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On the Pyritization of the Coastal Sediments in the Malay Peninsula during the Holocene and its Effects on Soil

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ABSTRACT

Much of the present coastal plains in the Malay Peninsula were inundated by seawater some 6,000 years ago. That was the time when pyrite is believed to have been mineralized in the sediments of the seawater. This paper attempts to explain the process of pyrite mineralization in the coastal sediments during the Holocene as well as to show how pyrite oxidation affects plants and aquatic life in their vicinity. At that point in time, the sea level was 3-5 meters above the present level. Under reduced conditions, Fe^{3+} ions existing in the sediments were reduced to Fe^{2+} ions, while SO_4^{2-} anions from seawater were reduced to S^{2-} ions. These reactions were promoted by microorganisms feeding on the organic matter provided by native vegetation. Finally, the ferrous and polysulfide ions reacted to form pyrite (FeS_2). Over the years, this pyrite accumulated in the sediments, occurring at varying depths. In some sediment of the coastal plains of the Malay Peninsula, there are considerable amounts of pyrite; however, they are environment-friendly. When the areas are developed for agriculture or otherwise, this pyrite is exposed to atmospheric conditions, resulting in its oxidation which in turn leads to acidity and the formation of yellowish jarosite $[KFe_3(SO_4)_2(OH)_6]$. Toxic amounts of Al and Fe are usually present in the soils and water in the area, affecting crop growth and aquatic life.

Keywords: **Acid sulfate soil, aluminum, Holocene, jarosite, Malay Peninsula, pyritization**

INTRODUCTION

The Malay Peninsula is bordered by Thailand in the North, Singapore in the South, the Straits of Malacca in the West and South China Sea in the East *(Fig. 1)*. Studies conducted in the peninsula found that the sea level rose to 3-5 m above the present high tide position, sometime during the Holocene (Tjia *et al.* 1977). The progradation of the sea many years later led to the formation of sandy beach ridges running parallel to the present shoreline (Roslan *et al.* 2010). The occurrence of these sandy beach ridges along the coastal plains provides evidence of the sea level rise during the Holocene (Tjia 1970). Carbon dating of the oldest sediments in the ridges occurring in the Sunda Shelf (the area between the Malay Peninsula and Borneo Island) indicates their age to be about 6000 years (Haile

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1970). This means that much of the present coastal plains in the Malay Peninsula were once under sea level. This was the time when pyrite was mineralized in the coastal sediments observed today. The soils that developed from pyrite-bearing sediments are termed as acid sulfate soils (Shamshuddin 2006).

Fig. 1: Map of South-east Asia

Acid sulfate soils occur along the coastal plains of the Malay Peninsula (Shamshuddin 2006), Kalimantan, Indonesia (Anda *et al*. 2009), Bangkok Plains, Thailand (van Breemen and Harmsen 1975) and Mekong Delta, Vietnam (Husson *et al*. 2000). These soils are characterized by the presence of pedogenic pyrite (FeS_2), which is readily oxides on exposure to atmospheric conditions, releasing sulfuric acid as well as Al and Fe into the environment. Finally, a new stable mineral called jarosite [$KFe_3(SO)_2(OH)_6$] is or can be formed (Shamshuddin and Auxtero 1991; Shamshuddin *et al.* 1995; Shamshuddin *et al*. 2004a). When this straw yellow mineral appears in the soil profile, the pH would be, almost certainly, below 3.5. In Soil Taxonomy, these soils are mostly classified as Sulfaquepts (Soil Survey Staff 2010).

Acid sulfate soils have been found to occur sporadically in the Kelantan Plains (Soo 1975). Using the soil map he produced, the government of Malaysia established the Kemasin-Semerak Integrated Agricultural Development Project (IADP) in 1982 so as to alleviate hard core poverty among the farming communities as well as mitigate flood by constructing drainage canals. Many years later some soils in the paddy fields were degraded due to excessive acidity related to oxidation of pyrite present in the soils. Later, a soil survey carried out by Shamshuddin *et al.* (2002) re-delineated the area covered by these acid sulfate soils.

Other areas in the peninsula where acid sulfate soils occur include the Kedah-Perlis Plains (Azmi 1982), Perak (Wan Noordin 1980), Pulau Indah, Selangor (Shamshuddin and Auxtero 1991) and Kuala Linggi, Melaka (Shamshuddin et al. 2004a). Some of these soils are utilized with mixed success for the cultivation of rice (Ting *et al*. 1993; Suswanto *et al.* 2007), cocoa (Shamshuddin *et al*. 2004b) and oil palm (Auxtero and Shamshuddin 1991). This paper attempts to explain the process of pyrite mineralization in the coastal sediments during the Holocene as well as attempts to show how pyrite oxidation affects plants and aquatic life in the vicinity.

Evidence Relating Pyrite to Seawater

Wan Noordin (1980) studied in great detail the occurrence of pyrite in some acid sulfate soils found in the Malay Peninsula. His study found remnants of diatoms in the sediments in close association with pyrite (*Fig. 2* modified). Diatoms are living creatures of the ocean. The diatoms could have survived only if the areas where the pyrite in the acid sulfate soils occurred were once inundated by seawater. As such, we conclude that the formation of pyrite found in the soils is closely related to the sea.

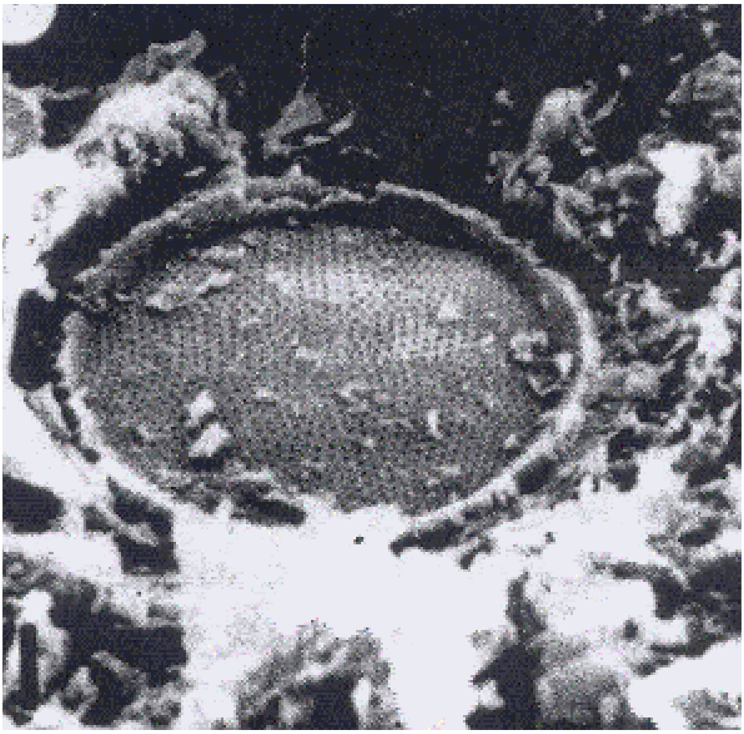


Fig. 2: Diatoms found in pyritic-bearing sediments

Pyritization of the Malay Peninsula

The Kedah-Perlis Plains

Azmi (1982) studied pyrite which is sporadically distributed in the paddy fields of the Kedah-Perlis coastal plains *(Fig. 3)*. The plains are situated in the north-western part of the peninsula, bordering Thailand *(Fig. 1)*. Drainage has oxidized the pyrite, decreasing pH and releasing a soluble form of Al into the soils in the vicinity leading to a reduction in rice yield. Some of the pyrite-bearing soils have been alleviated using lime, resulting in a small increase in rice yield. Some pyrite-bearing sediment was found a few kilometres away from the shoreline, indicating that the areas were once inundated with seawater. This could have occurred when the sea level was at its highest during the Holocene. The occurrence of pyrite in the soils can be used as an evidence of a sea level rise in the plains during the Holocene.

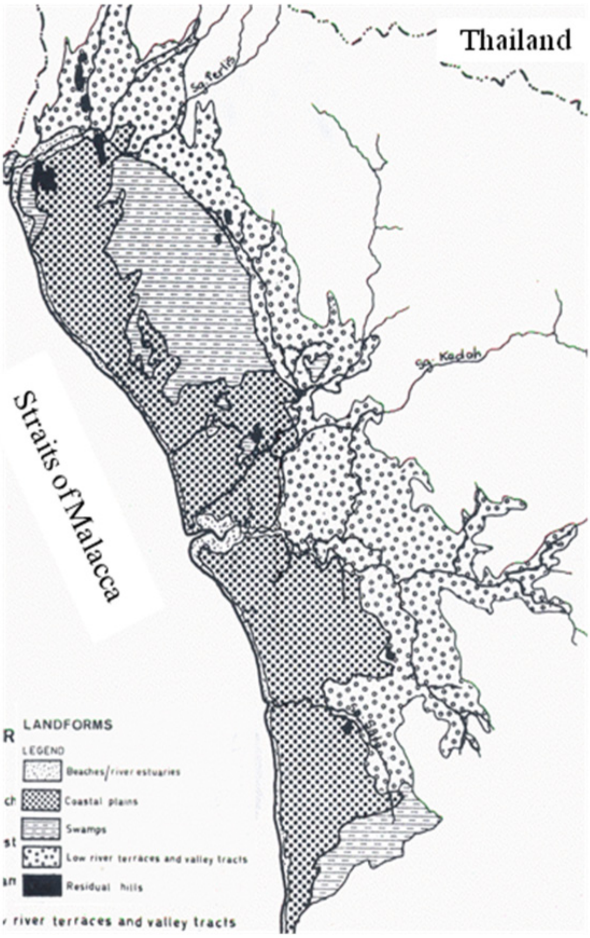


Fig. 3: The Kedah-Perlis coastal plains

Pulau Indah, Selangor

Pulau Indah is an island off the coast of Selangor, a few kilometres from Port Klang. It is only a small island with parts of it being occupied by farming communities *(Fig. 4)*. The areas utilized for crop production contain true acid sulfate soils with low pH and high concentration of Al in the soil solution, and which have an effect on crop growth. The rest of the area, intermittently inundated with seawater, is still occupied by native plant species. The soils, containing pyrite, are classified as potential acid sulfate soils (Auxtero *et al.* 1991). The whole island was completely submerged under seawater 1-2 metres above sea level when the sea level rose some 6,000 years ago. The pyrite in the areas occupied by the farmers was probably mineralized during that time.

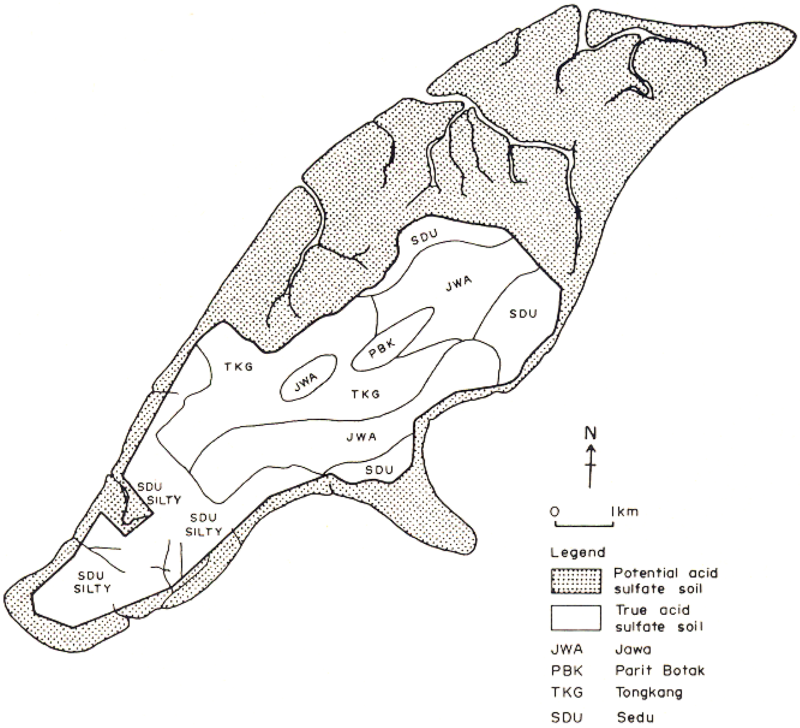


Fig. 4: A soil map of Pulau Indah

Kuala Linggi, Melaka

Shamshuddin *et al.* (2004a) studied the acid sulfate soils found at Kuala Linggi, Malacca. Samples for this study were taken from the Cg horizon of the Linau Series. *Fig. 5* shows the beautiful crystals of pyrite found in the samples. However, jarosite was found in the horizon immediately above the Cg horizon, instead of pyrite. The occurrence of jarosite in this horizon is a result of pyrite oxidation when the area was drained. The pH of the topsoil is very low with exchangeable Al being very high. No crop yield will be obtained without the soil being alleviated using lime or basalt. The pyrite found in the soil of Linau Series was mineralized at the same time as the soils of Pulau Indah, Selangor when the area was inundated with seawater.

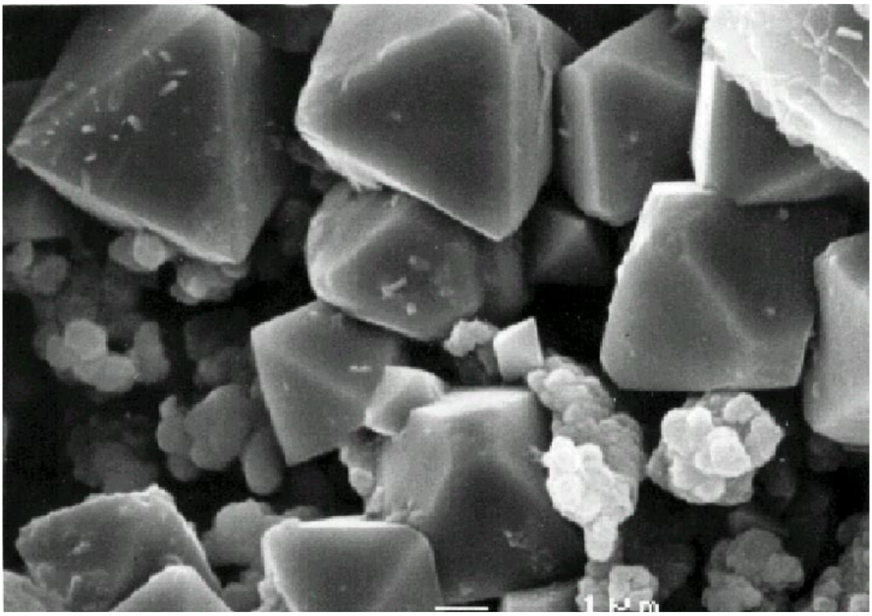


Fig. 5: SEM micrograph of fresh pyrite

The Kelantan Plains

Table 1 show the soil properties of the soils at selected points from three locations in the Kelantan Plains where detailed investigations were carried out were studied. By and large, the first type (pyrite occurred 2 m below the soil surface) was found mostly at Location a (the northern part), the second type (peat overlying the pyritic layer) was found at Location b (the partially drained peaty area - the middle part) and the third type (pyrite occurring in the topsoil) was found at Location c (southern part) (Enio *et al*. 2011) *(Fig. 6)*. At these locations, the pyrite containing layer was found to occur at different depths. Soils had low pH, ranging from 3.24 to 4.89, and contained remarkably high amounts of exchangeable Al (>5 cmol_c/kg). Soil pH values less than 3.5 are indicative of high acidity associated with the occurrence of pyrite/jarosite.

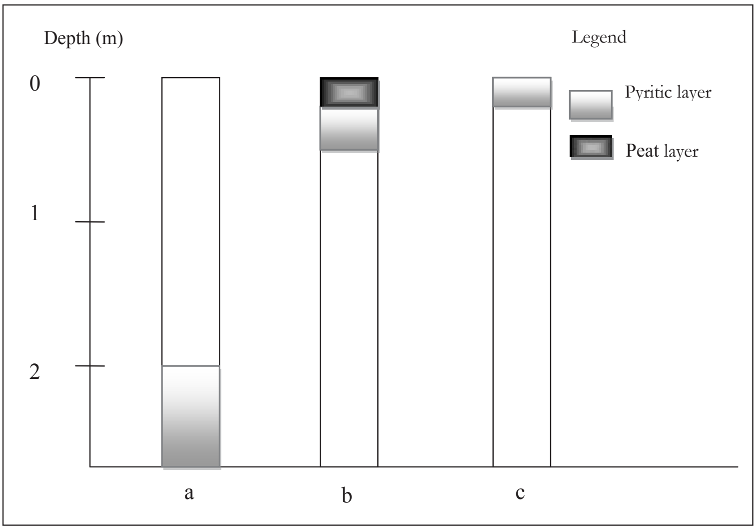
Pyrite-bearing sediments were found at many places in the plains (Enio *et al*. 2011). They were concentrated in the low lying areas (swales) adjacent to the sandy beach ridges *(Fig. 7)*. An imaginary line can be drawn to separate soils containing pyrite from those without (riverine alluvium). Pyrite occurred in the soils to the East of the line and this so-called pyrite border is consistent with the areas demarcated for acid sulfate soils found earlier by Shamshuddin *et al.* (2002). We can, thus, say that this line was more or less the former shoreline about 6,000

TABLE 1
Chemical properties of the soils

Samples	Depth	pH	EC	Exchangeable cations				CEC	Ext. Fe	Avail. P	Total N	Total C
				K	Ca	Mg	Al					
	(cm)	(H_2O)	(dS m^{-1})	(cmol$_c$ kg^{-1})						(mg kg^{-1})	(%)	
1(a)	0-15	4.44	0.14	1.07	0.57	0.26	5.10	11.64	1.96	18.55	0.37	4.64
	15-30	4.16	0.12	0.54	0.56	0.16	6.71	10.36	1.11	18.48	0.18	1.68
	30-45	4.03	0.10	0.50	0.73	0.20	6.78	9.64	1.06	16.80	0.10	1.11
	45-60	4.01	0.08	0.63	0.83	0.36	7.87	10.79	0.43	134.40	0.21	6.29
2(a)	0-15	4.24	0.10	1.23	2.97	1.67	2.66	8.36	2.65	16.66	0.17	1.96
	15-30	4.50	0.08	0.83	4.12	1.97	1.36	7.64	1.90	19.81	0.12	1.06
	30-45	4.89	0.10	0.88	7.09	2.15	0.63	9.43	1.51	11.62	0.11	1.40
	45-60	4.47	0.09	0.96	5.53	2.11	0.56	13.93	3.40	15.89	0.12	2.13
	60-75	4.17	0.15	1.29	8.17	2.19	1.12	12.14	2.61	19.81	0.19	4.32
3(b)	0-15	3.50	0.12	0.76	0.68	0.42	6.54	15.86	1.28	8.12	0.27	6.42
	15-30	3.52	0.17	0.52	0.45	0.42	6.45	13.64	0.49	9.31	0.14	2.68
	30-45	3.51	0.16	0.56	0.41	0.43	4.40	10.29	0.02	10.78	0.92	1.39
	45-60	3.23	0.18	0.48	0.50	0.52	7.61	27.43	0.13	10.64	0.20	8.76
4(b)	0-15	3.42	0.04	0.47	0.76	0.22	5.81	12.50	0.25	11.97	0.18	2.34
	15-30	3.21	0.06	0.52	0.47	0.18	4.47	9.79	0.24	9.73	0.11	1.25
	30-45	3.85	0.06	0.42	0.52	0.11	3.19	8.29	0.26	9.38	0.72	0.42
	45-60	3.72	0.06	0.40	0.35	0.09	2.87	7.71	0.15	10.01	0.66	0.37
5(c)	0-15	4.01	0.06	0.76	0.49	0.23	2.58	19.64	0.27	18.62	0.36	6.78
	15-30	3.80	0.08	0.43	0.44	0.14	6.04	13.64	0.10	10.92	0.14	2.53
	30-45	3.36	0.14	0.54	0.40	0.20	9.12	20.93	0.33	14.28	0.24	8.71
	45-60	3.24	0.19	0.36	0.35	0.23	3.36	20.36	0.12	11.62	0.24	7.91
6(c)	0-15	3.84	0.04	0.82	0.34	0.28	1.96	16.07	0.12	14.28	0.25	5.26
	15-30	3.61	0.04	0.45	0.41	0.12	1.79	11.93	0.23	9.87	0.12	2.07
	30-45	3.60	0.04	0.39	0.35	0.09	1.28	8.57	0.53	10.36	0.87	0.94
	45-60	3.20	0.02	0.16	0.32	0.06	1.04	3.67	0.54	15.75	0.05	0.89
7(c)	0-15	4.02	0.34	1.35	0.71	0.54	7.86	25.43	0.11	10.22	0.51	16.00
	15-30	4.09	0.42	1.37	0.68	0.70	10.81	17.21	0.10	23.10	0.29	11.40
	30-45	3.86	0.14	1.06	1.37	1.66	36.80	30.93	0.62	27.58	0.46	30.32
	45-60	3.24	0.42	0.83	2.88	2.05	92.00	8.16	0.15	23.59	0.47	36.43

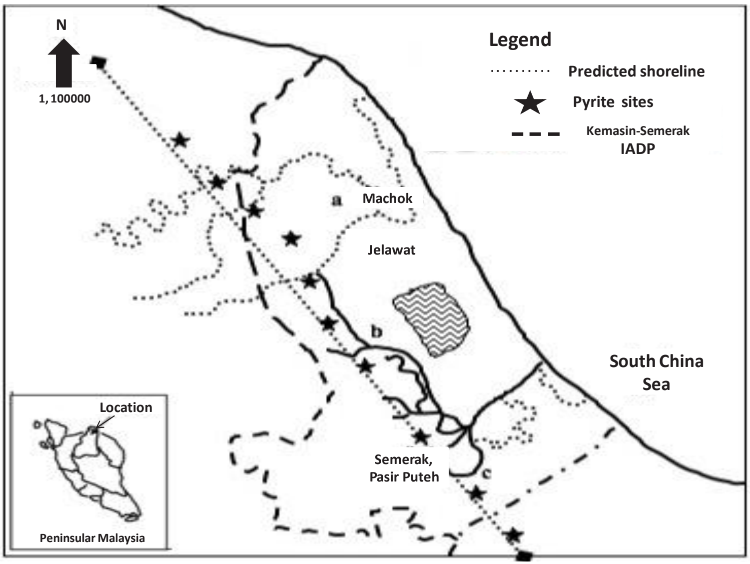
Where, location a (the northern part), the second type was found at location b (the partially drained peaty area-the middle part) and the third type was found at location c (southern part).

(*Source*: Modified from Enio *et al*. 2011) years ago. Therefore, the plains starting from this line eastwards were under sea level during that particular period of history.



(*Source*: Modified from Enio *et al*. 2011)

Fig. 6: The depth in soil profile below which pyrite occurs (The first type in which pyrite occurred 2 m below the soil surface was found mostly at Location a (the northern part), the second type where peat overlying the pyritic layer was found at location b (the partially drained peaty area-the middle part) and the third type in which pyrite occurring in the topsoil was found at Location c (southern part)



(*Source*: Modified from Enio *et al*. 2011)

Fig. 7: The predicted shoreline about 6000 years ago

years ago. Therefore, the plains starting from this line eastwards were under sea level during that particular period of history.

The distribution of pyritic layer in the soils of the Kelantan Plains with depth was studied. Some of the locations where pyrite was present in the sediments are marked as bold stars. It was found that the pyritic layer occurred at three conspicuous depths, namely below 2 m, between 0 to 50 cm and in the topsoil.

The pyritic layer in the soils of the northern part of the study area (near downtown Jelawat) was too deep to be identified by soil auger. For this location, samples were taken in the vicinity of the drainage canals as at the time of the survey, the local authority was cleaning, and widening the canals for maintenance purposes. The materials from the canal bed were dug up and placed on the shoulders of the canals. Samples were left to dry for a few days to determine if they contained pyrite. Clear yellowish mottles appeared within the samples while the pH was about 3. These yellowish mottles are actually jarosite formed from the oxidation of pyrite (Shamshuddin and Auxtero 1991; Shamshuddin 2006). As the drainage canals were more than 2 m deep, some samples were also taken from the area using the auger for physico-chemical analyses. The study results found that the soils from the top 75 cm had pH values above 3.5, giving the impression that the soils might not be acid sulfate soils (Enio *et al.* 2011).

The soils within the partially drained area of the Kelantan Plains are overlain by peaty materials of the Holocene age (Enio *et al.* 2011). During field work, samples were taken at intervals of 15 cm depth and subsequently analyzed to determine their physico-chemical properties. Within a depth of 50 cm, the pH of the soils was found to be generally below 3.5, consistent with the pH value for Sulfaquepts (acid sulfate soils) as defined by Soil Taxonomy (Soil Survey Staff 2010). The air-dried samples indicated the presence of yellowish jarosite mottles.

Soils in the southern part of study area were mostly acidic throughout the profiles. At some locations, yellowish jarosite mottles appeared on the surface of the soils. Clearly, in these soils pyrite occurred within the topsoil. Much of the area had been cropped to rice, but was abandoned when the soils became very acid and Al was found to be present at toxic level, rendering the soils unsuitable for rice production. Currently, the plant species growing in abandoned paddy fields is purun *(Eleocharis dulcis)*, which is Al-tolerant.

Process of Pyritization in the Coastal Plains of the Malay Peninsula

The pyritization process that began in the past is still ongoing in the peninsula. Pyrite forms when sulfate (SO_4^{2-}) from seawater and ferric ions (Fe^{3+}) from marine sediments are reduced to sulfide (S^{2-}) and Fe^{2+} ions, respectively. These reactions occur under anaerobic conditions (extremely reducing) where microorganisms feeding on organic matter present in the sediments play an important role in the reduction process.

Under flooded conditions and in the presence of organic matter, Fe^{3+} ions in the solution are readily reduced to Fe^{2+} ions with the help of microbes (Ivarson *et al.* 1982; Konsten *et al*. 1994). The organic matter needed for the reduction process will be provided by native vegetation. The microbes responsible for this reaction are most probably Desulfovibro desulfuricans (Bloomfield and Coulter 1973; Ivarson *et al.* 1982). The conditions in the area must have been strongly reducing, where Eh was less than -220 mV (De Coninck 1978).

The sea level at and around the Kelantan Plains about 6000 years BP was 3 to 5 m higher than at present (Tjia *et al.* 1977). The sea level here might have been similar to what had been proposed for the Bangkok Plains during that period of history by Pons *et al*. (1982). Carbon dating of the oldest sediments in the Sunda Shelf found their age to be about 6,000 years ago (Haile 1970). This means that much of the present coastal plains in Kelantan were submerged by the sea at that time.

Pyrite found in the study area was probably formed when the sea level was at its highest. The process of pyritization can be summarized as follows (Enio *et al*. 2011):

$$\mathrm{Fe^{3+}}\ \text{(present in the marine sediments)} \rightarrow \mathrm{Fe^{2+}}$$
$$\mathrm{SO_4^{2-}}\ \text{(from seawater)} \rightarrow \mathrm{S^{2-}}$$

Subsequently, the ferrous and sulfide ions had reacted to form pyrite (FeS_2). After a long period of time (probably a few thousand years), the amount of pyrite formed could be substantial in quantity. The overall reaction for the formation of pyrite in the soils of the peninsula can be described by the following equation (Pons *et al.* 1982):

$$\mathrm{Fe_2O_{3(s)} + 4SO_4{}^{2-}{}_{(aq)} + 8CH_2O + 1/2O_{2\,(aq)} \rightarrow 2FeS_{2(s)} + 8HCO_{3(aq)}{}^{-} + 4H_2O}$$

The above equation shows that it needs sufficient amounts of organic matter for the reduction process to proceed without interruption. Furthermore, there must be some oxygen available in the sediments for the microbes that help convert sulfate to sulfide.

This reaction is presumed to have occurred while the sea level was higher than that at present. As the sea prograded due to geological reasons, sandy beach ridges interspersed with swales were created (Roslan *et al.* 2010). Some of the soils in the swales between the ridges contain pyrite (Shamshuddin *et al.* 2002), confirmed by the authors during field work at another time.

Soils at Location a (the northern part of Kelantan, Malaysia) where the first type was found, have a pyritic layer deep down the soil profiles. How did this come about? The plausible explanation for the pyritization process at that location could be as follows. In the northern part of the study area, in the vicinity of Jelawat, the area bordering the shoreline was probably at a higher level. Seawater was able to seep through the porous riverine alluvial materials containing oxides of Fe at a few meters below the surface as evidenced by the presence of high amounts of extractable Fe in the soils of the area. Organic matter required by microbes is also present in sufficient amounts. Given time, pyrite was slowly but surely mineralized in the hydromorphic sediments in the presence of Fe and seawater.

At Location b, (the partially drained peaty area - the middle part) the landscape was totally different from that of Location a (the northern part of the Kelantan, Malaysia). Here, due to reasons unknown to us, swamps were formed when the sea was prograding. The water in these swamps was probably brackish due to intrusion of seawater from time to time, most probably during the monsoon months of November to January. Native plant species have probably been growing since time immemorial, providing enough organic matter for the microbes to survive while converting sulfate to sulfide. If sufficient amounts of Fe^{3+} are available, pyrite is consequently mineralized in the sediments. Given the waterlogged conditions, the organic matter from the plant species growing in the swamps had accumulated forming an organic layer above the mineral soils. Hence, the areas now have soils containing pyrite overlain by peaty materials. A big portion of this area has been gazetted as forest reserve, although it has been partly drained.

Location c (southern part of the area) is rather flat and located close to the present shoreline. The area is not swampy, but floods during the rainy season. Native local plant species also flourish here. Seawater must have reached this area a few thousands years ago, during which time pyrite was mineralized according to the mechanism mentioned above. Jarosite appeared in the topsoil in some localities; jarosite is or can be the product of pyrite oxidation due to drainage.

By identifying the locations where pyrite-bearing sediments are found, we can delineate areas containing pyrite (acid sulfate soils). We can then draw an imaginary line separating acid sulfate soils and the original riverine alluvial materials. This line can be assumed to be the position of the shoreline about 6,000 years ago. It is known that pyritization of the sediments requires sufficient amounts of seawater to supply the sulfate. This means that for the pyrite to be formed in the area, the sea level must have risen a few metres above the present level. As such, we can use the presence of pyrite as yet another piece of evidence for the rise in sea level in the Kelantan Plains during the Holocene. This argument on sea level rise during the Holocene is consistent with the evidence put forward by others, which is based on geological records of the area (Tjia 1970; Tjia 1973; Tjia *et al.* 1977).

Implications of Pyrite-Bearing Sediments for Agriculture

Sediments containing pyrite could be a major problem to the livelihood of the people living in the surrounding areas. Pyrite itself is stable under natural conditions (submerged under water). When the soils containing pyrite are drained for development (for agriculture or otherwise), the pyrite is exposed to atmospheric conditions and subsequently oxidized, releasing high amounts of acidity. The low pH that follows could accelerate the dissolution of silicates in the soils that result in the release of toxic metals such as Al and Fe into the soils and waterways.

This condition naturally affects both rice production (Elisa Azura *et al*. 2011) and aquatic life alike. The water in the paddy fields contains high amounts of Fe as shown by its reddish colour at the onset of the rice growing season.

The above process had taken place in the Malay Peninsula. Some paddy fields in the peninsula are degraded by soil acidity and marked by extreme levels of Al concentration. Growing rice on such unproductive land is uneconomical by any means as the yield is far below the national average of 3.8 t ha^{-1} (Eliza Azura 2012). In order to get a good crop of rice from paddy fields in such areas, dolomitic limestone has to be applied at high rates (Suswanto 2007; Elisa Azura 2012). Also, the soils can ameliorated with the application of ground basalt at appropriate rates (Shazana *et al*. 2013).

CONCLUSION

Pyrite-bearing sediments occur sporadically in the Malay Peninsula. This pyrite occurs at varying depths in the sediments; in the Kelantan Plains, it is below 2 m in the soils of the northern part and in the surface horizon in the soils of the southern part. Oxidation of this pyrite has caused untold damage to the productivity of paddy soils in the area. The pyrite occurring in the coastal sediments of the Malay Peninsula is assumed to have been mineralized about 6,000 years ago when the sea level rose 3 to 5 m above the present. The occurrence of pyrite in the soils is evidence of a sea level rise in the plains during the Holocene age.

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