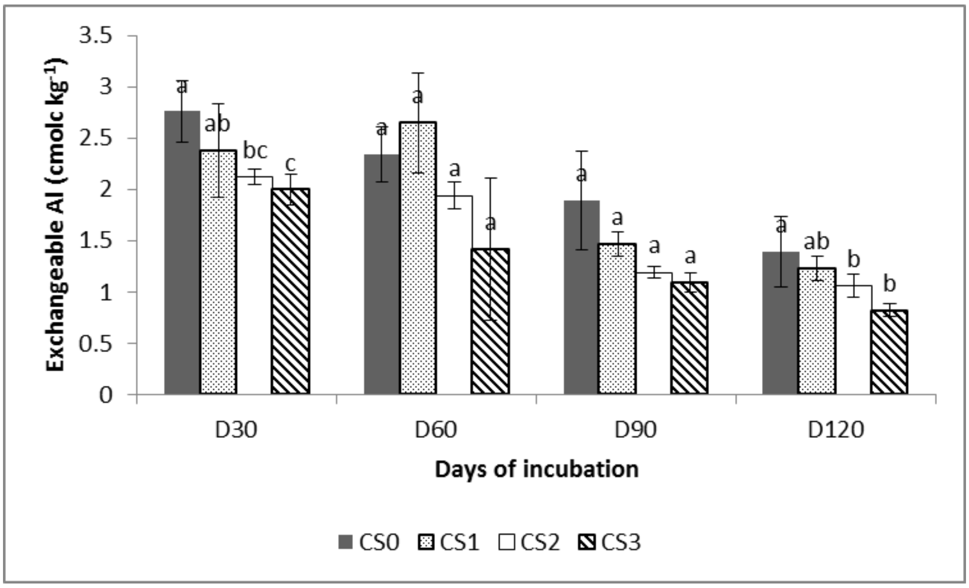

Figure 2. Effects of calcium silicate application on exchangeable aluminum. Means marked with the same letter for each incubation day are not significantly different at $p <$ 0.05 (Tukey`s Test).

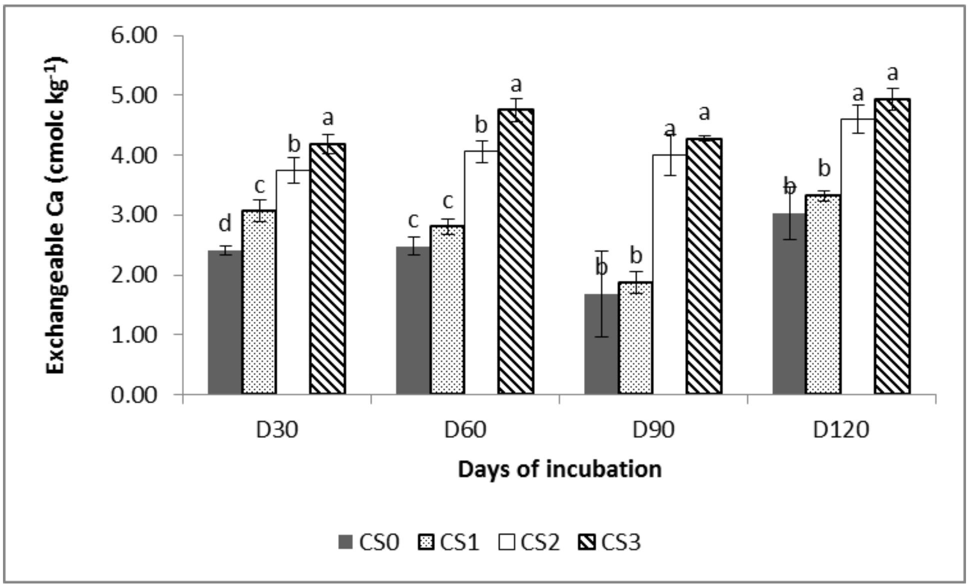

Figure 3. Effects of calcium silicate application on exchangeable calcium. Means marked with the same letter for each incubation day are not significantly different at $p < 0.05$ (Tukey`s Test).

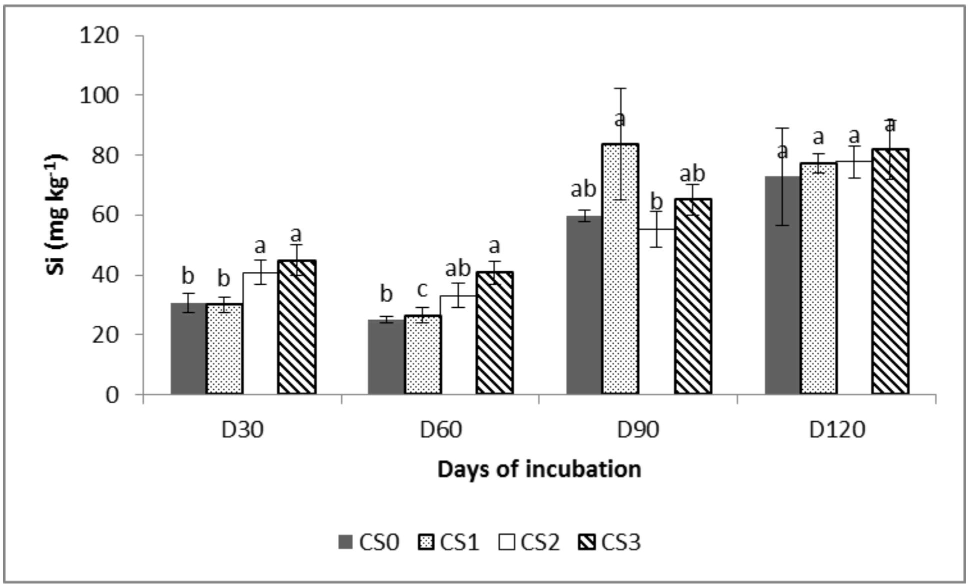

Figure 4. Effects of calcium silicate application on silicon content. Means marked with the same letter for each incubation day are not significantly different at $p < 0.05$ (Tukey`s Test).

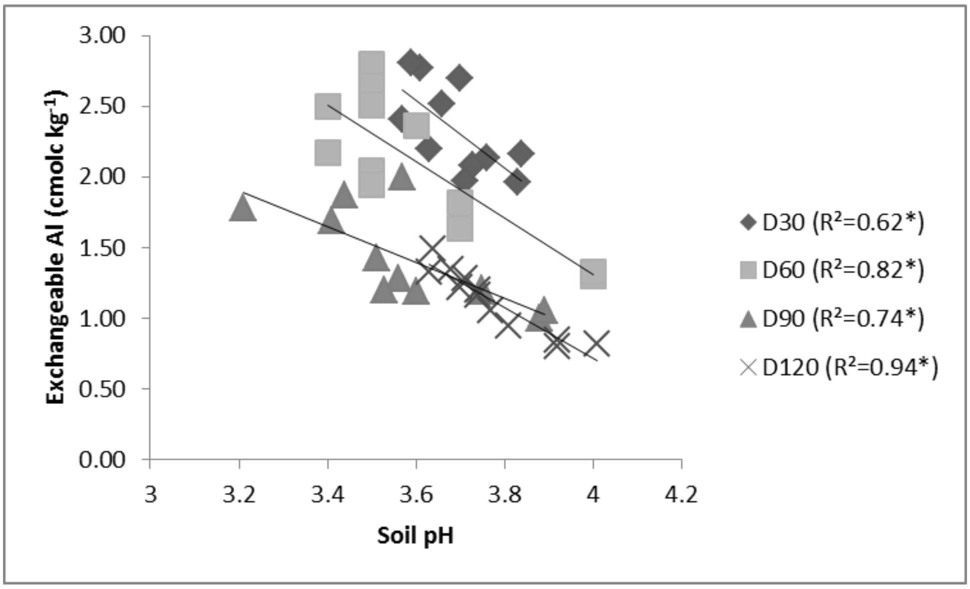

Figure 5. Relationship between exchangeable Al and soil pH (* $p < 0.05$)

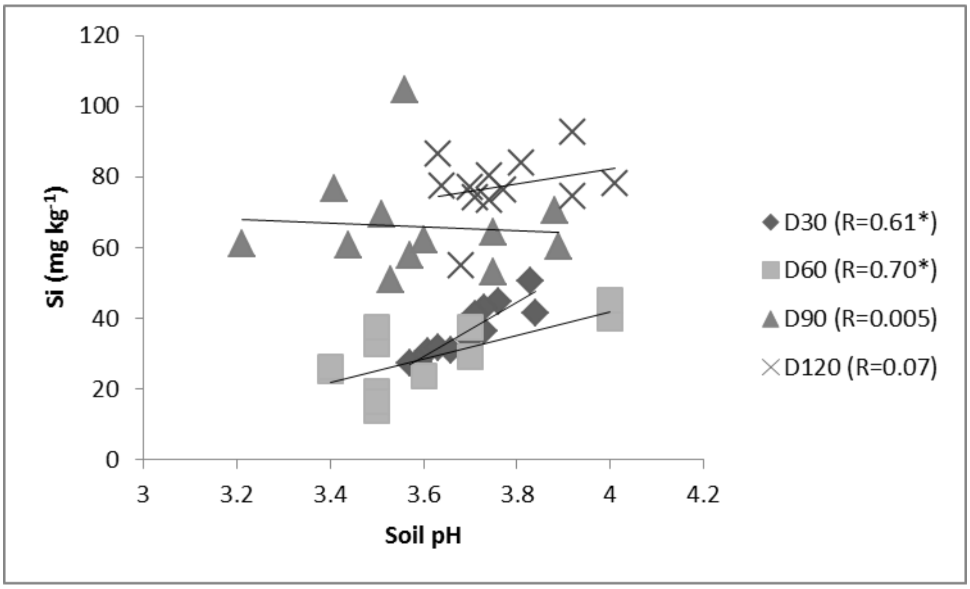

Figure 6. Relationship between Si content and soil pH throughout the incubation period (* $p < 0.05$)

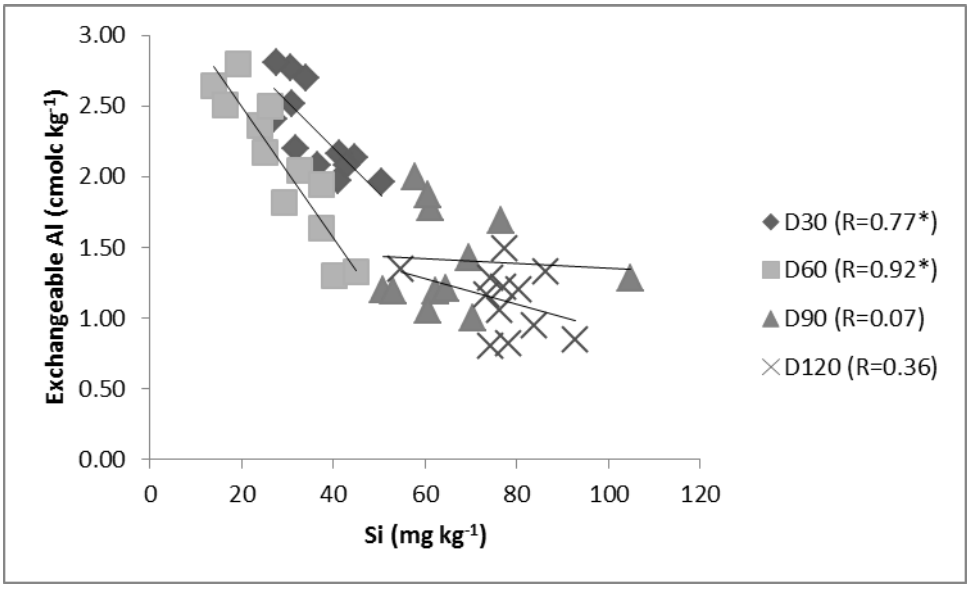

Figure 7. Relationship between exchangeable Al and Si content in the soil throughout the incubation period (* $p < 0.05$)