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THE CONTROLLING FACTORS OF SILICON SOLUBILITY IN SOIL SOLUTION

FAKTOR PENGENDALI KELARUTAN SILIKA DALAM LARUTAN TANAH

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ABSTRAK

Silika adalah unsur yang menguntungkan bagi tanaman padi, yang dapat mencegah penyakit blast, meningkatkan kekuatan batang, dan mengurangi abiotik stress. Silika (Si) dalam larutan tanah dipengaruhi oleh beberapa faktor, seperti pH, temperatur, bahan organik dan potensial redoks (Eh). Tujuan penelitian adalah untuk mengetahui faktor pengontrol kelarutan Si dalam larutan tanah. Penelitian ini menggunakan silika gel dan tanah Ultisols yang berasal dari Jepang. Pada Penelitian ini diamati pengaruh Ca (kalsium), Mg (magnesium) dan unsur lainnya terhadap kelarutan Si dilakukan di dalam laboratorium. Sepuluh gram sampel tanah dalam tabung plastik ditambah silika gel, Ca dan Mg, selanjutnya ditambah air hingga tergenang dan diinkubasi pada suhu 30^{o}C selama 29 hari. Kalsium dan Mg diaplikasikan pada tanah dengan konsentrasi 5, 10, 15 mg Ca L^{-1} (T2, T3, T4) dan 5, 10, 15 mg Mg L^{-1} (perlakuan T5, T6, T7). Sebagai kontrol adalah T0 (tanah) dan T1 (tanah + silika gel). Selama inkubasi, konsentrasi Si, Ca, Mg, Fe, dan Mn dalam air permukaan diukur menggunakan ICP spektroskopi pada hari ke 8, 15, dan 29. Hasil analisis tanah awal dikategorikan sedikit asam (pH 5,7) dan konsentrasi Si adalah 267,1 mg SiO_2 kg^{-1}. Konsentrasi tersebut termasuk dibawah batas kritis Si tersedia (300 mg SiO_2 kg^{-1}). Hasil penelitian menunjukkan bahwa total konsentrasi Ca dan Mg dalam larutan tanah lebih tinggi untuk perlakuan T4 dan T7 dibandingkan perlakuan lainnya. Pada 8 hari pertama inkubasi, pelepasan Si ke dalam larutan tanah lebih tinggi untuk T1 dan T2 dibandingkan dengan perlakuan lain. Kelarutan Si berkorelasi positif dengan Mn, Eh dan berkorelasi negatif dengan pH. Hal ini mengindikasikan bahwa faktor tersebut adalah yang mengontrol pelepasan Si dalam larutan tanah. Pada penelitian ini tidak ditemukan korelasi antara Si dan konsentrasi Ca atau Mg dalam larutan tanah.

Kata kunci: silika, kalsium, magnesium, larutan tanah

ABSTRACT

Silica is a beneficial element for rice plants which can protect from blast disease, increase stem strength, and alleviate abiotic stress. Silicon in soil solution is affected by several factors such as pH, temperature, organic matter, and redox potential (Eh). This study aims to investigate the controlling factor of Si solubility in soil solution. In the present study, Japanese silica gel (JSG) and Ultisols were collected from Japan. In laboratory experiment, the effects of Ca (calcium), Mg (magnesium) and others on solubility of Si (silica) were investigated. Under submerged condition, ten gram of soil with silica gel, Ca and Mg in plastic tube were incubated at 30^0C for 29 days. Calcium and Mg were applied into soil, at the concentration of 5, 10, 15 mg Ca L^{-1}(T2, T3, T4 respectively) and 5, 10, 15 mg Mg L^{-1}(T5, T6, T7 respectively). There was two controls as a follow T0 (soil) and T1 (soil + silica gel). During incubation, Si, Ca, Mg, Fe, and Mn concentrations in surface water were measured using ICP spectroscopy at day 8, 15, and 29. The results show the soil before treatment was slightly acidic (pH 5.7) and extractable Si concentration was 267.1 mg SiO_2 kg^{-1}. It was classified to be below critical level of available Si for rice (300 mg SiO_2 kg^{-1}). Total concentration of Ca and Mg in soil solution were highest for treatment T4 and T7, respectively compared with other treatments. On the first 8 days of incubation, Si released into soil solution was higher in T1 and T2 compared to other treatments. The solubility of Si was significantly positive correlated with Mn, Eh, and negatively correlated with pH, that indicated these were the controlling factors of the Si release in soil solution. There was no correlation between Si and Ca or Mg concentration in soil solution

Keywords: silica, calcium, magnesium, soil solution

INTRODUCTION

Silicon (Si) is a tetravalent metalloid and a ubiquitous element in soil (Liang et al., 2015). Silicon is beneficial element for plant, including rice, wheat, barley, and cucumber (Ma et al., 2001) by increasing available P in soil. Silicon increased soil respiration in P deficient soils (Schaller, 2019). According to Rodrigues and Datnoff (2015), the silicon for plants can reduce the damage caused by insects and grazing animals. Silicon can improve plant growth and enhance plant biomass carbon accumulation under abiotic and biotic stresses in terrestrial ecosystems (Li at al., 2018). Soils in tropical and tropical regions are generally acidic, with silicon (Si) solubility due to weathering and leaching associated with high rainfall and temperatures (Keeping, 2017).According to Haynes (2014), concentration of Si in soil solution is one of the important key parameters in relation to the plant-availabile Si. Silicate can be adsorbed into the surfaces of variable charged soil colloids such Fe and Al hydrous oxides. Silicon in soil solution in the form of H_4SiO_4 is present at concentrations normally ranging from 0.1 to 0.6 mM (Epstein, 1999). Silicon in soil solution is mainly present in the form of an uncharged monomeric molecule, silicic acid ($Si(OH)_4$) at a pH below 9.0 (Ma, 1990).

Plants uptake silicon from the soil in the form of monosilicic acid (H_4SiO_4) (Meena et al., 2014). Monosilicic acid is absorbed by plant roots along with other elements occurring in soil solution (Kamenik et al., 2013). The mechanisms by which silicon is absorbed by plants, i.e active, passive, and rejective. The plants that use active mechanism of silicon uptake have density of transporters in root and shoot. The transporters, a membrane protein, known as channel proteins or aquaporin were coded by low-silicon genes (*Lsi1* and *Ls2* in roots and *Ls6* in shoots). In passive mechanisms, the silicon uptake by plants through mass flow. While in rejective mechanism, the silicon plant uptake is characterized by the build-up of H_4SiO_4 in the soil solution results from the low concentration of silicon that absorbed by the plants (Tubana and Heckman, 2015).

Additions of $CaCO_3$ to soil reduced silica concentration in soil solutions (Roy, 1969). Silicon enhanced oxidation of Fe from ferrous iron to insoluble ferric iron. Therefore, excess Fe uptake was indirectly prevented by Si application (Ma, 2004). Monosilicic acid can interact with Fe and Mn to form slightly soluble silicates (Snyder et al., 2006). Fertilization with silica gel is an option to improve Si for growing of rice plants in soils with low levels of plant-available Si (Marxen et al., 2016). The release of Si from silica gel into soil solution must vary in different soil type. Factors controlling dissolution of Si include iron (Fe), calcium (Ca), manganese (Mn), pH, and particle size of the materials and presence of organic matter (Makabe et al., 2013; Kendrick, 2006). The key factor that controls soil nutrient availability is pH (Zhang et al., 2019). The pH of soil solution is the most relevant to soil and plant biogeochemical processes (Rengel, 2015).

According of Liang et al. (2015), the one important factor that influences the Si soil solubility is Eh. Flooding results in soil reduction, lowering Eh and leading to an increase in the concentration of available soil Si. The solubility of the soil silicon increases by the reduction in the Eh (Ponnamperuma, 1965), According to Greger et al. (2018), silicon affects the uptake of mineral nutrients in plants such as N, P, K, Ca, and Mg. In the present study, we examined the interaction between Si and Ca, as well as Mg in terms of the Si solubility and investigated the behavior of Si in soil solution in relation with Fe, Mn, Eh, and pH.

MATERIALS AND METHODS

Japanese silica gel (JSG) and red clayey soil were collected from Japan. Silica gel was ground into fine powder in agate grinding jars, using a mixer mill (MM 200, Retsch GmbH, Haan, Germany). The soil sample was air dried and passed through a 2 mm sieve. The exchangeable Ca, K, Mg and Na (Ex. Ca, K, Mg, Na) were extracted with 1 M ammonium acetate pH 7.0 and measured by Inductively Coupled Plasma Spectroscopy (ICPE-9000 Shimadzu, Kyoto Japan). The contents of available Fe and Mn were obtained by extraction with 0.1 N HCl and quantified using the ICP. Soil pH (H_2O) was determined on 1:2.5 (w/v) soil: water suspensions with pH meter (D-51, Horiba).

A 2.5 g of 2 mm sieved air-dried soil sample was mixed with 25 ml of 1.0 M acetate buffer (pH 4.0) in a 50 ml plastic bottle and kept in an incubator at 40°Cfor 5 h with intermittent shaking every 15 minutes. The solution was then filtered with a filter paper and the Si concentration of the solution was determined by using molybdenum blue method (Imaizumi and Yoshida, 1958).

Incubation experiment

The amount of dissolved Si, Ca and Mg in soil solution was evaluated by incubating under submerged condition. The soil sample was air dried and passed through a 2 mm sieve. Ten gram of air dried soil was placed into a centrifuge plastic tube, afterward 0.39 g of JSG and 40 ml Ca or Mg solutions were added. The solution was mixed thoroughly using glass stick. The concentration of Ca or Mg in solution were 0, 5, 10, and 15 mg L^{-1}(Table 1).

Table 1 Treatments

Treatments	
T0	Soil + DW
T1	Soil + Silica gel + DW
T2	Soil + Silica gel + 5 mg Ca L^{-1}
T3	Soil + Silica gel + 10 mg Ca L^{-1}
T4	Soil + Silica gel + 15 mg Ca L^{-1}
T5	Soil + Silica gel + 5 mg Mg L^{-1}
T6	Soil + Silica gel + 10 mg Mg L^{-1}
T7	Soil + Silica gel + 15 mg Mg L^{-1}

Note. DW: distilled water

The tube was covered with plastic para film and incubated at 30°C for 29 days. The experiment was replicated three times. At day 8, 15 and 29 of incubation, the redox potential (Eh) and pH of soil solution were measured with Eh and pH meter (TOA HM-14P and D-51 Horiba, respectively) without disturbing the soil. The surface water was collected by using pipette and the supernatant was obtained after filtration (paper filter Advantec No. 6). Silica, Ca, Mg, Fe, and Mn concentrations in supernatant were measured using ICPE-9000 Shimadzu. To resume the incubation, residue on the filter paper was washed back into tube with distilled water. Distilled water was added up to a total volume of 40 mL based on the weight (Makabe et al., 2013). The soil solution was replaced with distilled water at day (d) 8, 15, and 29 assuming field water replacement by drainage / leaching and irrigation in the field. Relationship between solubility of Si with Ca, Mg, Fe, Mn, pH and Eh was analyzed using the statistical package SPSS 22.

RESULTS AND DISCUSSION

Properties of soils

The soil was slightly acidic (pH 5.7) and relatively rich in available Fe and Mn (72.5 and 52.2 mg kg^{-1}). Exchangeable Ca, Mg, K, and Na were 4.3 ; 2.4 ; 0.7 and 0.3 $cmol_c kg^{-1}$, respectively. The extractable Si concentration was 267.1 mg SiO_2 kg^{-1}. According to Sumida (1992), it was classified to be below critical level of available Si for rice (300 mg SiO_2 kg^{-1}), but above deficiency criteria (<86 mg SiO_2 kg^{-1}) by Doberman and Fairhurst (2000).

Solubility of Si, Ca, Mg, Fe and Mn in soil solution

Figure 1 and 2 show the different Si, Ca, Mg, Fe and Mn concentration in soil solution. Soil solution was higher in Ca concentration for T4, followed with treatment T3, T2, and T1. As expected, the lowest Ca and Mg concentration was treatment T0. Meanwhile, Mg concentration was highest for T7 compared with T6, T5 and T1.

Although statistically not significantly different but Si release into soil solution (Fig 1b) was higher for T1 (1.336 mmol L^{-1}) and T2 (1.335 mmol L^{-1}) compared to T3 and T4 (1.163 and 1.244 mmol L^{-1}, respectively) on the first 8 days of incubation. Meanwhile, the Si concentration was higher for T1 compared to T5 – T7 (Fig 2b). This might be possible because Ca and Mg decreased Si release into soil solution for

T3 and T4 and T5 – T7 (1.255 – 1.276 mmol L^{-1}), respectively. It seems that Ca and Mg inhibited the solubility of Si by binding Si as $CaSiO_3$ or $MgSiO_3$ in soil/solid phase. In submerged condition and by the time of incubation, it is possible silica will be released into soil solution through the following reaction mechanism (Flint & Wells, 1934):

$$\text{Soil} + \text{water} \leftrightarrow \text{soil solution}$$

$$MgSiO_{3(s)} + 3\ H_2O \leftrightarrow Mg^{2+}_{(aq)} + H_4SiO_{4(aq)} + 2OH^-_{(aq)}$$

$$\textit{Or}\ MgOH^+_{(aq)} + H_4SiO_{4(aq)} + OH^-_{(aq)}$$

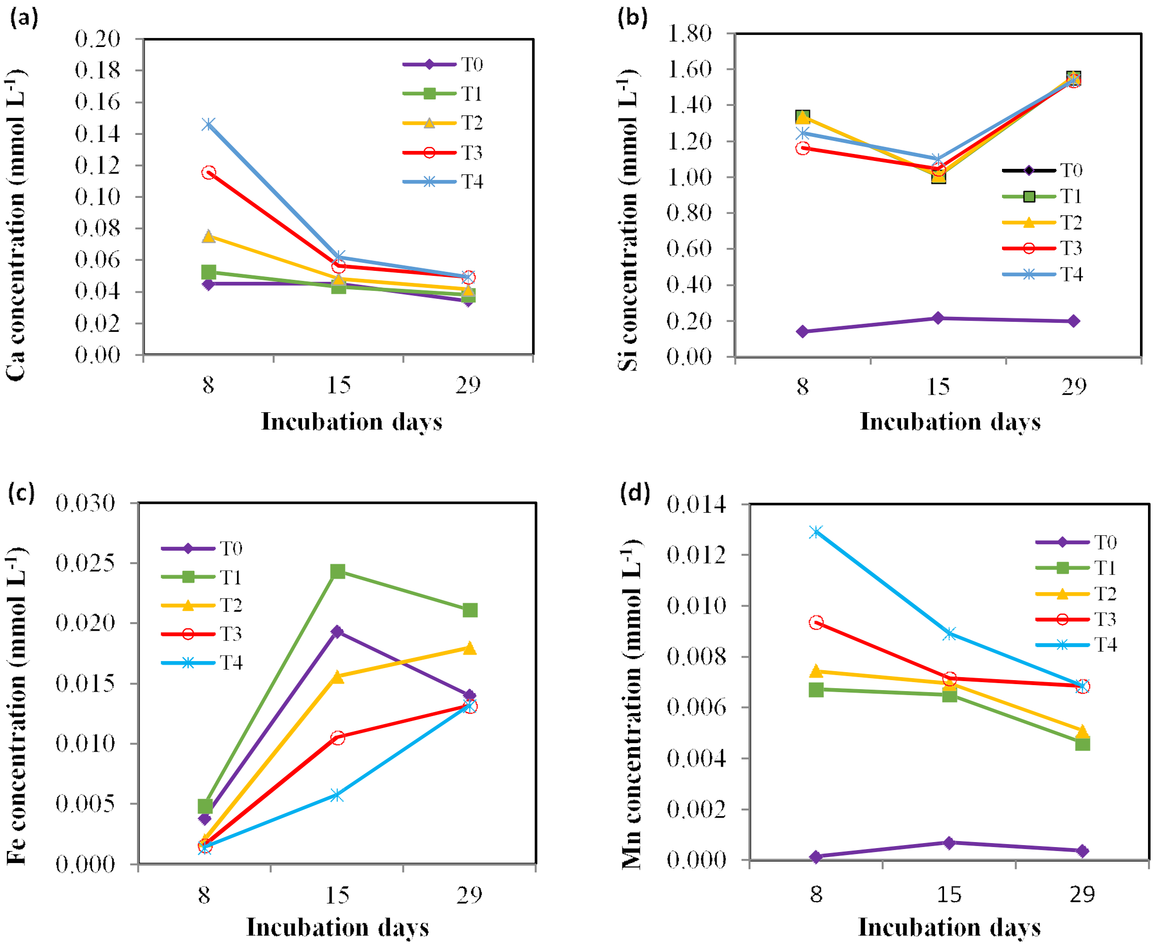

Figure 1 Release pattern of calcium (a), silicon (b), iron (c), and manganese (d) during 29 days of experimental time

In this research, most of Fe concentrations in soil solution were lower on day 8 compared to other days, specifically Fe was lower in T2 to T4 (Fig 1c) and T5 to T7 (Fig 2c) compared to T0 and T1. In contrast, Mn concentration was higher in treatment with Ca and Mg application (Fig 1d and 2d, respectively).

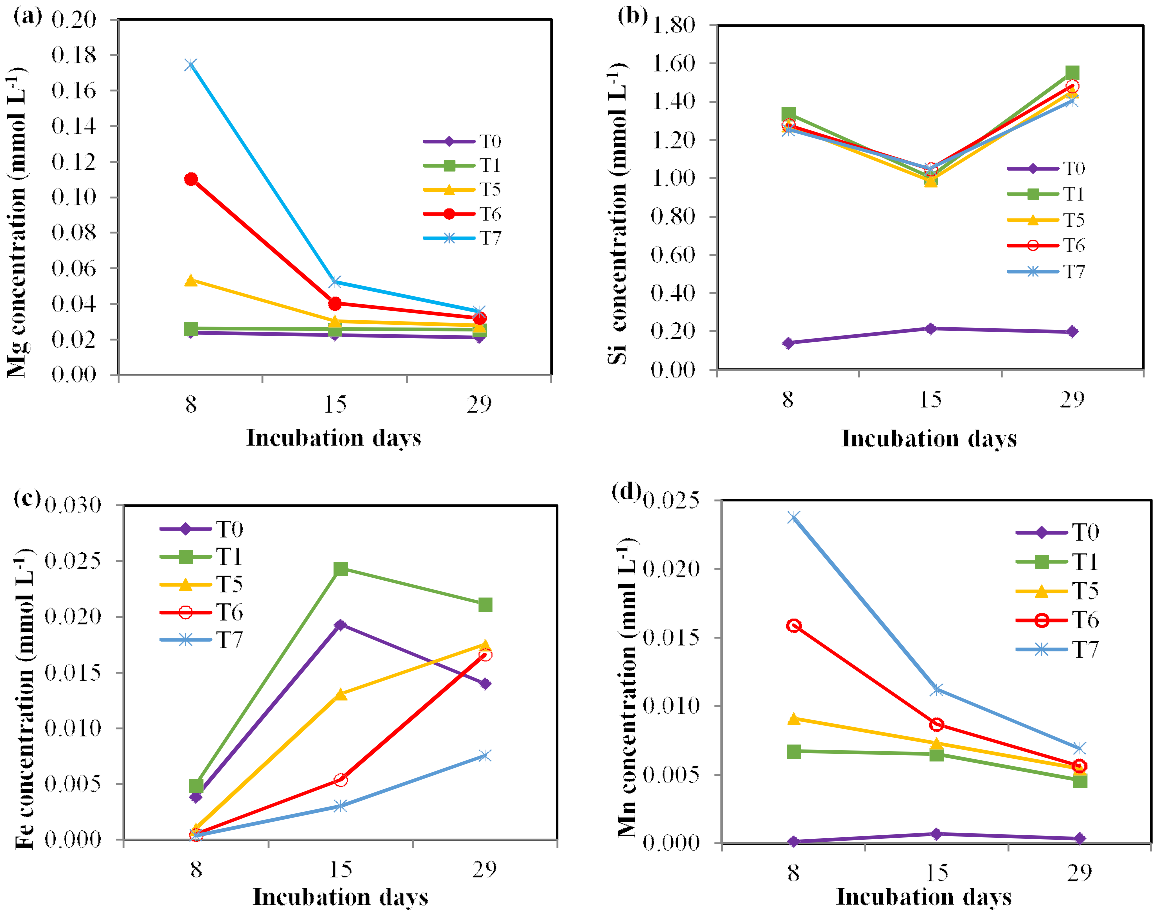

Figure 2 Release pattern of magnesium (a), silicon (b), iron (c), and manganese (d) during 29 days of experimental time

Silicon concentration in T1 and T7 decreased from day 8 to day 15 and then increased on day 29. Low Si concentration on day 15 was probably due to replacement of soil solution with distilled water and was not strong enough to release Si that is adsorbed by soil into soil solution, although the Si existed in easily releasable form (Makabe et al., 2013). Concentration of Si increased again on day 29, it might be due to the soil Si and Si from silica gel that were adsorbed by the soil could enter the soil solution when concentration of Si in soil solution decreases (Kato and Owa, 1997). According to Ponnamperuma (1972), the Si concentration in the solutions of submerged soils increases slightly after flooding. The release of Si after flooding may be due to reduction of hydrous oxides of Fe (III) sorbing Si and action of CO_2 on aluminosilicates (Briker & Godfrey, 1967). Liang et al. reported that concentration of Si slightly increased after flooding and then gradually decreased.

Furthermore, the release of Ca and Mg were higher on the first 8 days and sharply decreased, thus slightly decreased on day 29. We assume that the soil solution replacement with distilled water would reduce Ca and Mg concentration. However, the Ca and Mg concentration in the soil solution of T3 - T4 and T5 - T7, respectively were higher than those of other treatments.

Generally, Fe concentration increased during incubation day (Fig 1c and 2c), in which Fe was higher on day 15 and then decreased. These probably because soil was red clayey that contain more Fe, so need more time to reduce Fe in soil solution. The concentration of Mn (1d and 2d) tended to decrease during incubation days. In addition, Ca and Mg application decreased Mn concentration for T2 to T7. It might be due to displacement of Ca ion sites on soil colloids with Fe ions so that Fe in soil solution declined.

Meanwhile, Ca and Mg application increased Mn concentration for T1 to T7. This result related to pH of T1 to T7 which was lower compared to control (T0). Husson (2013) reported that solubilization of Mn^{2+} ions is a function of both pH and Eh. According to Schwab and Lindsay (1982), higher Mn bioavailability corresponds to a decrease in Eh. In this experiment, pH was the dominant factor for dissolution of Mn. Meanwhile Eh was not the main factor of the Mn dissolution as the Eh increased but concentration of Mn increased in soil solution. However disagreed with finding of Schwab and Lindsay (1982), the increase in Eh resulted in the increase of Mn in soil solution.

The Amounts of Si, Ca, Mg, Fe, Mn concentration, mean pH and Eh during incubation

The amount of Ca and Mn increased in soil solution which was subjected to Ca application. However, we could not explain how Mn concentration elevated in soil solution after the Ca application. Addition of Ca decreased Si concentration in the soil solution of treatment T3 and T4 (3.745 and 3.881 mmol L^{-1}, respectively) (Table 2). Similarly, the Fe concentration in the soil solution decreased with addition of Ca.

The solubility of Si (3.708 mmol L^{-1}) was lowest in which high concentration of Mg) was added to T7. The solubility of Mn in soil solution increased by the Mg treatment. In line with additional Ca, the increase Mg application doses led to the decrease in Fe concentration. An explanation for this finding might be that Eh was higher in soil solution with Ca and Mg application compared with control.

Table 2 Cumulative concentration of Si and other elements in supernatant during 29 days

Treatments	Si	Ca	Mg	Fe	Mn
			----------- mmol L^{-1} -----------		
T0	0.550	0.125	0.068	0.037	0.001
T1	3.893	0.134	0.077	0.050	0.018
T2	3.900	0.165	-	0.036	0.019
T3	3.745	0.221	-	0.025	0.023
T4	3.881	0.257	-	0.020	0.029
T5	3.715	-	0.112	0.032	0.022
T6	3.810	-	0.183	0.022	0.030
T7	3.708	-	0.263	0.011	0.042

Note. -: not determined

The amount of Ca and Mg concentration in supernatant was higher for T4 and T7 (0.257 and 0.263 mmol L^{-1}, respectively). The mean of pH (Table 3) in soil solution was lower for T2 to T7 than T1 or T0. It was similar with Si concentration. The explanation for these results might be that as alkali is consumed, the pH decreases, the concentration of Ca^{2+} increases, and then the concentration of Si in solution decreases. While, if alkali concentrations remain high, Ca concentrations are very low, and the loosened Si structure imbibes more liquid and hydroxyl ion that continue the process of dissolution of the Si (Helmuth et al., 1993). limiting factors of Si release in this soil. This finding is in accordance with finding's Liang et al. (2015) that the increase in Eh soil would increase the availability of Si.

The relationship between Si and Ca or Mg concentration in soil solution was not significant. However, Ca and Mg application reduced Si concentration in soil solution. Makabe et al. (2013) reported that Si and Ca were significantly negatively correlated in soil solution during incubation.

Meanwhile, Ca was negatively correlated with Fe and pH. Furthermore Ca was significant positively correlated with Mn (0.900) and Eh (0.694). As Ca and Mg was also positively correlated with Mn (r = 0.901) and Eh (r = 0.591). In contrast, Mg was significant negatively with Fe (r = 0.55) and pH (r = 0.49). Silicon present in soil solutions is in the form of monosilicic acid (H_4SiO_4) (Jones and Handreck, 1967; Epstein, 1994). According to Liang et al. (2015), monosilicic acids concentration is strongly depends on soil pH. Szulc et al. (2015) found that a factor significantly affecting the exchangeable silicon content of the soil pH. Tavakkoli et al. (2011) reported that concentration of Si solution was controlled by polymerization reaction in equilibrium with specific soil pH.

Table 3 The mean pH and Eh values of soil solution during 29 days

Treatments	T0	T1	T2	T3	T4	T5	T6	T7
pH	5.1	4.8	4.7	4.7	4.6	4.6	4.6	4.6
Eh (mV)	254	281	297	301	310	309	313	317

It shows that Ca and Mg increased Eh in soil solution. For this reason, Fe and Si solubility was low in soil solution under less reduction condition. Snyder et al. (2006) reported that Si release such as monosilicic acid and polysilicic acid that have high chemical activity can react with Fe in the formation of slightly soluble silicate.

$$2Fe_2SiO_4 + 4H^+ = 2Fe_2^+ + 2H_4SiO_4$$

Relationships between solubility of Si with Ca, Fe, Mn, pH and Eh

Generally, the addition of Ca or Mg into soil solution, the results indicated that Si concentration was significantly positively correlated with Mn concentration (r = 0.401), Eh (r = 0.381), and negatively correlated with pH (r = 0.671) in soil solution (Fig. 3). Those results indicate that Mn, Eh and pH are the

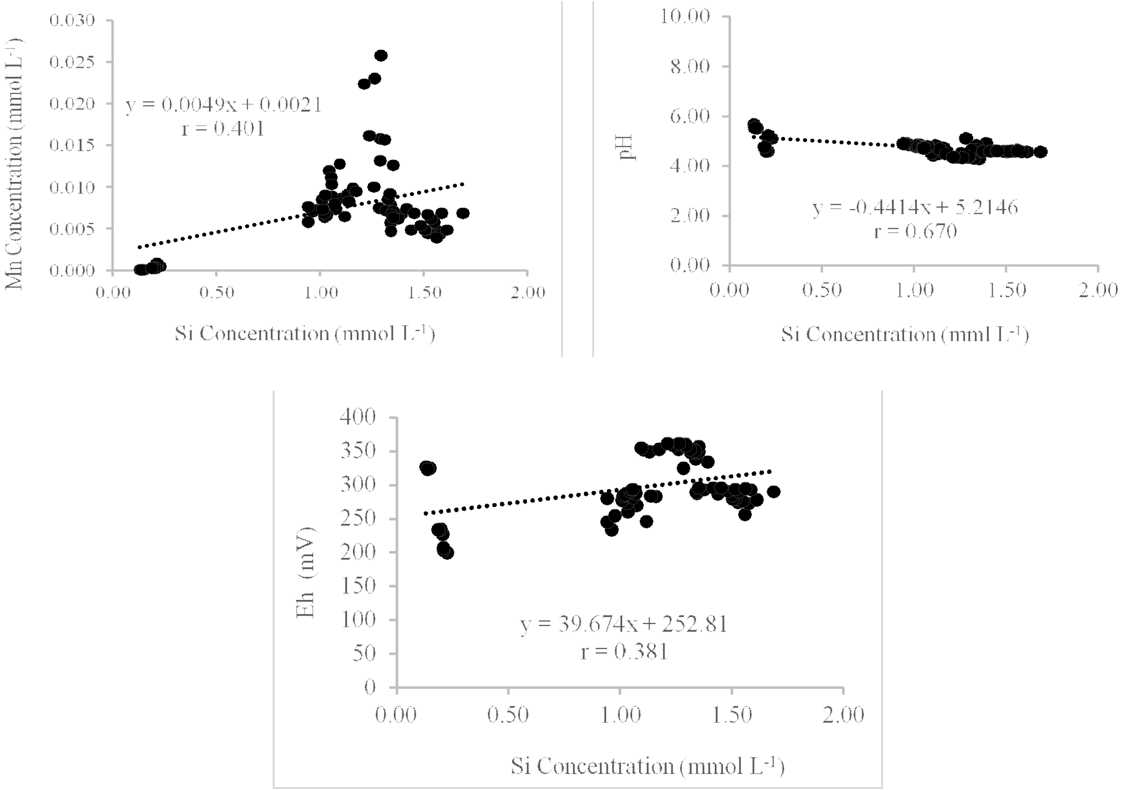

Figure 3 Correlations coefficient of Si concentration with Mn, pH and Eh in soil solutions

CONCLUSIONS

The solubility of Si was significantly positive correlated with Mn, Eh, and negatively correlated with pH, that indicated these were the controlling factors of the Si release in soil solution. There was no correlation between Si and Ca or Mg concentration in soil solution. However, Ca and Mg application reduced Si concentration in soil solution.

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