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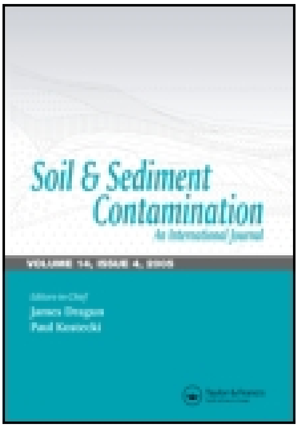

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Impact of Simulated Acid Rain on Trace Metals and Aluminum Leaching in Latosol from Guangdong Province, China

Jia-En Zhang [a] , Jiayu Yu [a] , Ying Ouyang [b] & Huaqin Xu [a]
[a] Department of Ecology , South China Agricultural University , Guangzhou , China
[b] USDA Forest Service, Mississippi State , Mississippi , USA
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Impact of Simulated Acid Rain on Trace Metals and Aluminum Leaching in Latosol from Guangdong Province, China

JIA-EN ZHANG,[1] JIAYU YU,[1] YING OUYANG,[2]
AND HUAQIN XU[1]

[1]Department of Ecology, South China Agricultural University, Guangzhou, China
[2]USDA Forest Service, Mississippi State, Mississippi, USA

Acid rain is one of the most serious ecological and environmental problems worldwide. This study investigated the impacts of simulated acid rain (SAR) upon leaching of trace metals and aluminum (Al) from a soil. Soil pot leaching experiments were performed to investigate the impacts of SAR at five different pH levels (or treatments) over a 34-day period upon the release of trace metals (i.e., Cu, Ni, Pb, Zn, and Fe) and Al from the Latosol (acidic red soil). The concentrations of trace metals in the effluent increased as the SAR pH level decreased, and were highest at the SAR pH = 2.0. In general, the concentrations of Cu, Pb, Fe, and Al in the effluent increased with leaching time at the SAR pH = 2.0, whereas the concentrations of Zn, Fe, and Al in the effluent decreased with leaching time at the SAR pH ≥ 4.0. The increase in electrical conductivity (EC) with leaching time at five different SAR pH levels was primarily due to the concentrations of Al and Fe in the effluent. There were good linear correlations between the effluent Al concentrations and the effluent pH at the SAR pH = 2.0 (R^2 = 0.87) and pH = 3.0 (R^2 = 0.83). More soil trace metals and Al were activated and released into the soil solution as the SAR pH level decreased.

Keywords Acid rain, electrical conductivity, trace metal, Latosol

Introduction

Trace metals are introduced into the environment naturally through the weathering of parent materials as well as from a variety of human activities. Some trace metals, such as Cu and Zn, are essential micronutrients required for the growth of organisms, while others, such as Pb and Cd, are not required for growth and have been considered to be noxious to human health and aquatic life. All trace metals exert toxic effects at high concentrations, including those that are essential micronutrients. Trace metal contamination in soils and waters is an issue of environmental concern. Such contamination is often caused by human activities, including mining, smelting, electroplating, and other industrial processes with metal residues in their wastes, and agricultural practices with chemical fertilizers and pesticides containing trace metals (Ouyang *et al.*, 2002; Wu *et al.*, 2007; Lin *et al.*, 2010).

Address correspondence to Jia-En Zhang, Department of Ecology, South China Agricultural University, Wushan Road, Tianhe, Guangzhou, China. E-mail: jeanzh@scau.edu.cn

Aluminum (Al) is the most abundant metal in the earth's crust and it accounts for about 7% of the mass (by weight) of Earth's surface. There are multiple forms of Al in the soils and most of them are not directly harmful to plants and organisms (Brady, 1984). Aluminum is usually bound to the negatively charged surface of soil particles. Under the influence of acid rain, adsorbed Al can be displaced by H^+ ion or other cations (Brady, 1984; Liu *et al.*, 1990). The excess Al in soil water is toxic to aquatic organisms and harmful to soil organisms and plants (Kochian, 1990; Menz and Seip, 2004).

Acid rain is rain containing an unnatural acidity and is one of the most serious ecological and environmental problems worldwide besides global warming and ozone zone damage (Galloway, 1995; Tao and Feng, 2000). The typical pH values of acid rain result from anthropogenic emissions ranging from 3.5 to 5.0 (Menz and Seip, 2004). Acid rain causes fish kills and plant deaths in water, and is unfavorable to human beings because we eat these fish, drink these waters, and consume foods from these plants. Acid rain with SO_2, NO_x, and NH_3 may form secondary pollutants such as fine (PM 2.5 or particles less than 2.5 μm in diameter) particles and nitrogen species that could react with organic compounds and contribute to ozone (O_3) depletion. The corrosion of many materials increases with SO_2 concentration in the atmosphere. Ozone and rain acidity may also contribute to material damage (Menz and Seip, 2004). About 40% of the world's arable lands and about 70% of non-arable lands are acidic. These lands are primarily distributed in tropic, sub-tropic, and temperate areas (Kochian, 1995).

Acid rain can mobilize base cations, decompose organic compound, and lose mineral structure in the soil. The H^+ ion in acidic water displaces the cations through cationic exchange from their binding sites, reduces the cation exchange capacity, and increases the concentrations of these cations in soil water (Brady, 1984; Liu *et al.*, 1990). Calace *et al.* (2001) reported that the structure composition of fulvic acid from organic matter changes when soil acidification increases and a shift toward smaller molecular weight compounds is observed due to the break-up of the original molecules in acidic conditions. The hydrolysis process leads to an increase in carboxyl groups accompanied with a decrease in molecular weight. The final result is an increase in molecular fulvic acid solubility and the leaching of some of the acids. Acid rain can also increase the weathering of silicate minerals, phosphates, vanadates, and carbonates in soils, which leads to a loss of mineral structure and possibly reduced soil fertility. Recently, Zhang *et al.* (2007) investigated impacts of simulated acid rain (SAR) on cation leaching from the Latosol in south China. A linear increase in effluent K^+ concentration was found at the SAR pH <3.0, whereas an exponential decrease in effluent Na^+ concentration was observed at all levels of the SAR pH. In general, leaching of Ca^{2+} and Mg^{2+} from the Latosol increases as the SAR pH decreases. Ling *et al.* (2007) reported that about 34%, 46%, 20%, and 77% of the original exchangeable soil Ca^{2+}, Mg^{2+}, K^+, and Na^+, respectively, are leached out by the SAR at pH 2.5 after 21 days. These studies have provided insightful information on the impacts of the SAR upon soil cations and nutrients.

A thorough literature search reveals that very few efforts have been devoted to investigating the impacts of acid rain on the mobilization of potentially harmful trace metals and Al from soils. Liao *et al.* (2007) investigated the effects of acid rain on competitive releases of Cd, Cu, and Zn from two natural soils and two contaminated soils in Hunan, China. These authors showed that the total releases are Zn > Cu > Cd for the natural soils and Cd > Zn > Cu for the contaminated soils, which reflected sensitivity of these metals to acid rain. Although this study has provided useful insights into the impacts of acid rain on certain trace metals' release from soils, more efforts are needed to fully understand the leaching patterns of trace metals under different pH levels.

The objectives of this study were therefore to: (1) investigate the leaching of trace metals including copper (Cu), iron (Fe), nickel (Ni), lead (Pb), and zinc (Zn) from the Latosol by simulated acid rain (SAR) with five different pH levels; (2) estimate the electrical conductivity changes under the influence of the SAR; and (3) evaluate the leaching of Al from the Latosol and its relationship with effluent pH associated with the impacts of the SAR.

Materials and Methods

Materials and Acid Rain Preparation

Top 20 cm Latosol collected from a forest garden located on the campus of South China Agricultural University, Guangdong Province, China, was used for the experiments. Latosol (acidic red soil) arises in the tropical rainforest biome, where high temperature and high precipitation occur throughout the year. Climatic conditions are related to the extensive chemical weathering of parent rock material, which results in the development of a deep soil profile (20 to 30 m). This soil has a loose structure and suffers from rapid erosion during rainfall, which results in leaching of nutrients and metals.

This soil has pH 4.3 with an organic matter content of 44.95 g kg^{-1}, a cation exchange capacity (CEC) of 6.69×10^{-2} mole kg^{-1}, and a base saturation (BS) of 12.9%. The soil has 1.75 g kg^{-1} total nitrogen, 0.41 g kg^{-1} total phosphorus, 6.22 g kg^{-1} total potassium, 11% sand, 28% silt, and 61% clay. The initial soil H^+ and Al^{3+} contents were 3.74 and 32.09 mmole kg^{-1}, respectively, whereas the initial soil Cu, Zn, Pb, Ni, and Fe were 20.01, 54.85, 30.14, 9.25, and 14.7×10^4 mg kg^{-1}. In south China, acid rain primarily consists of H_2SO_4 and HNO_3 with a ratio of 4 to 1 (Zhang *et al.*, 2007). In order to have SAR reflecting the natural conditions in south China, the stock acid solution was prepared using this ratio. Working solutions of acid rain with pH 2.0, 3.0, 4.0, 5.0, and 6.5 (CK or control) were prepared in volumetric flasks by diluting the stock solution with deionized (DI) water. Analytical-grade sulfuric acid (H_2SO_4) and nitric acid (HNO_3) used for preparation of stock solution were purchased as standard catalog items from a commercial supplier (Guangzhou Chemical Reagent Manufacture Plant, China).

Application of Simulated Acid Rain to Soil Pots

A pot with a top inner diameter of 19 cm and a bottom inner diameter of 14 cm was used to contain an 18-cm soil pot. 3000 g of air-dried soil, passed through a 2-mm sieve and mixed thoroughly, was then poured into the pot in 2-cm increments and stirred to prevent layering. The soil pot was tapped to settle the soil to a bulk density of 1.6g cm^{-3}. A hole with a diameter of 3.5 cm was drilled at the bottom-center of the pot to collect effluent. Prior to and after filling the pot, a piece of plastic filler and two pieces of paper filters were placed at both ends of the pot to prevent soil leakage. A total of five pot leaching treatments (i.e., one pH level for each treatment) with four replications were carried out in this study.

To reflect natural rainfall conditions, an intermittent influent application method was employed. That is, a 250-mL influent of the SAR was slowly sprayed on the top of the pot every 24 h. Leaching experiments were performed for 34 days with a total of 8500 mL influent of SAR. This volume of SAR was equivalent to three years of acid rain in south China.

The effluent samples from the bottom of the soil pots were collected every 48 h. It should be noted that the first effluent samples were collected after a four-day leaching because there was not enough effluent sample within the first four days. The contents of Cu, Fe, Ni, Pb, and Zn in the samples were analyzed using the HF–$HClO_4$–HNO_3 digestion method (Lu, 1999). Concentrations of the trace metals in solutions were determined using an Atomic Absorption Spectrophotometer (Hitachi Z-5300) and calibrated using the national standard samples obtained from the Ministry of Agriculture of China, whereas the contents of Al in the samples were analyzed using ICP-AES (Varian 710-ES) (Sparks, 1996). Effluent pH was measured with a soil-to-water ratio of 1:2.5 using a pH meter, and the electrical conductivity was measured with a DDS-11A meter. Comparisons of the differences among the five different SAR treatments were performed using DUNCAN statistics with SAS 8.1. All of the differences were statistically significant at $\alpha = 0.05$. The average values were used for plotting the figures.

Results and Discussion

Impact of the SAR on Trace Metal Leaching

Copper and Ni are essential micronutrients for growth of plants and other organisms (Kabata-Pendias and Pendias, 1991). However, the excess Cu and Ni presented in the soil are harmful to plant growth, while the extra Cu and Ni accumulated in the food chain could pose a health threat to humans (Li, 1996). Leaching of Cu from the soil pots by the SAR at five different pH levels is shown in Figure 1A. The concentrations of Cu in the effluent samples increased as the SAR pH level decreased. For example, there was about 4.5-fold increase in the Cu concentration at day 28 when the SAR pH level decreased from 6.5 (CK) to 2.0. This occurred because more Cu in the soil was activated and released to the soil solution as the SAR pH level decreased. The decrease in the SAR pH level increased the soil solution H^+ content. The H^+ ion in the acidic water displaced the cations from their binding sites, reduced the cation exchange capacity, and increased the concentrations of these cations in the soil water (Brady, 1984). Overall, leaching of Cu from the soil was highest at the SAR pH = 2.0 and the maximum Cu concentration in the effluent was 0.06 mg/L at day 28 for this pH level. Figure 1A further demonstrated that the concentrations of Cu in the effluent, in general, increased with SAR leaching time. The highest concentration of Cu in the effluent samples was found at the end of the experiment for all of the treatments, except for the pH = 2 treatment, which was found at day 28. The results implied that more Cu in the soil was released into the soil solution as the leaching time increased when the SAR pH level was 2.0.

Leaching of Ni from the soil pots by the SAR at five different pH levels is shown in Figure 1B. Analogous to the case of Cu, the concentration of Ni in the effluent increased as the SAR pH level decreased, and was highest at SAR pH = 2.0. There was about a 4.9-fold increase in Ni concentration at day 4 when the SAR pH level decreased from 6.5 (CK) to 2.0. The results revealed that more Ni in the soil was activated and released to the soil solution as the SAR pH level decreased, for the same reasons as in the case of Cu. In addition, the concentration of Ni in the effluent showed a decreasing pattern with leaching time for the SAR at pH = 2.0 and a fluctuated pattern with leaching time for the SAR with pH ranging from 3.0 to 6.5.

Lead is not required for the growth of plants and other organisms, and it is one of the most toxic trace metals in the environment (Kabata-Pendias and Pendias, 1991). Lead in the bloodstream has been shown to produce toxic effects in children, such as lowered

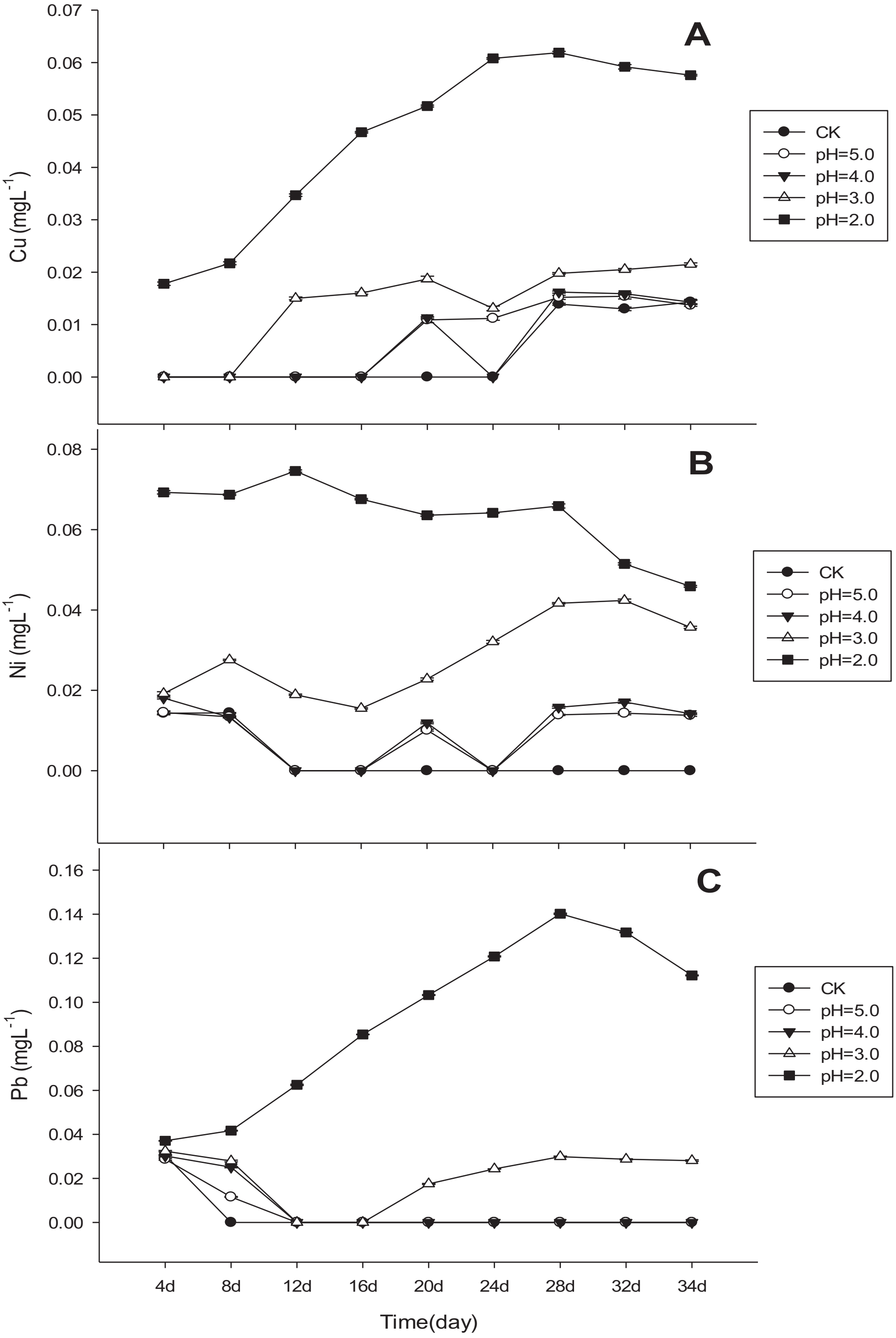

Figure 1. Effluent concentrations of Cu (A), Ni (B), and Pb (C) at five different simulated acid rain pH levels. CK denotes control treatment at pH = 6.5.

intelligence and behavioral dysfunctions. In senior adults, Pb exposure causes high blood pressure, which is associated with increased risks of heart attacks, strokes, and death (Driscoll *et al.*, 1992). Leaching of Pb from the soil pots by the SAR at five different SAR pH levels is shown in Figure 1C. Analogous to the case of Cu, the concentration of Pb in the effluent increased as the SAR pH level decreased and was highest at SAR pH = 2.0. Unlike the case of Cu, however, the concentration of Pb in the effluent showed a mix pattern: increased from day 4 to 28 and decreased from day 28 to the end of the experiment at the SAR pH = 2.0; decreased from day 4 to 16 and increased from day 16 to the end of the experiment at the SAR pH = 3.0; and decreased with time for the SAR pH level $\geq$3.0. There was about a 4.7-fold increase in Pb concentration at day 28 when the SAR pH level decreased from 3.0 to 2.0. A comparison of Figures 1A and Figures 1C shows that both the Cu and Pb in the effluent had a similar leaching pattern and reached their maximums at day 28 at SAR pH = 2.0. It is apparent that more Pb in the soil is activated and released to the soil solution when the SAR pH level is 2.0. Although the exact reasons for the decrease of Pb in the effluent after 28 d (Figure 1C) remain to be investigated, a possible explanation could be the depletion of soil Pb with less soluble Pb available for leaching.

Zinc is an essential micronutrient for the growth of plants and other organisms (Lin *et al.*, 2010). Leaching of Zn from the soil pots by the SAR at five different pH levels is shown in Figure 2A. Analogous to the case of Cu, the concentration of Zn in the effluent increased as the SAR pH level decreased and was highest at SAR pH = 2.0. Unlike the case of Cu, the concentration of Zn in the effluent decreased with leaching time for all five treatments. For example, the concentration of Zn in the effluent was about 1.1 mg/L at day 4 and about 0.62 mg/L at day 34 with SAR pH = 2.0. In addition, there was about a 2.4-fold increase in Zn concentration at day 8 when the SAR pH level decreased from 6.5 (CK) to 2.0. This occurred because more Zn in the soil was activated and released to the soil solution as the SAR pH = 2.0, resulting from the H^+ ion in the soil solution that displaced Zn from its binding sites and reduced the Zn exchange capacity. It is noted that at the lowest SAR pH = 2.0, the content of almost all trace metals, as well as Al, increased with time. Only for Zn, and to a lesser extent for Ni, did the concentrations decrease with time at pH = 2.0. Taking into consideration that adsorption of Zn cations or precipitation of Zn hydroxides occurs at higher pH values, this effect cannot be explained and requires further investigation. A comparison of leaching experiments between Cu and Zn revealed that the total release was Zn (1.04%) > Cu (0.13%). This finding was consistent with that reported by Liao *et al.* (2007).

Iron is an essential macronutrient for the growth of plants and other organisms. Leaching of Fe from the soil pots by the SAR at five different pH levels is shown in Figure 2B. The concentration of Fe in the effluent increased from day 4 to 16 and decreased from day 16 to the end of the experiment for the SAR pH from 3.0 to 6.5. Analogous to the case of Cu, the concentration of Fe in the effluent increased as the SAR pH level decreased and was highest at SAR pH = 2.0. There was about a 9.9-fold increase in Fe concentration at day 32 when the SAR pH level decreased from 6.5 (CK) to 2.0. The results suggest that more Fe in the soil was activated and released into the soil solution as the SAR pH level decreased, for the same reasons as that of Cu.

Compared with the Chinese surface water quality standards, the maximum concentrations of Cu, Zn, and Pb measured during the experiments were found to exceed the Level I criteria of the Chinese surface water quality standards, which correspond to 0.01 mg L^{-1} for Cu, 0.05 mg L^{-1} for Zn, and 0.01mg L^{-1} for Pb. Thus, further study is required to obtain more accurate estimates about potential adverse environmental impacts of acid rain upon ground- and surface water quality.

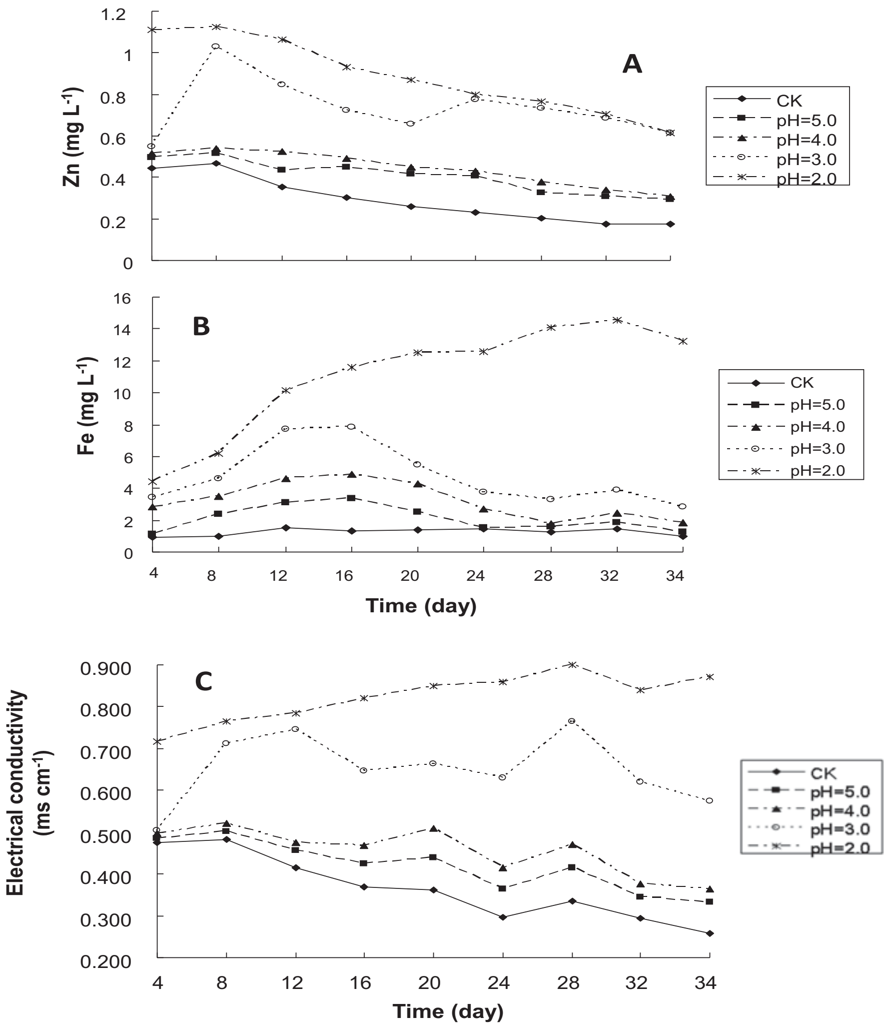

Figure 2. Effluent concentrations of Zn (A) and Fe (B) as well as electrical conductivity (C) at five different simulated acid rain pH levels. CK denotes control treatment at pH = 6.5.

Impact of SAR on Electrical Conductivity

Changes in electrical conductivity (EC) in the effluent from the soil pots at five different SAR pH levels are shown in Figure 2C. The following two distinct patterns were observed: (1) the EC increased with leaching time at the SAR pH = 2.0; and (2) the EC decreased with leaching time when the SAR pH was ≥ 4. The first pattern was similar to those of Cu, Pb, Fe, and Al (see Figures 1a, Figures 1b, Figures 2b and Figures 3a) at SAR pH = 2.0, while the second pattern was more or less similar to those of Zn and Al at SAR pH ≥ 4 (see Figures 2a and Figures 3a). On a molecular basis, the concentrations of Cu, Ni, and Pb did not exceed the level of 1.5×10^{-6}mol L^{-1} and that of Zn did not exceed the level of 17×10^{-6}mol L^{-1}, while Al and Fe were found to vary within the ranges 19-3700 × 10^{-6}mol L^{-1} and 18-250 × 10^{-6}mol L^{-1}, respectively. It is thus evident that the changes in EC pattern are primarily dictated by the changes in the concentration of Al and, to a lesser extent, of Fe.

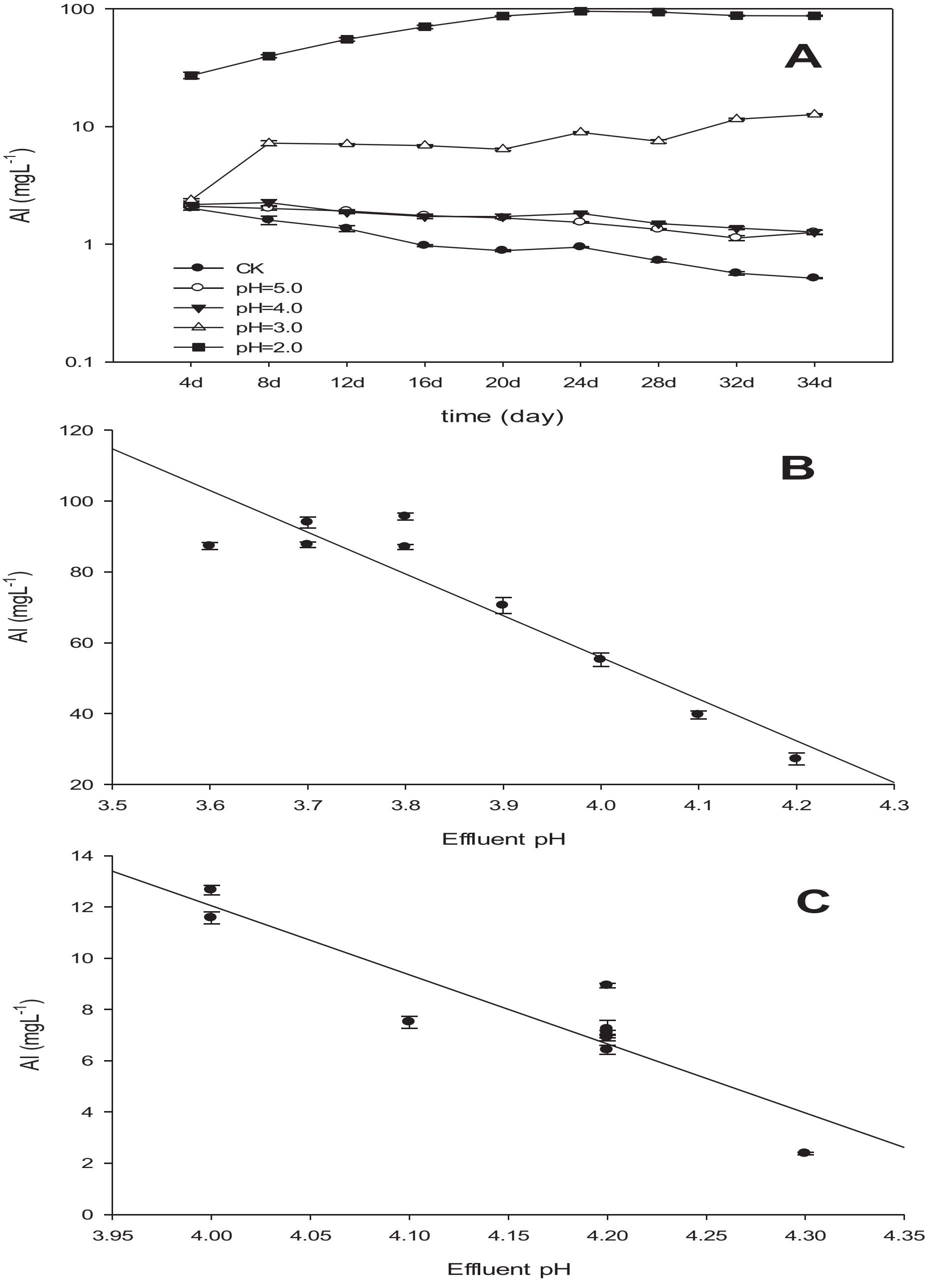

Figure 3. Effluent concentrations of Al (A) at five different simulated acid rain pH levels as well as relationships between the effluent Al content and the effluent pH at the simulated acid rain pH levels of 2.0 (B) and 3.0 (C). CK denotes control treatment at pH = 6.5.

Impact of the SAR on Al and Effluent pH

Leaching of Al from the soil pots by the SAR at five different pH levels (or treatments) is shown in Figure 3A. Two distinct leaching patterns were observed: one for the SAR pH from 4.0 to 6.5 and the other for the SAR pH from 2.0 to 3.0. For the SAR pH levels from 4.0 to 6.5, the concentration of Al in the effluent decreased with time. For example, the content of Al in the effluent was 2.1 mgL^{-1} at pH 5.0 on day 4 and was 1.3 mgL^{-1} at the same pH level on day 34. The former was about 1.6 times greater than the latter. The opposite results were obtained for pH levels from 2.0 to 3.0. The concentration of Al in the effluent increased with leaching time. For instance, the concentration of Al in the effluent was 2.3 mg L^{-1} at pH 3.0 on day 4 and was 12.7 mg L^{-1} at the same pH level on day 34. The former was about 5.5 times less than the latter.

A probable explanation for this trend is that the soil contains relatively soluble Al mineral phases, such as amorphous, low crystallinity gibbsite, etc. At higher SAR pHs (i.e., 4, 5, and 6.5), these phases may be converted to thermodynamically more stable Al oxyhydroxides or oxides (e.g., well crystalline gibbsite, bohemite, etc.), a process that is followed by a decrease in Al concentrations. On the contrary, at lower SAR pH values (i.e., 2 and 3), the preponderant process seems to be the gradual dissolution of the relatively soluble Al mineral. The results show that the impact of acid rain on soil Al release was profound.

What is more interesting from this study, however, are the correlations between the Al concentration and the effluent pH at different SAR pH levels. Figures 3B and Figures 3C show the plots of the concentrations of Al in the effluent against the effluent pH at SAR pH levels of 2.0 and 3.0. It should be pointed out here that the SAR pH was the pH in the influent used to leach the soil pots, whereas the effluent pH was the pH in the effluent that flowed out of the soil pots. The effluent pH increased as the effluent Al content decreased at both the SAR pH levels (i.e., 2.0 and 3.0). There were good linear correlations between the effluent Al concentration and the effluent pH at SAR pH levels of 2.0 ($R^2 = 0.87$) and 3.0 ($R^2 = 0.83$). Calculations were carried out considering the total concentration of Al in the aqueous phase. However, it should be noted that Al exists in the form of many different hydrolysis species in the aqueous solutions (e.g., Al^{3+}, $Al(OH)^{2+}$, etc. According to Sarpola (2007), the preponderant forms are Al^{3+} below pH 3 and $Al(OH)^{+2}$ between pH 4 and 5. Detailed speciation calculations could provide more accurate predictions about the effect of pH on Al release. In any case, the experimental results suggest that acidic rains can cause an important mobilization of Al from Latosols.

It is worthwhile to mention that Latosol presents a certain acid-buffering capacity, which is evident when the soil is treated with the two acidic SAR pHs, 2 and 3. Namely, when SAR pH is 2, the effluent pH ranges between 4.2 and 3.6, and when SAR pH is 3, effluent pH varies between 4.3 and 4.0. It is evident that the soil neutralizes part of the acidity introduced with the repeated applications of SAR (at low SAR pHs). On the contrary, when SAR pH is 4.0, 5.0, and 6.5, the effluent pH is always close to natural soil pH 4.3, which means that the inherent acidity of the soil overcomes the acidity introduced with the SAR solution.

Conclusions

Laboratory experiments were performed to investigate the impacts of simulated acid rain (SAR) at five different pH levels over a 34-day period upon leaching of trace metals and Al from the Latosol (acidic red soil). Our study showed that the concentrations of

Cu in the effluent samples increased as the SAR pH level decreased, which occurred because more Cu in the soil was activated and released to the soil solution as the SAR pH level decreased. In general, the concentration of Cu in the effluent increased with SAR leaching time, and the leaching of Cu from the soil was highest with SAR pH = 2.0.

Analogous to the case of Cu, the concentrations of Ni, Pb, and Fe in the effluent increased as the SAR pH level decreased, and were highest at the SAR pH = 2.0. Unlike the case of Cu, the concentrations of Zn in the effluent samples decreased with leaching time for all five treatments.

Two distinct patterns were observed for electrical conductivity (EC): (1) the EC increased with leaching time at SAR pH = 2.0; and (2) the EC decreased with leaching time when the SAR pH was ≥ 4. The changes in EC patterns could be primarily attributed to the concentrations of Al and Fe in the effluent.

Two distinct leaching patterns also were observed for Al. For SAR pH levels from 4.0 to 6.5, the concentration of Al in the effluent decreased with time. The opposite results were obtained for pH levels from 2.0 to 3.0, where the concentration of Al in the effluent increased with leaching time. There were very good linear correlations between the effluent Al concentration and the effluent pH at SAR pH levels of 2.0 and 3.0. The results suggest that impact of SAR on soil Al released was profound.

Further study is warranted to investigate the rainwater and soil oxidation-reduction potential, soil mineralogy, ionic equilibrium, and elements speciation on trace metals and Al leaching in Latosol under the influence of acid rain.

Acknowledgments

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