



## CHEMICAL CONDITIONS IN ACIDIC WATER IN THE PLAIN OF REEDS, VIET NAM

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**Abstract**—In the Plain of Reeds in the Mekong delta (Viet Nam) about 600,000 ha is covered by pyrite soils. At the beginning of the rainy season acidic material is leached from the soil to water in the canal system. Evaluation of water chemistry data indicate that jurbanite is the acidic mineral which is accumulated at the soil surface and subsequently leached to the canals during the early rains. The results obtained correspond well with published results for acidic soils in other areas. It has earlier been assumed that Fe(II) is the major acidic ion in the water and oxidized slowly. The measurements made in acidic canal indicate that Fe(II) is oxidized to Fe(III) according to a pseudo first-order reaction with a half-time of about 4 h and precipitated as hydroxide. Instead aluminium, produced by weathering, has proved to be the most acidic ion in the water.

**Key words**—acidic soils, acidic water, aluminium, iron oxidation, jurbanite, pyrite

### INTRODUCTION

The Plain of Reeds (PLR) in Viet Nam is situated NW of Ho Chi Minh City adjacent to the Mekong River and covers an area of about 800,000 ha and is latticed with canals leading Mekong water through the area towards the sea (Fig. 1). It is under influence of tropical climate with two seasons, one represents the summer monsoon season, hot and humid and one the dry winter (Toan, 1983). About 10 million ha in coastal plains and estuaries in the tropical zone (Pons *et al.*, 1982), about 600,000 ha in the PLR, are greatly influenced by acid sulphate soils containing pyrite (FeS<sub>2</sub>). Pyrite, the origin of acidity, was mainly produced after the last glaciation period in areas with high production of organic matter such as mangrove, which reduced the marine sulphate to sulphide reacting with iron. During the dry season, these soils dry out and cracks form, which give way for oxygen penetration to deeper layers. As a result, oxidation of pyrite will occur leading to acidification. The leaching of acid products starts with the first heavy rains in April. Drainage water is collected in shallow ditches, then, with the following rains, this acid water, with high concentrations of Al<sup>3+</sup>, Fe<sup>2+</sup>, Fe<sup>3+</sup>, SO<sub>4</sub><sup>2-</sup> and pH values as low as 2, drains into the canals.

These processes can be presented by the model in Fig. 2 and may be described as:

Oxygen diffusion leading to oxidation of pyrite and release of acidity and Fe(II).

Released acid reacts with aluminium silicate minerals forming soluble products as aluminium and other cation sulphate salts leaving secondary minerals such as kaolinite or montmorillonite (Eriksson, 1981).

The soluble products move upwards due to capillary forces and evaporation at the soil surface, and eventually precipitate as basic aluminium sulphate Al(OH)SO<sub>4</sub>.

Ultimately rain leaches and drains the acid products into the canal where a chemical equilibrium is established in the water.

In the following will this equilibrium be discussed. Also the iron(II) oxidation under oxygen rich conditions in acid water will be dealt with.

The purpose of this study was to elucidate processes leading to leaching of acidic substances and the fate of these in the canals. This is of importance for the development of agriculture in the whole Mekong delta.

### MATERIALS AND METHODS

#### *The study area*

The Plain of Reeds (PLR), particularly the Tam Nong district, is the center of actual and potential acid sulphate soils. At this time the Hoa Binh canal and its neighbourhood was one of the heaviest affected areas, it can be seen as the main source of acidic water in the whole area. Because of its location close to the two main canals, Hong Ngu and Duong van Duong, which take their water from the Mekong river the acidic water propagates downstream.

For these reasons the Hoa Binh canal and connecting canals were chosen for the study during a field campaign in

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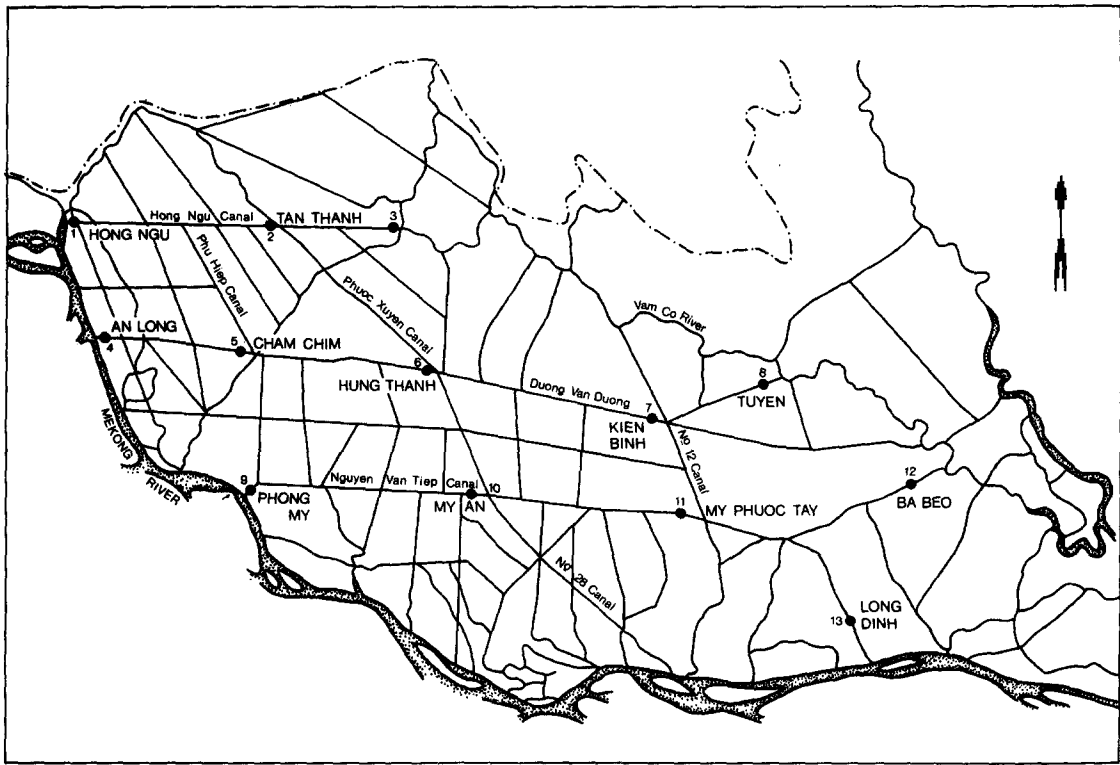


Fig. 1. Map over the Plain of Reeds in the Mekong delta, Viet Nam.

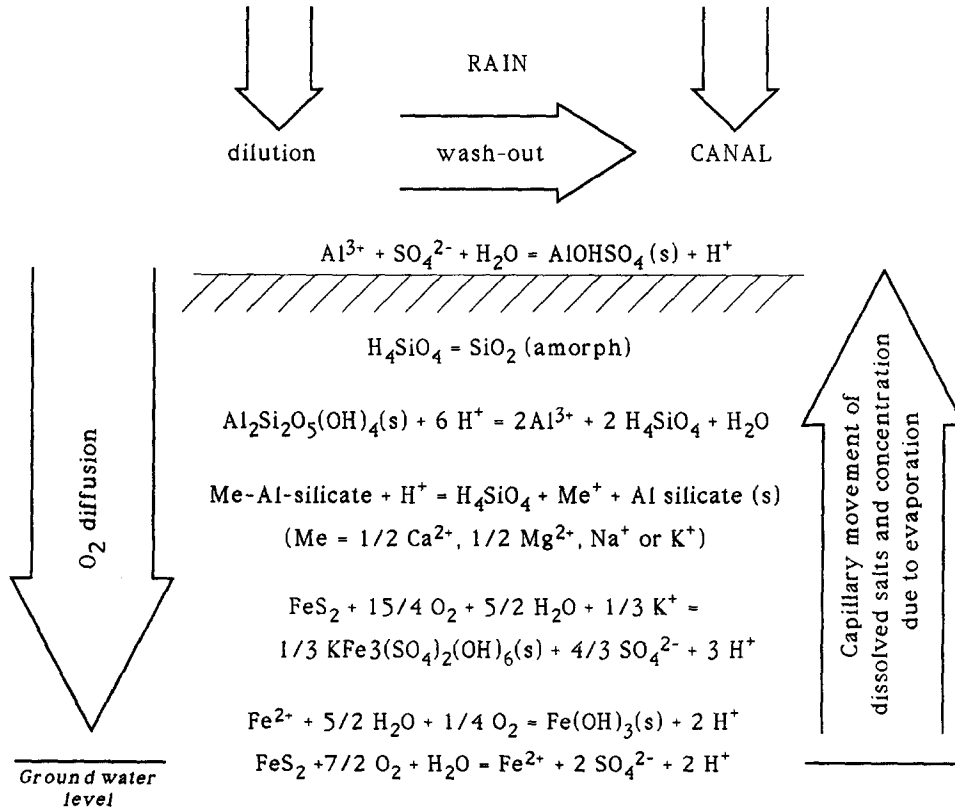


Fig. 2. Schematic outline of processes in acid sulphate soil in PLR.

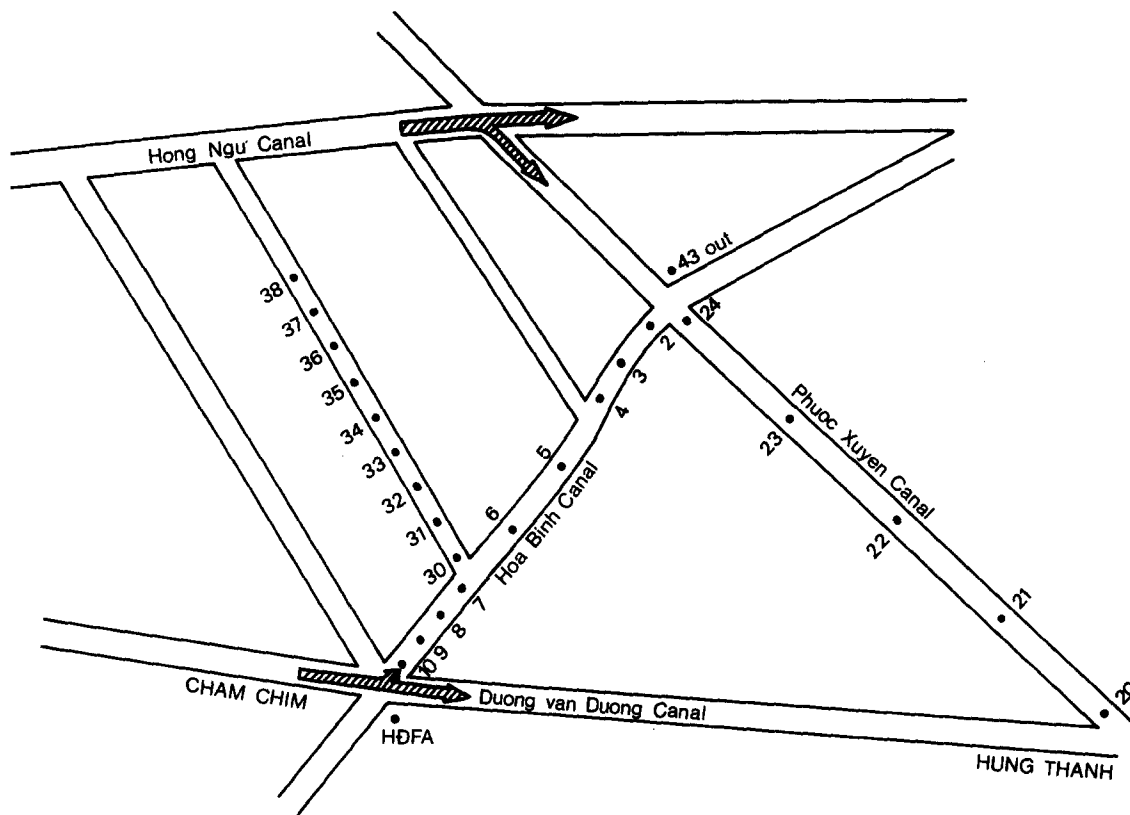


Fig. 3. Map over sampling points during field campaign.

1987 (Fig. 3). In addition the whole area was monitored monthly at 13 main stations (Fig. 1) by the Water Quality Monitoring Programme (Quang, 1988).

#### Sampling and methods

Samples were collected in the field and a 100 ml sub-sample filtered through a 0.45  $\mu\text{m}$  membrane filter and acidified with 5 ml of HCl (1:1) for later analysis of metals at the laboratory.

pH measurements were made in the field using a WTW pH-meter with separate glass and calomel Radiometer electrodes and calibrated against at least two buffer solutions (Standard Methods, 1985).

Conductivity was either determined in the field or in the laboratory soon after sampling with a WTW conductivity meter (ISO 7888). Major cations were analyzed using either a flame photometer or an atomic absorption spectrophotometer (Standard Methods, 1985). Sulphate and chloride were determined by cation exchange and titration of strong acid according to Mackereth. Chloride was determined using an automatic potentiometric titrator (Radiometer) with Ag/AgCl as indicator electrode and AgNO<sub>3</sub> as titrant. The concentration of sulphate was calculated as the difference between the two measurements (Mackereth, 1955).

Aluminium was determined on filtered and acidified samples using a segmented flow method based on pyrocatechol violet (Røgeberg and Henriksen, 1985).

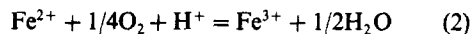
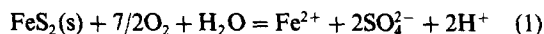
Iron(II) was measured spectrophotometrically using the orthophenanthroline method (Standard Methods, 1985) and total iron with the same method after the reduction with hydroxylamine under slight boiling for 20 min (Standard Methods, 1985). Oxygen determinations were made with an oxygen electrode (ISO 5814). Acidity was determined by potentiometric titration with NaOH to pH 5.6.

The activities of Al<sup>3+</sup>, OH<sup>-</sup> and SO<sub>4</sub><sup>2-</sup> ions were calculated using the Davies equation for the determination of

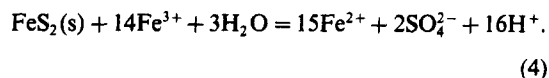
activity coefficients (cf. Stumm and Morgan, 1981) and the ionic strength calculated from the concentrations of the major constituents or estimated by using an empirical relation between electrolytical conductivity and ionic strength.

#### SOURCES OF ACIDITY

The primary source of acidity is pyrite. Oxidation of pyrite is complex, but may be characterized by the following reactions (cf. Nordstrom, 1982a; Taylor *et al.*, 1984)



The oxidation of pyrite (1) releases dissolved iron(II) and acidity to the water, the rate of this process is limited by the diffusion of oxygen from the air into the pyrite soil layer as stated by Dent (1986). The dissolved iron(II) is, according to (2), oxidized to iron(III) and then hydrolyzed to form iron(III) hydroxide (3) thus releasing more hydrogen ions to the water and coating the canal bed with a brown yellow precipitate. Alternatively iron(III) can be reduced by pyrite as described by the following equation (Singer and Stumm, 1970)



Released acidity, along with iron(II) ion may reenter the cycle through reaction (2). The process of oxidation of iron(II) is usually catalyzed by autotrophic

iron bacteria such as *Thiobacillus ferrooxidans* (Zajic, 1969; Singer and Stumm, 1970; Lundgren and Dean, 1979). Under severely acid conditions; Eh

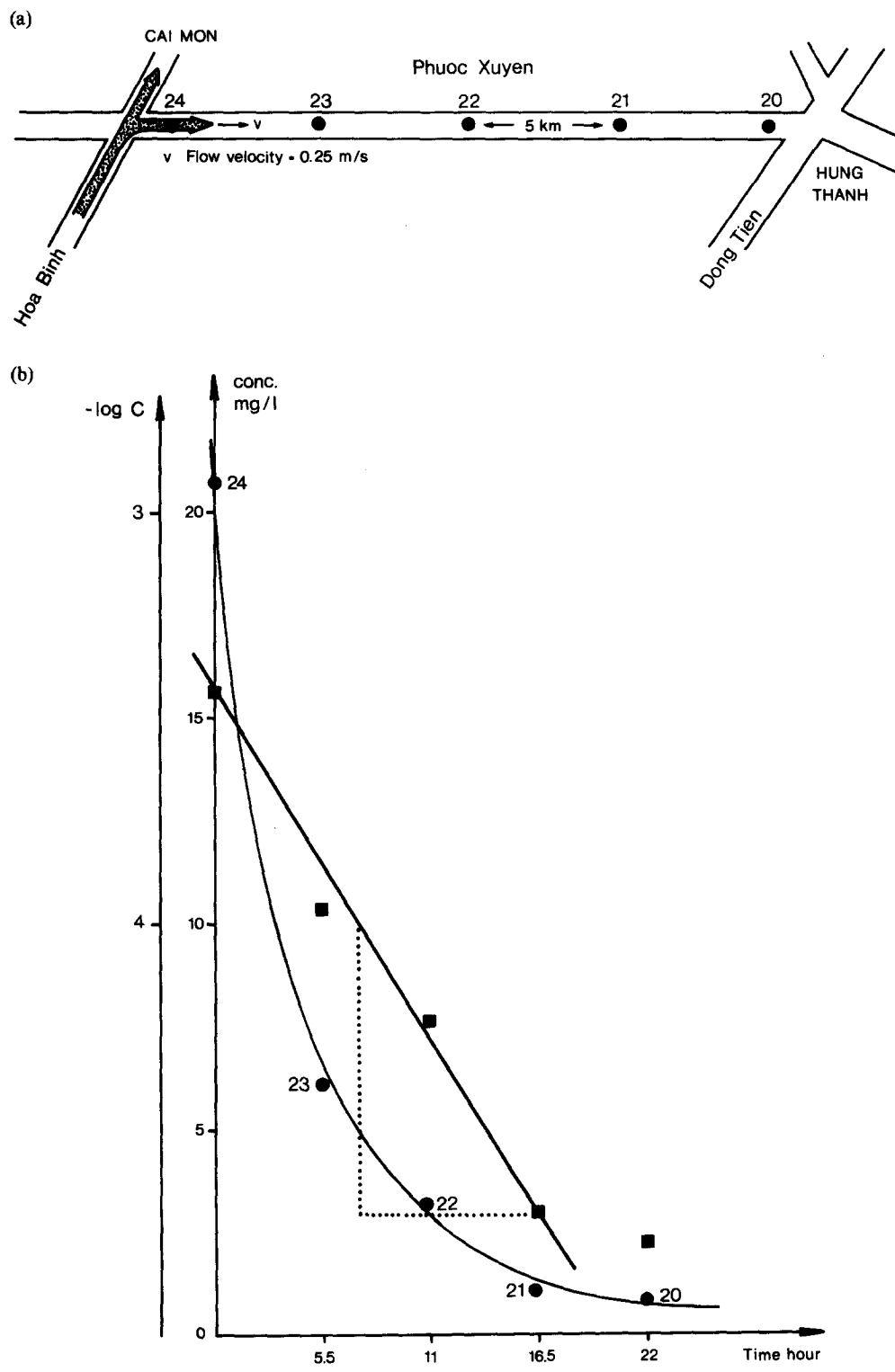
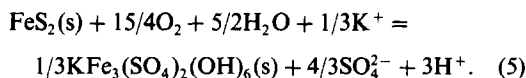
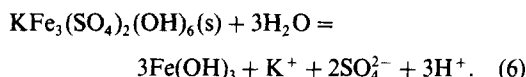


Fig. 4. (a) Map Phuoc Xuyen canal showing main source of acidic water. (b) Changes in iron concentrations along Phuoc Xuyen canal.

should be higher than 400 mV and pH less than 3.7, the formation of jarosite from pyrite may occur (Nordstrom, 1982b, Dent, 1986) according to



The oxidation of 1 mol of pyrite to form 1/3 mol of jarosite  $[\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6]$  produces 3 mol of acidity. At higher pH values, jarosite is metastable with respect to goethite and ultimately it is hydrolyzed to iron hydroxide (van Breemen, 1973)



The hydrolysis of 1 mol of jarosite releases 3 mol of acidity. This mineral can be seen at some depth in the soils and has been considered as the major secondary source of acidity in the PLR (Le huy Ba, 1982).

#### Oxidation of iron(II) in the Plain of Reeds

Iron(II) is stable only in anoxic waters, however at some sources of acidity, the concentrations can be very high as shown in Table 1. Our measurements show that iron(II) does not exist for a long time in the canal system in the PLR, it is quickly oxidized and hydrolyzed to form iron(III)hydroxide. In an attempt to measure the situation under field conditions, the Phuoc Xuyen canal [Fig. 4(a)] was chosen as subject of study. This canal has the following characteristics:

The main source of acidic water with a high concentration of iron is the Hoa Binh canal [the arrow in Fig. 4(a)].

No supplementary source of acid water.

During the travel by boat, water samples were taken every 5 min in the middle of the stream at 0.5 m below the surface. At each station subsamples were filled in two 125 ml bottles, one for immediate EC, Eh and pH measurements, and the second subsample prepared for iron determinations. The results of the chemical analyses are given in Table 2.

From Fig. 4(b) it is obvious that the iron(II) concentration decreases rather fast and as a pseudo first-order reaction. The concomitant decrease in total Fe concentration is caused by the sedimentation of iron(III) hydroxide.

The iron(II) oxidation kinetics is complicated but generally considered to follow the rate law

$$\frac{-d[\text{Fe}^{2+}]}{dt} = \frac{k\text{H}[\text{O}_2(\text{aq})][\text{Fe}^{2+}]}{[\text{H}^+]^2}. \quad (7)$$

Table 1. The concentration of iron and aluminium at two sources of acidity (newly dug pits) in the PLR

Sample	pH	Fe <sup>2+</sup> mg l <sup>-1</sup>	Tot Fe mg l <sup>-1</sup>	Al <sup>3+</sup> mg l <sup>-1</sup>
43 out	2.32	447	891	436
HDFA	2.74	147	680	241

Table 2. Concentrations of iron compounds along the Phuoc Xuyen canal. Sampling sites are shown in Fig. 4

	Sampling site				
	24	23	22	21	20
Fe <sup>2+</sup> , mg l <sup>-1</sup>	20.8	6.21	3.30	6.21	0.93
Tot. Fe, mg l <sup>-1</sup>	31.6	29.9	20.8	29.9	12.1

Singer and Stumm (1970) estimated kH to  $3 \times 10^{-12} \text{ mol l}^{-1} \text{ min}^{-1}$  at 20°C (cf. Stumm and Morgan, 1981). The oxidation rate is considered to become independent of H<sup>+</sup> at pH values < 3.5, leading to an equation (Singer and Stumm, 1970)

$$\frac{d[\text{Fe}^{2+}]}{dt} = k\text{H}[\text{O}_2(\text{aq})][\text{Fe}^{2+}] \quad (8)$$

where  $k\text{H}' = 1.0 \times 10^{-7} \text{ atm}^{-1} \text{ min}^{-1} = 7.3 \times 10^{-5} \text{ l mol}^{-1} \text{ min}^{-1}$  (Singer and Stumm, 1970). The rate increases about 10-fold for every 15°C raise in temperature (Stumm and Morgan, 1981).

In the Phuoc Xuyen canal the concentration of dissolved oxygen  $[\text{O}_2(\text{aq})]$  was 5.9 mg/l and the pH 3.1. Now the kH [equation (7)] can be calculated

$$k\text{H}' = \frac{2.93 \times 10^{-3}(6.3 \times 10^{-7})}{(0.184 \times 10^{-3})} \\ = 1.0 \times 10^{-5} \text{ mol l}^{-1} \text{ min}^{-1} \quad (9)$$

The half-time of the reaction is calculated to

$$t_{1/2} = \frac{\ln 2}{k} = \frac{0.693}{2.93 \times 10^{-3}} = 237 \text{ min} \sim 4 \text{ h}. \quad (10)$$

In the same way the value for kH' using equation 8 can be found. These results are compared with those given by Singer and Stumm (1970) in Table 3.

Our estimated rate constant was seven orders of magnitude higher than the abiotic homogeneous rate constant found by Singer and Stumm (1970). However this rate is low in comparison with those in seawater with  $t_{1/2}$  as low as 3.3–5.5 min as quoted by Sung and Morgan (1980).

From these results some conclusions can be drawn:

The results given by Singer and Stumm (1970) were obtained under laboratory conditions, with pure chemicals containing no catalytic substances, as a result the oxidation is slower. In areas with high concentrations of iron(II), such as the Hoa Binh canal iron(II) will be oxidized rather quick, as the half-time of the reaction is estimated to about 4 h. A proof of the rapidity of this oxidation is the observation that in more distant canals iron was mainly present as iron(III) in solid form, while iron(II) concentrations were 1–4 mg/l.

Table 3. The rate of iron oxidation in PLR according to equations (7) and (8) compared with data from Singer and Stumm (1970)

	Singer and Stumm	This study
kH (mol l <sup>-1</sup> min <sup>-1</sup> )	$3 \times 10^{-12}$	$1.0 \times 10^{-5}$
Half-time $t_{1/2}$ (h)	$1.3 \times 10^7$	4
kH' (l mol <sup>-1</sup> min <sup>-1</sup> )	$7.3 \times 10^{-5}$	16

The above field result refers to quite different conditions:

In presence of light, the reaction is approximately two to three times faster than in darkness (Sung and Morgan, 1980).

Suspended solids and goethite may play a catalytic role.

Microbial oxidation, by the autotrophic iron bacteria, *Thiobacillus ferrooxidans*, may be important (Taylor *et al.*, 1984) and speed up the oxidation by 5–6 orders of magnitude (Singer and Stumm, 1970; Nordstrom, 1982).

Recently Barry *et al.* (1994) presented a study of iron oxidation kinetics and divided the oxidation under natural conditions into a first order reaction [equal to equation (8)], an heterogenous abiotic surface reaction, and a biotic (microbial) reaction.

They found that the addition of 0.49 g TS/l of sterile sediment increased the value of the rate constant 10 times and that the same amount of unaltered sediment increased the value four orders of magnitude. The concentration of suspended matter in the canals were in the same range, generally between 0.1–0.5 g/l.

Even though the value of our rate constant was seven orders of magnitude higher than the abiotic homogenous rate constant found by Sung and Morgan (1980) is our estimated rate of iron oxidation under natural conditions is corroborated by the measurements of Barry *et al.* (1994).

#### ALUMINIUM IN ACIDIC WATER

Aluminium as a toxic element in acidic water is well recognized and it is considered as the most

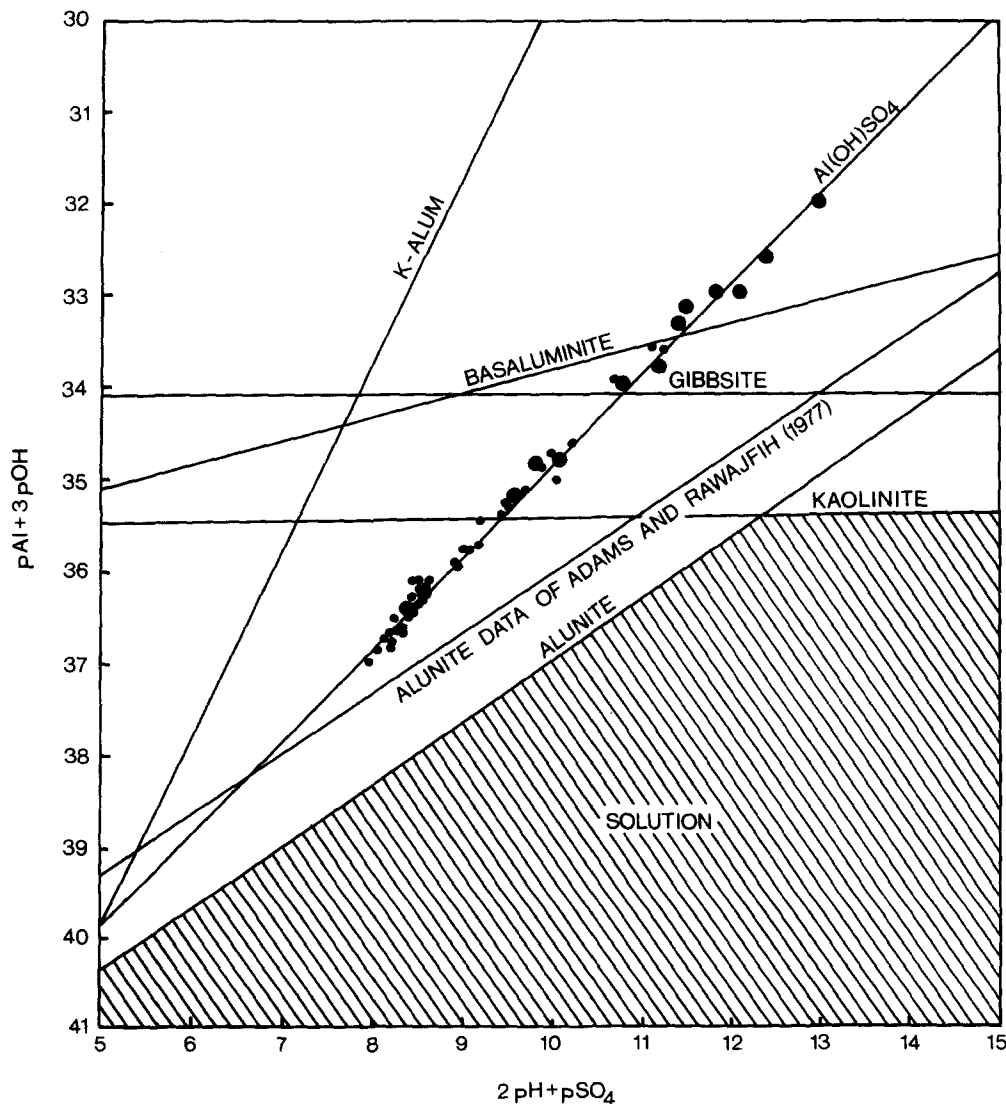


Fig. 5. Data for acidic water from the PLR inserted into a solubility diagram for aluminium compounds (from van Breemen, 1973).

important toxic element for freshwater biota under acid conditions. The toxicity of aluminium compounds is very complex and has been reviewed by among others Dickinson Burrow (1977) and Rosseland *et al.* (1990).

Aluminium occurs in primary minerals such as feldspars, micas, and hornblende. The acid produced by pyrite oxidation will release aluminium through weathering and it will be found either in dissolved form or as hydroxide or in secondary clay minerals as kaolinite or montmorillonite, possibly also as basic salts. The solubility of aluminium is highly depending on pH.

van Breemen (1973) discovered in acid sulphate soil water from Thailand, Sarawak, Kentucky and Pennsylvania a nearly perfect relation, suggesting a solid phase of the composition  $\text{Al}(\text{OH})\text{SO}_4$ , jurbanite

$$p\text{Al} + p\text{OH} + p\text{SO}_4 = 17.3. \quad (11)$$

Later Nordstrom (1982a) estimated the  $pK_{sp}$  to 17.8. The data from our measurement campaigns in the Plain of Reeds in 1986 to 1988 were analysed and compared with data collected by van Breemen (1973) (Fig. 5). The results show that all values lie close to the solubility line for jurbanite. In our case, we obtain

$$p\text{Al} + p\text{OH} + p\text{SO}_4 = 16.96. \quad (12)$$

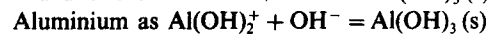
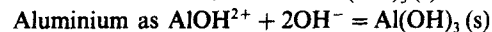
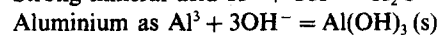
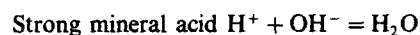
#### State of aluminium as estimated by acidity determination

The acidity of a water sample is equal to the amount of alkali required for the neutralization to a selected end point. In this study the endpoint for the acidity titration is chosen at 5.6 for the following reasons:

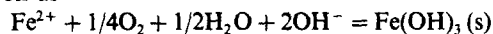
To prevent significant formation of aluminate ion.

The remaining concentration of ionic aluminium will be low enough to be neglected. Be in agreement with the endpoint for the alkalinity titration used.

Thus acidity here consists of



Iron as



In unpreserved samples the concentration of total dissolved iron has diminished significantly after two weeks of storage in the laboratory. As shown previously this reaction also takes place in the canals. At the same time an equivalent amount of  $\text{H}^+$  is produced. The corresponding acidity is the present as strong mineral acid and aluminium species leading to the relation

$$\text{total acidity} = \text{H}^+ + 3\text{Al}^{3+} + 3\text{AlOH}^{2+} + \text{Al}(\text{OH})_2^+ \quad (13)$$

and thus

$$\text{Al (weak) acidity} = \text{total acidity} - \text{H}^+ \quad (14)$$

where  $\text{H}^+$  = strong mineral acidity, mainly  $\text{H}_2\text{SO}_4$ . In order to estimate the existing protolytic state of aluminium in solution, the equivalent weight (EW) can be used

$$\text{EW} = \text{conc. Al} / \text{Al acidity}. \quad (15)$$

If EW is 9 (27/3) then aluminium exists mainly as  $\text{Al}^{3+}$ , if  $9 < \text{EW} < 27$  then there is a mixture of  $\text{Al}^{3+}$ ,  $\text{AlOH}^{2+}$ , and  $\text{Al}(\text{OH})_2^+$ , and if EW is close to 27 will  $\text{Al}(\text{OH})_2^+$  predominate. Some of the calculations are presented in Table 4 indicating that the major portion of acidity is aluminium ions as  $\text{Al}^{3+}$ .

Table 4. Values on acidity [cf. equations (13) and (14)] and the calculated equivalent weight [cf. equation (15)], EW (g equivalent<sup>-1</sup>) for acidic water in the PLR, June 1987

Sample No.	pH	EC mS m <sup>-1</sup>	Acidity				
			Mineral meq l <sup>-1</sup>	Total meq l <sup>-1</sup>	Weak meq l <sup>-1</sup>	Al conc mg l <sup>-1</sup>	EW g eq. l <sup>-1</sup>
2	2.95	141	1.56	5.95	4.39	44.2	10.0
3	2.96	150	1.54	5.98	4.44	48.0	10.8
4	3.03	153	1.31	6.12	4.81	46.5	9.6
5	3.10	126	1.09	5.29	4.20	39.6	9.4
6	3.14	128	0.995	6.13	5.14	54.4	10.4
20	3.08	133	1.147	4.33	3.18	32.7	10.2
21	3.09	143	1.13	4.55	3.42	35.2	10.3
22	3.13	115	1.00	3.56	2.56	25.6	10.0
23	2.94	165	1.64	7.25	5.61	54.3	9.6
24	3.19	109	0.87	4.07	3.2	30.9	9.6
30	2.93	218	1.84	9.62	7.78	83.1	10.6
31	3.01	257	1.49	9.23	7.74	87.4	11.3
32	3.05	260	1.36	8.51	7.15	76.9	10.7
33	3.04	256	1.39	7.22	5.83	72.6	12.4
34	3.03	261	1.43	6.06	4.63	58.9	12.7
35	2.98	254	1.60	6.35	4.75	62.1	13.0
36	3.07	232	1.28	5.48	4.2	58.1	12.4
37	3.0	236	1.50	5.13	3.63	52.9	14.6
38	3.14	226	1.08	4.5	3.42	49.4	14.4

## CONCLUSIONS

The genesis of acidification in the PLR is pyrite oxidation, which consists of a series of processes. Iron(II) is produced in the first step of oxidation, but does not exist for a long time in oxygenated acid water. The half time for the iron(II)oxidation in the Plain of Reeds was determined to about 4 h, thus much faster than expected from literature data for pure solutions at a pH value of 3. After oxidation iron(III)hydroxide will precipitate along the canals. The suspected toxicity from dissolved iron(II) was thus found to be of less importance. Also the importance of iron as a major acidic component was disproved. Thus steps to improve iron oxidation will not be actualized.

It is now clear that aluminium becomes the main subject for research on effects in the canal water of the pyrite oxidation, and will, together with strong mineral acidity be the major criterion for water quality for fishery and agricultural purposes.

Previously jarosite was considered as the main intermediate source of acidity in the Plain of Reeds. This study has shown that jurbanite, probably present on the soil surface, is the main source of acid releasing acidity with the first rains.

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