Quantifying the release of acidity and metals arising from drainage of acid sulfate soils in the Kelantan Plains, Malaysia

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Abstract

Purpose. This paper discusses the release of acidity and toxic metals from acid sulfate soils in the Kelantan plains due to the oxidation of pyrite.

Methods. A study was conducted in the field and laboratory using leaching column to quantify the release of acidity and metals from these acid sulfate soils.

Findings. The highest pyrite content of 6.3% was recorded in the subsoil in the south region. Titrable actual acidity was significantly high (23.2 cmol H^+/kg) in soils of the southern part where jarosite occurred in the topsoil. Most of the acidity has been released at the beginning of the laboratory leaching experiment, indicating that in the real field situation, the oxidation of pyrite occurred immediately after the area was drained

Originality. The depth at which pyrite layer occurs in the soils has important implication on land quality and crop production in the area. High amount of Al (0.36-0.81 $cmol_c$/kg) and Fe (0.35-0.7 $cmol_c$/kg) were found in the leachates.

Practical implications. Opening an area of acid sulfate soils in the Kelantan Plains for agriculture had oxidized pyrite in the sediments, resulting in the release of high acidity and toxic level of Al, Fe and trace elements into the soils and waterways, which can affect crops and aquatic life. This is made worse by the release of As, B, Cu, Mn and Zn into the soil solution and waterways.

Keywords: acid sulfate soils, jarosite, pyrite, soil acidity, toxic metal, trace metals

1. Introduction

Coastal sediments rich in sulfide contents cover large areas of the beach regions throughout the world. These sediments would form acid sulfate soils upon reaction with seawater in the presence of organic matter with the help of living microorganisms. Acid sulfate soils occupy almost 50 million ha worldwide, including Southeast Asia, Australia, West Africa and Scandinavia [1]-[3]. In Malaysia, this kind of soils was estimated to cover about 0.5 million ha; they are mostly found in the Malay Peninsula [4], [5].

When coastal areas are drained for agriculture or otherwise, severe acidification will occur due to the oxidation of pyrite present in the sediments. This process normally releases considerable amount of acidity and subsequently harmful metals will be made available, such as Al and Fe [3], [6]. The high amount of acidity and Al and/or Fe, accompanied by the presence of trace metals will affect the soils and the surrounding environment [7]-[9]. Natural phenomena, drainage programmes and land development are among the usual processes known to be the main causes of the worsening

condition in areas occupied by acid sulfate soils throughout the world, especially in Southeast Asia [3].

The released acids and large amounts of toxic soluble ions presumably affect plant growth as well as the aquatic biosphere surrounding the soil systems [5], [10], [11]. Such conditions will affect agricultural activities as well as the livelihood of the farming communities in the areas. The surrounding environment and water sources might be polluted with these soluble ions; hence, agricultural production could not be sustained.

Occasionally, pyrite crystals are contaminated with trace elements (e.g. Ni, Co, Cu, Zn, Pb and As) due the substitution of Fe by these elements [12]. High concentrations of water-soluble As, Cd, Cu, Mo, Ni, Pb and Zn were found in the pyritic soils of the Bangkok Plains [13]. Research on the release of acidity and potentially toxic trace elements from drained acid sulfate soils in Malaysia is rather uncommon. As such, for proper management of acid sulfate soils in the country and to improve their productivity, the release of acidity, Al, Fe and the associated water-soluble trace metals as well the

anions arising from pyrite weathering should be determined. Thus, a study was carried out on the acid sulfate soils in the Kelantan Plains, Malaysia, to determine the release of acidity, Al and/or Fe and potentially toxic trace elements that cause environmental pollution and/or inhibition of crop growth.

2. Materials and methods

2.1. Site information

North eastern Peninsular Malaysia is well known to have experienced heavy rainfall during monsoon times, especially in November to January each year. During these periods, heavy rain floods almost all rivers in Kelantan Plains and many other parts of the state. The mean annual precipitation in the plains ranged from 2000 to 2500 mm in 2010-2012. The annual rainfall was 2306 mm in 2012, which was recorded at Kota Bharu Meteorological Station and 2250 mm recorded at Gong Kedak Meteorological Station [14]. Almost all rainfall (> 90%) is concentrated in the rainy season between August and November of each year. During this rainy season, river water sometimes overflows and often the people there are placed under flood warning situation. The average river water levels reach the danger alert state, which is approximately 2 and 1.5 m higher than that of the normal water level at the river gauge station of the Kelantan river near Kota Bharu and Semerak river (in Pasir Puteh), respectively [15].

The acid sulfate soils for this study were located in the Kelantan Plains, Peninsular Malaysia (Fig. 1). The area is adjacent to sandy soils formed on beach ridges studied by [16]. The formation of both soils is associated with the sea level rise during the Holocene. There were three sampling areas chosen within the Kemasin-Semerak IADP (Fig. 1), based on the occurrence of pyrite within the soil profiles; these areas were in the northern part (Bachok), forest reserve area called Saliran Gelam Utama (SGU) which was situated in the middle of the plain and Tok Bali area, in the district of Pasir Putih (in the southernmost of the plain). Special drainage canals were constructed within SGU to get rid of excess water during rainy season. Based on the earlier study [17], it was found that the soils in Bachok area have pyritic layer > 1 meter below the mineral soil surface.
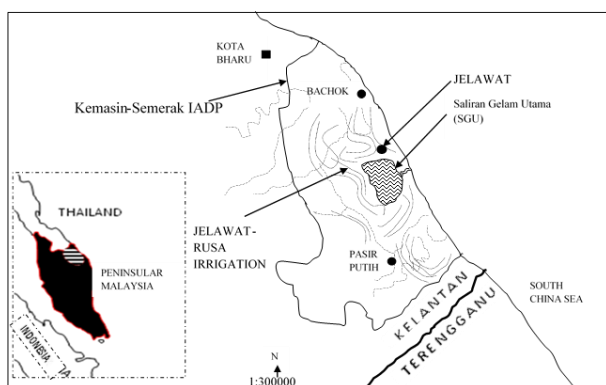

Figure 1. The sites of the studied in the Kelantan Plains, Malaysia

Some thirty years ago, the area under study was constantly flooded during the monsoon months, leading to the government decision to set up a drainage project in order to prepare the site for agricultural production, aiming to alleviate poverty among the farmers. However, the agricultural productivity of the area was low due to serious soil acidity problems arising from drainage; thus, a large portion of the drained land was abandoned. Furthermore, some parts of the pyrite-bearing sediments were overlain by peaty materials (SGU), which had added more problems to the already acidic soil condition. Soils in this part of the Kemasin-Semerak IADP (Fig. 1) were regarded as peat soils having sulfidic materials (pyrite); hence, they are qualified to be classified as acid sulfate soils. About 30 water samples were collected from the drainage canals and waterways across the SGU area. In Tok Bali area which was earlier studied by [18], the soil samples were collected from abandoned paddy fields where the pyritic layer was found to exist in the surface layer.

2.2. Soils and sampling information

All the published soil maps of the area under investigation were studied in detail before conducting the field work. The semi-detailed soil map of Kemasin-Semerak IADP produced by [19] was referred to during the soil sampling. For ease of movement, a four-wheel drive vehicle was used to comb the areas suspected of containing soils with pyrite in the Kemasin-Semerak IADP. Generally, it was found that pyrite occurred at varying depths (close to the surface, below 2 m and in between the two). Additionally, the soils with pyrite at the study sites showed different degree of association with peaty materials (organic matter). Soil profiles were dug and were described at each site (Table 1). At selected locations, soil samples were taken at 0-15, 15-30, 30-45 and 45-60 cm depths using an auger. Altogether, there were 109 soil samples taken from several sampling periods and represents all the three sites. These soil samples were subjected to determination of soil chemical properties (Table 2). In Jelawat, the depth of the peaty materials and sulfuric horizon was determined and recorded. The samples were placed in plastic bags and were sealed before transporting them to the soil laboratory at Universiti Putra Malaysia, Serdang. Most of the soil samples were air-dried, pounded and sieved using 2 mm sieve.

To assess the effects of acid sulfate soils drainage, the drained areas in the middle of the plain called Saliran Gelam Utama (SGU) near Jelawat-Rusa Irrigation Scheme (JRIS) were compared with those undrained acid sulfate soils in Bachok as well as in Tok Bali which was situated near Pasir Putih in the southern part of the Kelantan Plains. About 30 water samples were collected from the drainage canals and waterways across the SGU area. The location of each soil and water sampling site was recorded using a Geographical Positioning System (GPS) as shown in soil maps. The water samples were stored in plastic bottles before transferring to the laboratory at Univesiti Putra Malaysia at Serdang. Before analysing the water samples was filtered using 0.25 µm membrane filters. In Tok Bali area which was earlier studied by [18], the soil samples were collected from abandoned paddy fields where the pyritic layer was found to exist in the surface layer.

2.3. Column leaching experiment

A leaching experiment using soil column was setup in the laboratory using the soil samples taken from the three sites. A solution of artificial rainwater was prepared with major cations and anions composition with volume similar to that of typical Malaysian rainfall data derived from Malaysian Meteorological Department, but without organic components or particulates.

Table 1. Profile description and morphological characteristics of the soils

Sites/ Horizons	Depth, cm	Colour	Texture [1]	Structure [2] (grade, size, type)	Consistency [3] (moist, dry) (moist, dry)	Boundary [4]	Remarks
Bachok							
Typic Sulfaquent							
A	0-15	10YR5/6	C	1, c, gr	w, fr	asb	Abundant coarse sands, very few fine roots
B	15-55	10YR5/8	C	2, m, sb	lw, fr	asb	Few fine roots, few clay coatings, few fine pores
Bw	55-150	10YR5/1	ZC	3, c, ms	fm, s, h	gsb	Very few fine roots, few yellowish nodules
Cg	>150	10YR5/2	ZC	3, c, ms	fm, s, h	cont	Abundant bluish grey nodules (5BG 6/1)
Jelawat							
Typic Sulfihemists							
Oe	0-20	2.5YR3/2	LP	1, f, gr	w, vfr	csb	Many medium-fine roots, very few fine pores
A	20-30	2.5YR3/3	LP	1, f, gr	w, vfr	csb	Many medium-fine roots, few fine pores
Bwg	30-45	10YR3/3	ZC	3, c, ms	fm, s, h	gsb	Few fine roots, few yellowish nodules
Cg	45-60	10YR3/3	ZCL	3, c, ms	fm, s, h	csb	Abundant bluish grey nodules (5BG 6/1)
Pasir Putih							
Typic Sulfaquept							
Ap	0-10	10YR6/1	C	3, m, ps	fm, s, h	asb	Rotten egg smells, 2.5Y 8/6 mottles, few fine roots
Bj	10-35	10YR5/1	C	3, m, ps	fm, s, h	cwb	Yellowish mottles (2.5Y 8/6), few fine roots
Bw	35-45	10YR3/3	C	3, c, ms	fm, s, h	gsb	Few fine roots, few clay coatings, few fine pores
Cg	45-75	10YR3/3	C	3, c, ms	fm, s, h	csb	Abundant bluish grey nodules (5BG 6/1)

(1) C – clay; ZC – silty clay; ZCL – silty clay loam; LP – loamy peat;

(2) grade: 1 – weak; 2 – moderate; 3 – strong; size: f – fine; m – medium; c – coarse; type: gr – granular; sb – subangular blocky; ps – prismatic; ms: massive;

(3) moist: w – weak; lw – lightly weak; fm – firm; s – sticky; dry: fr – friable; vfr – very friable; h – hard;

(4) asb – abrupt smooth boundary; gsb – gradual smooth boundary; csb – clear smooth boundary; cwb – clear wavy boundary; cont – continuous.

Table 2. Chemical properties of the soils in the studied areas

Sites/ Horizons	Depth, cm	pH	EC, dS/m	Exc. Al, cmol+/kg	K	Ca	Mg	CEC	Ext. Fe	Avail. P, mg/kg	Total N	O.C.	Total S	Sand	Silt	Clay
Bachok																
Sulfic Endoaquept																
A	0-15	4.9	0.14	7.9	1.23	2.97	1.67	10.0	2.3	17.6	0.3	13	0.1	32.3	12.2	55.3
B	15-55	3.8	0.12	6.0	0.83	4.12	1.97	9.0	1.5	19.1	0.2	14	0.3	31.4	26.3	42.3
Bw	55-150	3.9	0.10	15.7	0.88	7.09	2.15	9.5	11.3	14.2	0.1	33	2.1	9.5	52.1	38.4
Cg	>150	4.2	0.08	10.2	0.96	5.53	2.11	12.4	11.9	75.1	0.2	42	2.3	1.0	57.4	42.1
Jelawat																
Typic Sulfihemists																
Oe	0-20	3.2	0.12	16.2	0.76	0.68	0.42	14.2	10.8	10.0	0.2	44	2.6	n.a	n.a	n.a
A	20-30	3.1	0.17	15.5	0.52	0.45	0.42	11.7	10.4	9.5	0.1	23	3.2	1.6	51.2	47.3
Bwg	30-45	3.1	0.16	13.8	0.56	0.41	0.43	9.3	8.1	10.1	0.8	39	3.1	4.9	58.7	36.4
Cg	45-60	3.3	0.18	15.2	0.48	0.50	0.52	17.6	9.1	10.3	0.4	46	3.5	n.a	n.a	n.a
Pasir Putih																
Typic Sulfaquept																
Ap	0-10	4.0	0.34	8.1	1.35	0.71	0.54	20.4	10.2	14.4	0.4	39	3.1	3.7	20.5	75.8
Bj	10-35	3.5	0.42	6.2	1.37	0.68	0.7	14.3	10 .1	14.6	0.2	35	3.2	4.0	21.7	74.3
Bw	35-45	3.1	0.14	15.7	1.06	1.37	1.66	20.1	10.5	17.4	0.5	13	3.2	0.7	25.8	73.5
Cg	45-75	2.8	0.42	32.1	0.83	2.88	2.05	10.7	10.3	17.0	0.3	15	1.7	n.a	n.a	n.a

*EC – electrical conductivity; CEC – cation exchange capacity; Exc. – exchangeable; Ext. – extractable; Avail. – available; O.C. – organic carbon

The solution was prepared by combining various stock standard solutions by the method of [20]. The prepared artificial rainwater solution was then diluted to a final volume of 50 L with distilled water and acidifying with hydrochloric acid. The composition of the rainwater (μmol/l) was Ca^{2+}, 8.29; $Cl^−$, 36.38; Mg^{2+}, 4.28; NH_4^+, 2.08; $NO_3^−$, 4.09; K^+, 13.17; SO_4^{2-}, 2.84 and Na^+, 28.06 respectively [14]. The pH and electrical conductivity (EC) of the artificial rainwater were 5.9 and 0.28 dS/m, respectively. Each leaching experiment was performed on duplicate samples of each of the soil

layers in all three soil profiles packed into polypropylene columns (10 cm diameter, 10 cm depth). The bottom of each column was plugged with plastic mesh to aid drainage and a filter-paper was placed on top of the soil to assist in the distribution of the leaching solution over the surface. The columns were covered with polyethylene sheeting to prevent contamination during the course of the experiments. Five hundred g of air-dried soil sample from each layer was placed and a volume of 100 ml per day of artificial rainwater was applied manually to each column, for 12 periods each of 5 days. This corresponds approximately to the application of average 1 month's rainfall per 5 days' period. Leachate was collected in amber-coloured polypropylene bottles and pooled for analysis every 5 days. After filtering through a 0.2 μm membrane filter (Omnipore membrane; Nihon Milipore, Tokyo, Japan), the volume, redox potential, pH and conductivity of the leachates were measured. A portion was stored for the determination of SO_4^{2-}, Cl^- and NO^{3-} by ion chromatography and the remainder was acidified to 1% HNO_3 and stored at 4°C prior to determination of Zn, Cu, Mn, Pb and As.

2.4. Laboratory analyses

2.4.1. Chemical analyses

Soil pH was determined in distilled water using soil to solution ratio of 1:2.5. pH KCl was determined after extracting the soil with 1:40 1M KCl solution. Electrical Conductivity (EC) was carried out using saturated paste. Cation Exchange Capacity (CEC) was determined by 1 M NH_4OAc solution buffered at pH 7 [21]. Exchangeable Al was extracted by 1 M KCl and the Al in the solution was determined by AAS. Total carbon (TC) was analyzed by the dry combustion method using a CNS analyzer. Total N was determined by the Kjeldahl method and available P was extracted with a mixture of HCl and NH_4F [22]. Titratable actual acidity (TAA) was determined by the method of [23]. This was done by titration of 1M KCl extracted soil solution to pH 6.5 using standardized 0.5 M NaOH solutions. Total S was determined by combustion method using CNS analyser and pyrite content was determined by the method of [24]. The pyrite content was calculated based on the difference between pyritic-Fe and non-pyritic Fe in the soil samples after digestion. About 2 g of soils were digested with 6 ml concentrated HCl before being left overnight after which 23 ml distilled water were added for both pyritic and non-pyritic Fe determination. The next day, samples for non-pyritic Fe were boiled for about an hour before the tube was made up to 75 ml with 1:10 HCl, while the other samples were added with 25 ml of 3N HNO_3 and were made up to 75 ml by adding 2 N HNO_3 while boiling for 30 minutes. Both sets of samples were filtered with Whatman No. 2 filter paper before subjecting to Fe measurement by ICP-AES.

Chloride (Cl^-), nitrate (NO^{3-}) and sulfate (SO_4^{2-}) ions in the leachates and the drainage water samples were measured using ion chromatography (882 Compact IC Plus 1); Methorhom, Switzerland). The eluent was 3.2 mmol/L Na_2CO_3; 1.0 mmol/L $NaHCO_3$ at a flow rate of 0.6 mL/min. Calibrant solutions for ion chromatography were prepared by dissolution of sodium chloride, sodium nitrate and sodium sulphate of analytical reagents grade from Merck. Aluminium, Fe, As, Cd, Cr, Cu, Mn, Ni, Zn and Pb in the leachates and water samples were determined by ICP. Calibrant solutions

for ICP were prepared by serial dilution of 1000 mg Spectrosol solutions of the appropriate elements (Merck, Lutterworth, UK) with the artificial rainwater. The geochemical speciation for the drainage water was carried out using GEOCHEM-EZ software [25].

2.4.2. Statistical analysis

Significant differences in the mean values of leachates parameters between the three sites were evaluated by one-way ANOVA for repeated measurement using Microsoft Excel Stat for Windows. The relationships of trace metals and properties of leachate were evaluated by Spearman Rank correlation.

3. Results and discussion

3.1. Pyrite content and acidity in the soils

Results of the study showed that the soil samples in Bachok area had pHKCl ranging from 3.2 to 4.5 (Table 3).

Table 3. Pyrite content and acidity status of the soils in the studied area

Site	Depth, cm	*pH$_{KCl}$	Pyrite, %	*TAA, cmol H^+/kg	Total S, %
Bachok	0-15 (sandy mineral soil)	4.5	0.09	2.8	0.1
(Undrained)	15-55 (sandy mineral soil)	4.2	0.00	6.4	0.3
	55-150 (clay soil)	3.3	1.71	8.8	2.1
	> 150 (clay soil)	3.2	0.81	9.3	2.3
*SGU	0-20 (peat soil)	3.2	0.96	16.0	2.6
(Drained)	20-30 (peaty mineral soil)	3.1	1.62	14.9	3.2
	30-45 (clay soil)	3.1	3.08	17.7	3.1
	45-60 (clay soil)	3.3	2.25	17.5	3.5
Tok Bali	0-10 (clay soil)	3.3	1.39	23.2	3.1
(Undrained)	10-35 (clay soil)	2.9	6.31	22.5	3.2
	35-45 (clay soil)	3.3	1.01	8.8	3.2
	45-75 (clay soil)	3.5	1.21	8.2	1.7

*SGU – Saliran Gelam Utama; TAA – Titratable actual acidity; pH$_{KCl}$ – pH in 1M KCl

The pH was lower in the subsoil compared to that of the topsoil, which was consistent with the increasing total actual acidity (TAA) down the soil profiles. The TAA increased from 2.8 to 9.3 from topsoil to the bottom, showing an increasing trend with depth. It was observed that pyrite was mostly concentrated in the subsoil with value between 0.8 and 1.7% and hence total S (mostly from pyrite) was mainly present in that zone, with values ranging from 2.1 to 2.3%. The topsoil in Bachok area did not contain any pyrite or jarosite mottles in the upper 100 cm depth. Pyrite was only found to occur below this depth. The topsoil was rather sandy with clay content increasing with depth (Table 2). It is believed that the continuous deposition of the fluvial materials on the top of the pyrite-bearing sediments during flood seasons since time immemorial had resulted in the current

morphological features of the soil profiles. Agriculturally, this area can sustain crop production as the pyritic layer is always below the water table, except during extreme dry condition which seldom occurs in the area.

In the SGU area, the topsoil contained peaty materials and at some sampling points, they were mixed with clayey materials. This area was mapped as peat soils [26] and under the Malaysian System of Soil Classification it can be named as Nipis Series [27]. Based on our data, the soils can be tax-onomically classified as Typic Sulfihemists [28]. We found that the pHKCl of the soils in the SGU ranged from 3.1 to 3.3, indicating extreme acidic condition throughout the soil profiles. This acidity might have been generated by the peaty materials in combination with pyrite oxidation during the dry months of the year when the water table dropped. During the survey and soil sampling, pyrite was found to be abundant in the subsoil, starting from 20 to 45 cm depth. The pyrite per-centage was very high in these sulfidic horizons, especially at the 30-45 cm depth, with value exceeding 3% (Table 3).

The topsoil in Tok Bali area contained yellowish jarosite mottles and the soil emitted bad odour during the sampling time. This phenomenon had already been mentioned in the paper written by [18], as the sign of pyrite disintegration into jarosite. Determination of the pyrite content showed that the top 10 cm of the soil profile contained as much as 1.4% pyrite and this amount increased up to 6.3% at 35 cm depth (Table 3).

The lower content of pyrite in the top-compared to that of the subsoil was due to its oxidation on exposure to the atmos-pheric conditions. The values obtained by this study are con-sistent with those of the acid sulfate soils elsewhere in the country as reported by [4]. Because of the presence of extreme acidity and too much Al and/or Fe in the soils, rice growth was seriously affected. Rice grows healthily if water in the field has pH of about 6 and Al and/or Fe concentration of < 20 μM [3]. The acid sulfate soils in Tok Bali area cannot be used for sus-tainable rice cultivation without proper ameliorative program; hence, currently most of the rice field are abandoned.

3.2. pH, EC and redox potential of the leachates

The leachates pH showed increasing trend from 12 batch-es collections throughout the experiment for all the soil lay-ers (Fig. 2). The pHs increased gradually from 30 cm depth downwards, while for the above two layers, the pHs in-creased abruptly starting from 4.1 to 4.6 at 4th batch and 3.8 to 4.4 at 6th batch for the second and first layer, respectively. The high amount of organic matter in the peat layer could have supplied bonding sites for Al^{3+} and H^+ in the soils up to these times, after which the pH increased quickly due to lower rate of leaching of these acidic species after a series of extreme flushing during the first 4 batches. However, the organic topsoil tended to show lower trend of pH values across the leaching experiment even after the pH increased at batch 6 as compared to the second soil layer which slowly joined the other two layers after a sharp pH increase at batch 4. The plausible reason could be the organic acids contribu-tion towards the persistent acidity in the layer. Electrical conductivity (EC) of the leachates from all soil layers de-creased from one leaching batch to another, indicating solu-ble ions that were leached out. The EC ranged from 0.31 to 1.22 dS m^{-1} and the similar changes occurred for pH during the first six collections for both two upper layers. Redox potential of the leachate ranged from 234 to 372 mV, which was consistent with the findings of [29].
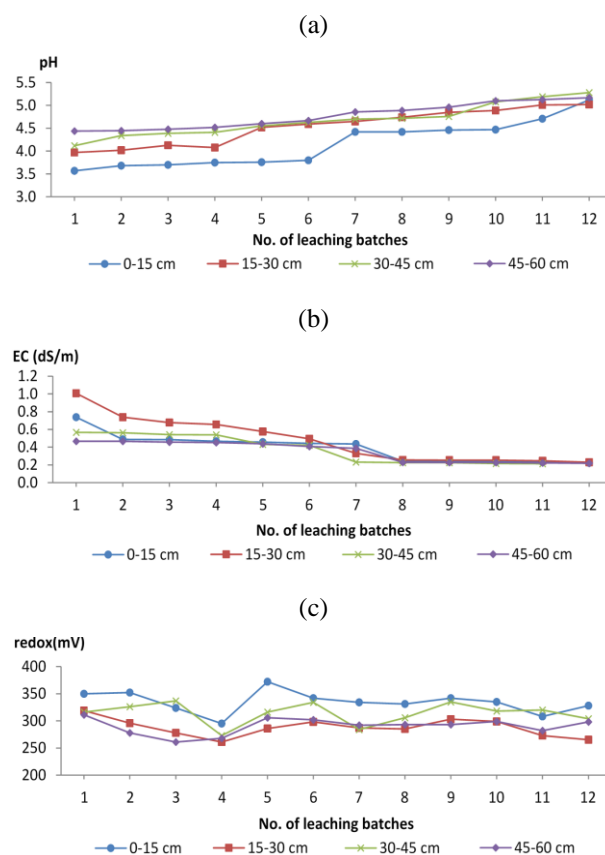

Figure 2. Properties of the leachates: (a) pH; (b) EC; (c) redox potential

3.3. The concentration of Al and Fe in the leachates

In Figure 3, the downward arrow denotes the number of leaching batches carried out for each sample. The mean con-centrations of Al and Fe were 0.36 and 0.51, 0.81 and 0.70, 0.35 and 0.49 cmolc kg^{-1} for soils in Bachok area, SGU and Tok Bali, respectively. Generally, the concentration of Al and Fe decreased from the first leachate collection to the end of experiment. The Al concentration ranged from 0.06 to 0.98, 0.34 to 1.24 and 0.11 to 1.08 cmolc·kg^{-1} in Bachok, SGU and Tok Bali, respectively, while the Fe concentration ranged from 0.11 to 1.08, 0.12 to 1.8 and 0.05 to 1.88 cmolc·kg^{-1}. These wide ranges between leached amount of Al and Fe were particularly an effect of different soil textural classes that present in the soil profiles. The Al and Fe in the samples from SGU showed significantly ($p < 0.05$) higher concentration in all horizons as compared to the other two sites and t-test showed the concentration of Al and Fe was in the order of SGU > Tok Bali = Bachok. The amount of Al and Fe leached out from the upper two layers of SGU showed increasing trend from early to mid-leaching period and then dropped down towards the end, but the leached amount were still high even though the leaching study ap-proached the end. Similar leaching trends of Al and Fe were found from the earlier studies on acid sulafte soils in Thai-land [29] and Indonesia [30]. On average, the amount of Al and Fe that have been leached out from this study were 5 and 8%, 6 and 7.3%, 3 and 5% from the total concentrations present in the soils of Bachok, SGU and Tok Bali, respec-tively. However, these amount of leached Al and Fe were calculated from cumulative 60 days' time of leaching and taken from only 500 g of soil samples from each soil layer.
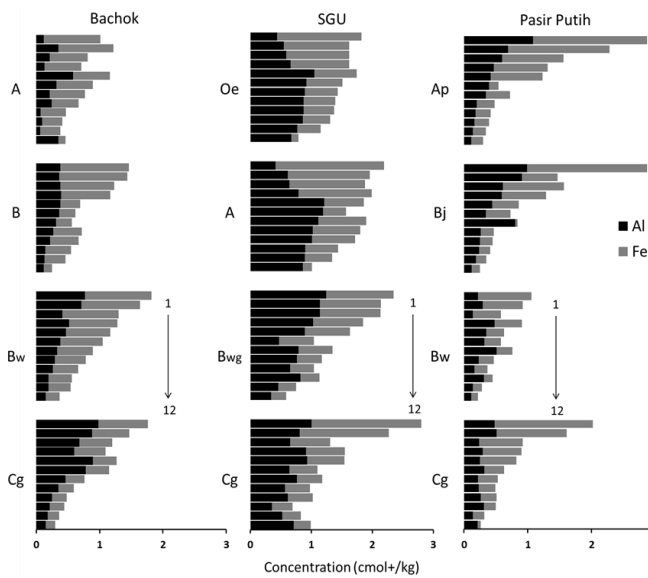
Figure 3. Concentration of Al and Fe in the leachates

Hence, the field leaching activities could be assumed to happen to a greater extent, due to extensive wet-dry conditions and higher precipitation. Thus, this leaching percentage was somehow showing an insight of the leaching phenome-non in the field and could be a reference value for future management strategies.

The pyrite concentration was higher in the soils of Tok Bali, whereas the leaching of Al and Fe was found to be higher in SGU. This was due to the drainage programmes in SGU where the pyritic topsoil was exposed to oxidation. Then, the leaching would easily flush out the Al and Fe from the soil colloids mixed with peat soils. Similar results were observed by [31]; the organic acids from peat and acidic chemical species from acid sulfate soils might have reacted with the organic matter as well as soil minerals to release Al and Fe, particularly from tropical areas where conditions were favourable for weathering.

3.4. The concentration of anions in the leachates

This study found that SO_4^{2-} ions were leached abundantly from the soils from the three sites, especially from the sulfidic layers with different leaching patterns (Fig. 4). Soils from Bachok area showed an increasing amount of leached SO_4^{2-} and NO_3^- going down the soil profile, with an average of 50.3 and 22.2 mg/l of SO_4^{2-} and NO_3^-, respectively. The highest leached amount of SO_4^{2-} was collected from layer 4 with maximum value of 103.9 mg/l and the concentration decreased over time up to the last batch where the leached amount was equal to the amount added in the ARW.
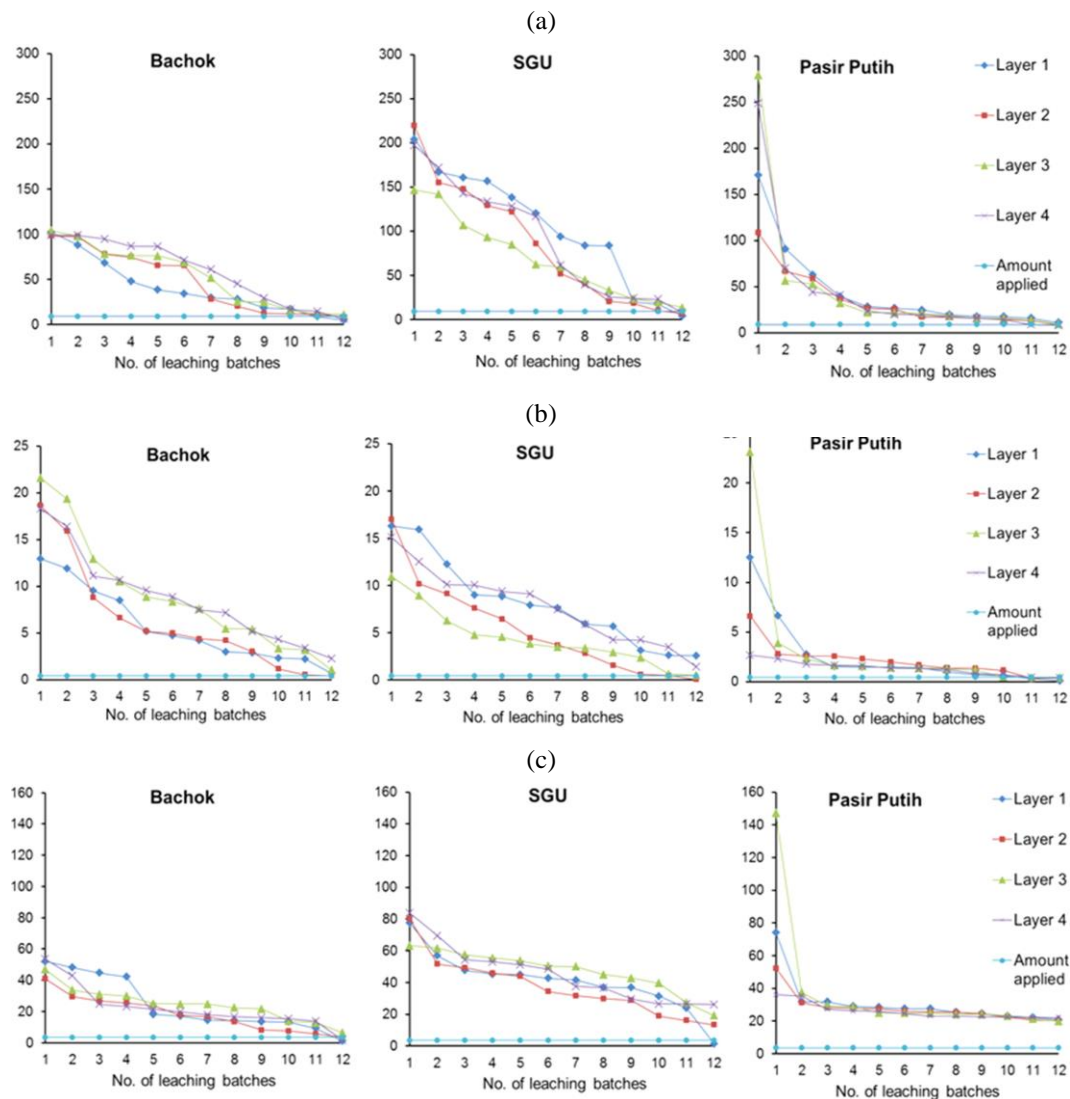

Figure 4. Concentration of anions in the leachates (mg/l): (a) SO_4^{2-}; (b) NO_3^-; (c) Cl^-

Soil layers from SGU released significantly higher amount of SO_4^{2-} ($p < 0.05$) than the other two profiles and the surface organic rich soil mixed with pyrite dominated the leachates throughout the twelve 5-days leaching collections, with an average of 86.6 mg/l of SO_4^{2-} and ranged between 13 and 204 mg/l. Soils in Tok Bali tended to release relatively high amount of SO_4^{2-}, Cl^- and NO^{3-} ions at the beginning of leaching and then the amount leached started to remain constant from the fifth batch towards the end. The third and bottom layer of Tok Bali soils released 279 and 248 mg/l of SO_4^{2-} during the first leachate collection, followed by a sharp drop in the second batch.

The SO_4^{2-} concentrations in the leachate tended to follow the concentration and position of pyrite in the soil profiles. For SGU and Tok Bali soils, pyrite was concentrated in the upper horizons, where the concentrations of SO_4^{2-} were high. On the other hand, the condition was the opposite for the soils in Bachok area. The disintegration of pyrite in the surface layers of SGU and Tok Bali soils released higher amount of SO_4^{2-} due to pyrite oxidation. This oxidation; however, did not occur in the pyritic bottom horizons of Tok Bali soils. Hence, the SO_4^{2-} ions were in negligible amounts.

3.5. Trace metals in the leachates

Besides acidic and anionic species, the oxidation of pyrite and the subsequent reactions of the products with minerals in the soils would release significant amount of trace metals into the environment. For the current study, the concentrations and correlation of metals in leachate samples with pH and their ratio according to the Malaysian standard of groundwater quality set by Malaysian Ministry of Health are shown in Table 4. The concentrations of Al, Cr, Cu, Fe, Mn and Zn showed significantly ($p = 0.05$) strong negative correlation with pH. Big ranges with more than a hundred folds difference were observed in the concentrations of Al, As, Fe, Mn, Ni, Zn and NO^{3-}. All elements measured were within the range of Malaysian standard of Ground Water Quality, except for the concentration of Al, As, Fe, Mn and EC of the leachates that exceeded the benchmark level by 17.6, 1.2, 38.5, 1.6 and 40 folds, respectively. According to [32], increase in heavy metal concentrations often created health risks for aquatic environments because of toxicity, bioaccumulation and persistence of these metals. This is exactly the situations in the SGU area, where no fishes or any aquatic organisms were observed in the rivers and waterways during soil/water samplings.

Table 4. Leachate properties of the soils in the Kelantan Plains

Element	KP Concentration (mg/l)			CorrS(pH)	MS[*]	KP median/MS
	median	max	min			
Al	3523.6	16490	65.1	-0.76	200	17.6
As	11.7	31	0.1	-0.44	10	1.2
B	113.5	618	9.9	-0.69	500	0.2
Cd	0.6	2	0.2	-0.59	3	0.2
Cr	0.6	3	0.1	-0.86	50	0.1
Cu	10.4	49	0.6	-0.82	1000	0.01
Fe	11541.3	94200	500	-0.94	300	38.5
Mn	162.5	291	2.3	-0.82	100	1.6
Ni	1.7	17	0.1	-0.07	20	0.09
Pb	3.2	18	0.3	-0.70	10	0.3
Zn	33.5	220	0.7	-0.92	3000	0.01
Nitrate	4213.8	32970.3	203.3	-0.03	10000	0.4
Sulfate	50775.9	200895	5780.6	-0.08	250000	0.2
Chloride	29246.8	94874.1	7402	0.30	250000	0.1
EC (dS/m)	0.4	1.01	0.21	0.73	0.01	40
pH	3.8	5.2	3.2		6.5-9.0	

[*]MS – Malaysian standard based on National Groundwater Quality Standard (KKM, 2000); KP – Kelantan Plains

The concentration of As in the leachates for the soils of Bachok showed the same trend as that of the leaching of Mn where the highest release was observed from the lowermost zone of the profile and it decreased upwards (Fig. 5). The mean concentrations of leached As and Mn in Bachok from the lowest horizon to the topsoil were 17.6, 13.4, 11.13, 6.9 and 65, 45.4, 24 and 20.6 µg/l, respectively. This was consistent with the soil textural class, where the topsoil was rather sandy, while the bottom layers were more clayey. Thus, leaching might have occurred extensively in the field especially for the sandy topsoil. For the soils in the SGU area, the concentration of leached As was higher in the subsoil (layer 2 and layer 3) compared to that of the topsoil. The organic topsoil (layer 1) had the lowest As concentration with mean value of 9.05 µg/l. This possibly was attributed to the low prevailing As concentration in the organic soils, or its strong binding with the solid phase of the organic matter as suggested by [33] and [34]. Generally, the leached concentrations of As from the soils in Bachok, SGU and Tok Bali were

about 3.2, 7 and 5%, respectively. The highest percentage of As leaching from SGU was particularly related to the higher amount of pyrite within most layers of the soils (Table 3) as well as due to the drainage programmes in that areas. Based on these leaching patterns, it was reasonable to say that there would be lower risk of As leaching from the peaty acid sulfate soils as compared to those acid sulfate soils without peat.

The leaching of Mn, however, did not show any trend where its respective mean concentration with depth was of 49.6, 53.4, 54.0 and 30.3 µg/l, from the top to the lowest layer. The leachates for the soils from Tok Bali followed the same pattern as that of As and Mn released by the soils from SGU area, although the concentration dropped drastically after batch 6 and batch 2, respectively. Note that the acid sulfate soils in SGU and Tok Bali areas were clayey in nature. We believed that these fine-grained sediments could be significant binding sites for the trace metals. This notion is in line with the findings of [35] who discovered that clay particles had strong relation with the soil metals like As and Mn.
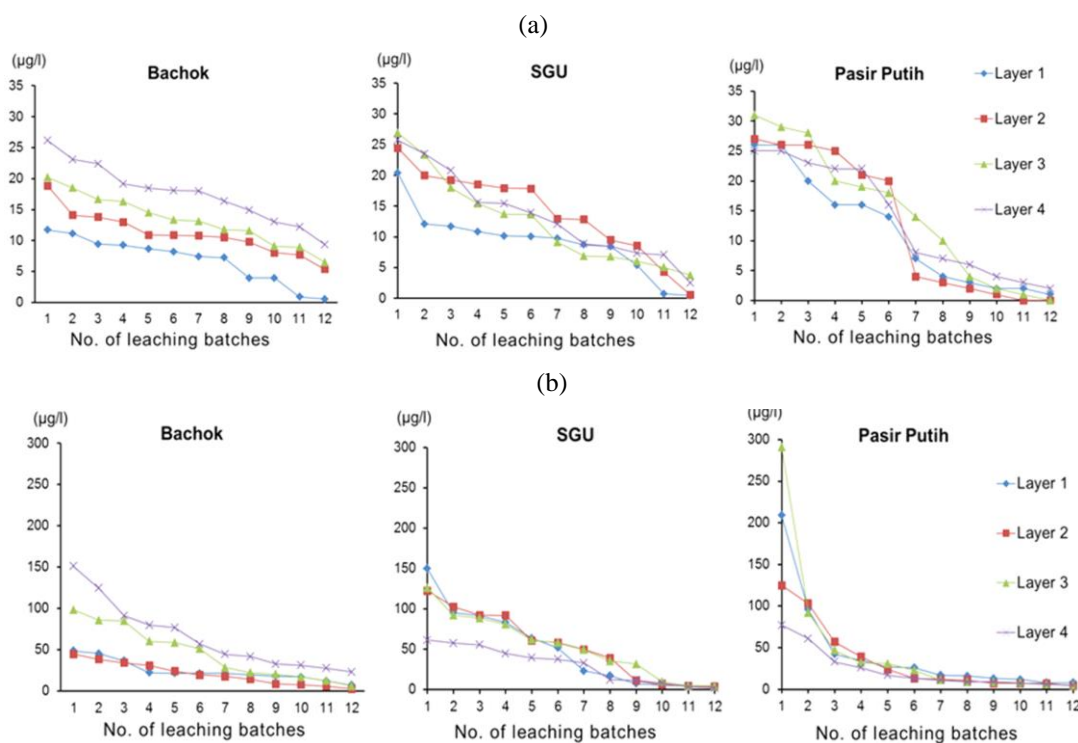
(a)

(b)

Figure 5. The concentration of metals in the leachates: (a) As; (b) Mn

3.6. The chemical properties of the waterways

The water samples and ionic species data were compared with those of the National Water Quality Standard (NWQS) for surface water set by Department of Environment, Malaysia [36] (Table 5). The pH and EC of the water samples were within the range of the standard set for surface water class II, III and IV. However, the chemical species in the water were high enough that it can no longer be classified as water sources class II and III, which can be treated for domestic uses. High concentration of Al and Fe in the water together with the extremely low pH made the waterways unsuitable for any aquatic organisms. The only living things found in the waterways were the acid tolerant plant species, like Eleocharis dulcis and "benta" grass (Fig. 6).

Based on these concentrations of metals and anions, the water quality falls into class V, where it cannot be used, either for domestic use after treatment or even for irrigation purposes.

Table 5. Ionic species and trace metal contents in the waterways of the acid sulphate soils

Samples	pH	EC, dS/m	Al, mg/l	Fe, mg/l	Mn, mg/l	Zn, mg/l	As, mg/l	NO_3^-, mg/l	SO_4^{2-}, mg/l	Water class
W1	5.6	0.7	18.2	23	0.7	0.5	0.2	4.5	22	Class V
W2	5.2	0.8	9.5	19	0.2	1.2	0.5	11.3	8.1	Class V
W3	5.2	1.4	25.3	21	0.2	0.9	0.2	3.4	15.2	Class V
MS[a]	6-9	1.0	0.05	1	0.1	5.0	0.05	7.0	250	
MS[b]	5-9	1.0	0.05	1	0.1	0.4	0.05	0.0	0	
MS[c]	5-9	6.0	0.5	5	0.2	2.0	0.1	5.0	0	
MS[d]	< 5	> 6	> 0.5	> 5	> 0.2	> 2	> 0.1	> 5	> 0.01	

MS – National Water Quality Standards for surface water (DOE, 2012); a – Class II (need conventional treatment); b – Class III (need extensive treatment); c – Class IV (for irrigation use); d – Class V (none of the above)
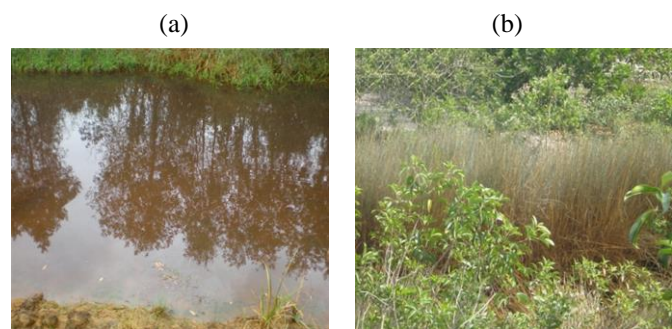
(a) (b)

Figure 6. Iron-stained waterways in the studied area (a); acid-tolerant plant species growing in the drains of the acid sulfate soil area (b)

3.7. Water in the drainage canals of the Kelantan Plains in comparison with others

The chemical properties of the water in the drainage canals in the Kelantan Plains were compared with those of Indonesia and Vietnam (Table 6). Most of the studies in the two countries were concerned with acidic chemical species with a few focused on the release of trace metals. The pH of the water in the drainage canals of the Kelantan Plains was 2.8, which was among the lowest in ASEAN. This very low pH was also reported for the water of the drainage canals in the acid sulfate soils of the Mekong Delta, Vietnam [29]. Consistent with the very low pH, the concentration of Al and Fe as well as EC of the water in the Kelantan Plains was

higher than the other drainage canals compared in this study. This indicates that the properties of the water in the drainage canals of the Kelantan Plains were significantly governed by the acidic species, particularly the Al and/or Fe. Regardless of the high amount of cationic species, the concentration of anions, like SO_4^{2-}, Cl^- and NO^{3-} was comparable to those of the Kalimantan, Indonesia as reported by [37] and [38]. It was found that the As concentration in the water of the canals of the Kelantan Plains was three times higher than that of the Mekong Delta studied by [39]. However, it was not the case for the concentrations of Mn and Zn.

The geochemical speciation model shows that in the drainage canals of the Kelantan Plains, under such low pHs (2.8-4.6), all the elements measured present in water-soluble forms, except for Al which forms some precipitate (53-65%) with OH^- and SO_4^{2-}. In SGU, where drainage canals were subdivided into primary, secondary and tertiary, the amount of dissolved elements correspond well to the dilution and convergence with receiving waterways after drainage canals end. In the primary canals, where the pH was found to be the lowest among all, there was about 46% dissolved Al. From this amount, 18% was in the form of free metal and the rest existed in complexes. While these canals contain rather high dissolved Al content, its activity also may reach up to 55 μM, which is detrimental to most plants and aquatic organisms. In the secondary and tertiary canals, the dissolved Al decreased to 35 and 5%, respectively.

Table 6. Comparison of water in the drainage canals of the studied area with that of the other ASEAN countries

pH	EC, dS/m	Al, mg/l	Fe, mg/l	SO_4^{2-}, mg/l	Cl^-, mg/l	NO_3^-, mg/l	As, mg/l	B, mg/l	Cd, mg/l	Cr, mg/l	Cu, mg/l	Mn, mg/l	Ni, mg/l	Pb, mg/l	Zn, mg/l	Sources
2.8	1.40	13.8	8.2	23	17	3	9.5	7.2	0.4	0.7	10.4	21.5	0.04	1.3	4.3	R[0]
5.6	0.06	0.5	5.8	1.15	3.6	5.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	R[1]
7.5	1.20	3.2	3.6	n.d.	7.0	0.5	3.1	n.d.	0.1	6.1	3.5	73.8	3.1	n.d.	10.2	R[2]
2.8	0.20	10.0	n.d.	10	4.0	0.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	R[3]
3.2	0.40	9.9	1.7	106	45.5	0.6	n.d.	n.d.	n.d.	n.d.	n.d.	0.03	n.d.	n.d.	n.d.	R[4]

n.d. – not determined; R[0] – this study; R[1] – Markus and Djadja, 2013 (Kalimantan, Indonesia); R[2] – Gert-jan wilbers, 2014 (Mekong, Vietnam); R[3] – Kawahigashi, 2012 (Mekong, Vietnam); R[4] – Markus, 2009 (Kalimantan, Indonesia)

3.8. Draining acid sulfate soils and its implication to the environment

The results from this study suggested draining of acid sulfate soils in the Kelantan Plains, particularly the SGU area, is not a good idea. It would certainly cause environmental degradation that affects crop production and aquatic lives. The agricultural lands and waterways had also suffered from existence of high amounts of acidity and Al that curtailed the growth of rice. The only living thing that survived the severe acidity is acid-tolerant plant species (Fig. 6). Due to high amount of Fe released by the oxidation of pyrite, iron staining is very common in the rivers and waterways in the area. The high concentration of Al and Fe in the streams and other water sources leads to rust formation (Fig. 7), affecting structures as well as the people's daily lives.

(a)   (b)

Figure 7. Rust formed on the iron pipe (a); rust formed on the galvanized pipes (b)

The quality of water source in the area under study becomes the most concern aspect of the farming communities in the area. Besides rusts and acidity, the presence of trace metals in the water sources could indirectly affect human health. Arsenic and Mn present in the waterways surrounding the SGU area could contaminate water used for domestic purposes. Drinking water containing high amount of trace metals may cause several health hazards/problems in human, including shortness of breath and various types of cancers [40]. These findings have to be taken into consideration by the powers-that-be if the areas under acid sulfate soils in the Kelantan Plains are going to be opened for development.

4. Conclusions

Acid sulfate soils are common in the coastal plains of Kelantan, Peninsular Malaysia. When the area was developed for agriculture, pyrite in the sediments was oxidized, resulting in the release high acidity and toxic level of Al and/or Fe into the environment. Results of the field study showed that pyrite in the soils of the plain occurred (concentrated) at various depths: Below 1 m in Bachok area (north) and in the topsoil in Tok Bali (south). For the soils in the forest reserve (middle), it was at 20-60 cm depth. The highest pyrite content of 6.3% was recorded in the subsoil of the south region. It is believed that the depth at which pyrite layer occurs in the soils has important implication on the land quality and crop production. Opening the land in the middle and southern parts of the Kelantan Plains for development had been found to result in pyrite oxidation that released high amount of acidity and toxic metals (Al and Fe) as well as trace elements into the soils and waterways, which was confirmed by the results of the leaching experiment in the laboratory. Most of the acidity has been released at the beginning of the leaching experiment, indicating that in the field situation the oxidation of pyrite occurs immediately after the area is drained. High amount of Al and Fe were found in the leachate, consistent with their high concentration in the soils that affect crops and aquatic life. This is made worse by the presence of some trace metals in the soil solution and waterways which are potentially toxic to human being.

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Аналіз виходу кислот і металів в результаті осушення сірчасто-кислотних ґрунтів у районі Келантанських рівнин, Малайзія

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Мета. Визначення виходу кислот і токсичних металів з сірчасто-кислотних ґрунтів у результаті окислення піриту в районі Келантанських рівнин.

Методика. Дослідження проводилися в натурних і лабораторних умовах із використанням колони вилуговування для вимірювання рівня кислотності та присутності металів у сірчасто-кислотних ґрунтах.

Результати. Зафіксовано, що у південному регіоні найбільш високий вміст піриту (6.3%) в шарі підґрунтя. Рівень кислотності був дуже високий (23.2 кмоль H^+/кг) в ґрунтах південного району, де у верхніх шарах ґрунту зустрічається ярозит. Встановлено, що пірит у ґрунтах рівнини концентрується на різних глибинах: нижче 1 м в районі Бачок (північ) і у верхньому шарі в Ток Балі (південь), для ґрунтів в лісовому заповіднику – 20-60 см. Виявлено, що вихід здебільшого кислот спостерігався на початку лабораторного експерименту з вилуговування, що вказувало на те, що в реальних натурних умовах окислення піриту відбувається відразу після осушення території.

Наукова новизна. Експериментально доведено, що якість ґрунту та врожаю на території значною мірою визначаються глибиною залягання піриту в ґрунті. У дренажних водах виявлено високий вміст алюмінію (0.36-0.81 кмоль/кг) і заліза (0.35-0.70 кмоль/кг).

Практична значимість. Використання частини сірчасто-кислотних ґрунтів для сільськогосподарських потреб призвело до закислення відкладень піриту й підвищення кислотності ґрунтів, а також до їх токсичного забруднення, що негативно впливає на врожай, флору і фауну. Отримані показники забруднення ґрунтів дозволять розробити ефективні заходи щодо протидії закислення та токсичного впливу.

Ключові слова: : сірчасто-кислотні ґрунти, ярозит, пірит, кислотність ґрунту, токсичні метали, сліди металів

Анализ выхода кислот и металлов в результате осушения сернисто-кислотных почв в районе Келантанских равнин, Малайзия

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Цель. Определение выхода кислот и токсичных металлов из сернисто-кислотных почв в результате окисления пирита в районе Келантанских равнин.

Методика. Исследование проводились в натурных и лабораторных условиях с использованием колонны выщелачивания для измерения уровня кислотности и присутствия металлов в сернисто-кислотных почвах.

Результаты. Зафиксировано, что в южном регионе наиболее высокое содержание пирита (6.3%) в подпочвенном слое. Уровень кислотности был весьма высок (23.2 кмоль H^+/кг) в почвах южного района, где в верхних слоях почвы встречается ярозит. Установлено, что пирит в почвах равнины концентрируется на разных глубинах: ниже 1 м в районе Бачок (север) и в верхнем слое в Ток Бали (юг), для почв в лесном заповеднике – 20-60 см. Выявлено, что выход большей части кислот наблюдался в начале лабораторного эксперимента по выщелачиванию, что указывало на то, что в реальных натурных условиях окисление пирита происходит сразу после осушения территории.

Научная новизна. Экспериментально доказано, что качество почвы и урожая на территории в значительной мере определяются глубиной залегания пирита в почве. В дренажных водах обнаружено высокое содержание алюминия (0.36-0.81 кмоль/кг) и железа (0.35-0.70 кмоль/кг).

Практическая значимость. Использование части сернисто-кислотных почв для сельскохозяйственных нужд привело к закислению отложений пирита и повышению кислотности почв, а также к их токсическому загрязнению, что негативно влияет на урожай, флору и фауну. Полученные показатели загрязнения почв позволят разработать эффективные мероприятия по противодействию закислению и токсическому воздействию.

Ключевые слова: сернисто-кислотные почвы, ярозит, пирит, кислотность почвы, токсичные металлы, следы металлов

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