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MINE WATER QUALITY DETERIORATION DUE TO ACID MINE DRAINAGE

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ABSTRACT

Water quality deterioration due to acid mine drainage is of concern in the northeastern coalfield of India as the water in this area is severely polluted. The mine drainage water emanating from various collieries are highly acidic in character and contain high hardness, sulphate, total dissolved solids and iron coupled with low pH values - which further results in contamination of trace (heavy) metals at significant levels. Trace metals are highly toxic and undesirable and are injurious to human health.

These acidic waters are also typically hard in character because of iron sulphate content rather than common Ca-Mg bicarbonate type hardness.

INTRODUCTION

Acid mine drainage (AMD) gives rise to several problems of environmental degradation - especially pollution of aqueous environment. The problems of AMD are intensely localised in the northeastern coalfield of India, where ecology of the surrounding area is badly disrupted. Rawat and Gurdeep Singh (1982-1983) have reported on the nature and occurrence of AMD from some northeastern coal mines in India.

Coal mining operations cause considerable changes in the amount, distribution and quality of water in surrounding areas, as indicated by Gorbett (1977), Cairney and Frost (1975) and Rogowski et al (1977). The contamination of trace metals along with other weatherable minerals in AMD which are acid soluble and are leached from the coal and associated strata during mining operations, result in severe water quality deterioration in the mining areas. Polluted water of this type will not support aquatic life, destroys mining equipment, increases the cost of water treatment for various water supplies and also leaves the water unacceptable for drinking and recreational purposes. This paper presents and discusses the effect of AMD on water quality deterioration at some of the northeastern coal mines of India.

EXPERIMENTAL PROCEDURE

Mine water samples were collected from various sumps of Ledo (Tirap), Baragolai, Jeypore and Tipong collieries of northeastern coalfield of India.

Standard methods were employed for estimation of various constituents in mine water samples (American Public Health Association, 1971). The pH and conductivity values were measured using Philips pH-meter and Systronics conductivity-bridge, respectively. The quantitative analyses of trace metals were done using Atomic Absorption Spectrophotometer, SP 1900, Pye Unicam. Dilution has been done wherever necessary in case of some samples.

Elemental Analysis of Coal Samples - Coal samples were powdered to about - 100 mesh and filled in carbon cavity. These samples were analysed on Hilger and Watts emission spectrograph using DC arc anode excitation. Exposure time to 12 seconds were given and pre-exposure time of arching was 2 seconds. Carbon electrodes were used for purpose of arcing.

GEOLOGY

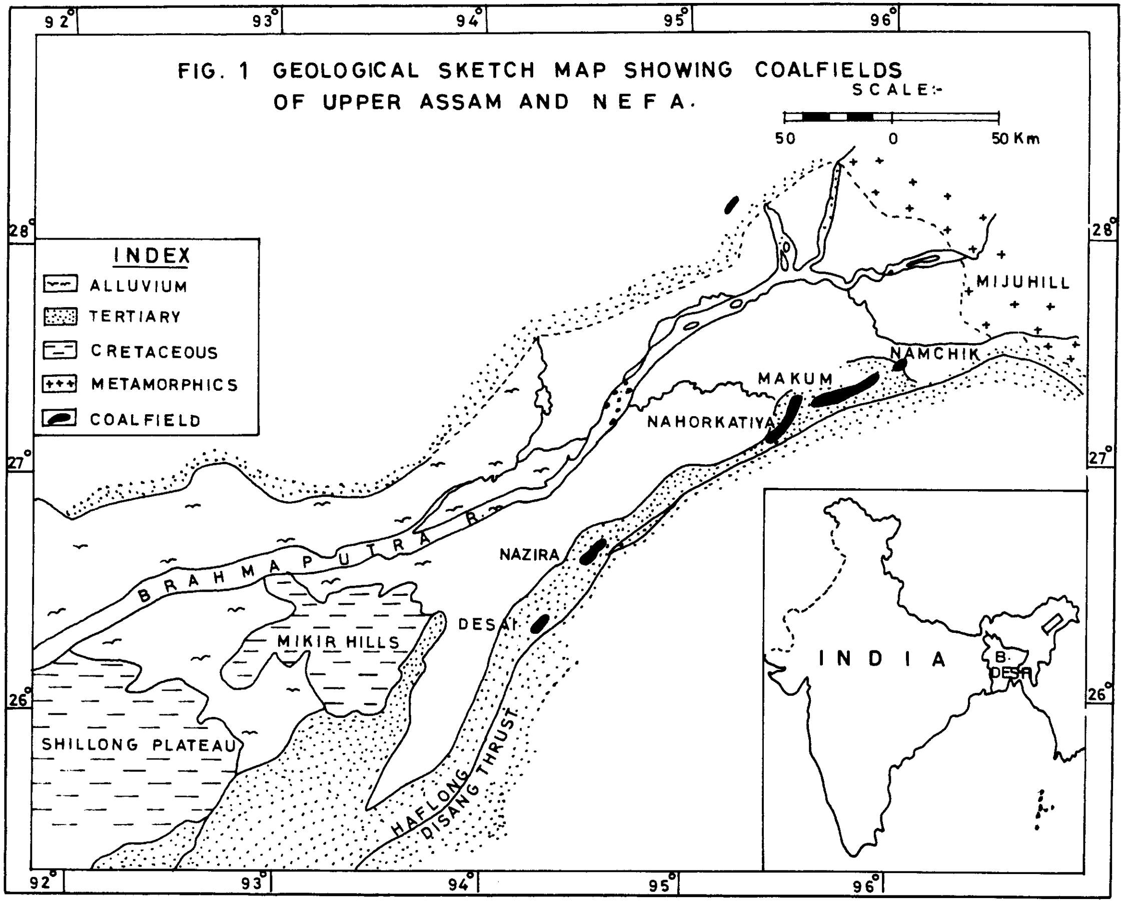
Geological map of the northeastern coalfield of India is given in Figure 1. Geology of northeastern coalfield is peculier in nature and is not found in other coal bearing areas of the country. A narrow belt of Tertiary rocks is exposed in the southern foot-hills of the Shillong plateau throughout the greater part of its length west to east and continues north-eastward through the north Cachar hills and the Mikir hills (Evans 1932). The Tertiary formations containing coal seams occur within a long narrow belt trending NE-SW (alongated syncline known as Namdang syncline and an anticline on the north, called the Ledo anticline). The whole coal-bearing formation lies between two major thrusts, known as Margherita or Naga thrust (concealed beneath alluvium) to the north and the Haflong-Disang thrust on the south. Within this belt again there are several minor thrusts generally parallel to the major thrusts. The formations in general show very steep south-eastern dip.

Lithologically, both the Baragolai and the Tikak Parbat stages are very much akin to each other but thick coal seams are present only in the latter. The seams of the Nazira, Makum, Jeypore and Namchik-Namphuk areas are included in the basal 120 m of Tikak Parbat stage. The present studies were concentrated mainly on the collieries existing in the Makum and Jeypore coalfield, where major mining operations are being carried out.

Makum Coalfield

The coal bearing rocks in this field belong to Varail series, and are represented by a thick sequence of sedimentaries of varied facies, comprising predominantly of medium to coarse grained ferruginous sandstones, bluish to greyish clays, sandy clay, clayey sandstones, thinly bedded greyish shales and carbonaceous shales with fine workable coal seams. Coal reserves occur mostly in synclines in the rocks of two different lithological units - the Baragolai and Tikak Parbat stages of the Varail series of Tertiary age. Among the two stages of the Barail series of coal seams of workable thickness are present in the Tikak Parbat stage at depths varying from surface outcrop to over 300 m.

FIG. 1 GEOLOGICAL SKETCH MAP SHOWING COALFIELDS OF UPPER ASSAM AND N E F A.



It is in this field where the Oligocene coals have attained maximum development. Of the fine coal seams occuring in the field 1st and 3rd from bottom upwards are major seams, the average thickness of the seams and partings varies within wide limits.

Jaypore Coalfield

In this coalfield rock formations of only Tikak Parbat stage of Barail series have been met. Lower most part of the Tikak Parbat stage and the older formations have been truncated by Haga thrust that runs almost parallel to the strike of the beds. The Tikak Parbat stage consists of light grey to brownish, fine to medium grained well-bedded sandstones, light grey to clays, fine shale bands to the coal seams. Sandstones belonging to lower portion of the stage are carbonaceous as described by Puri (1968). Regional strike of the beds is NNE-WSW. Dips are southerly, varying from 25° to about 80° but usually 45°.

Six coal seams have been recorded in Jaypore and Jeypore colliery areas. As there is an unconformity towards the top of the Tikak Parbat stage and as the lower portion of this stage has been truncated by Naga thrust, complete section is nowhere exposed.

Seam IV is 1.51 m thick, underlain by dirty white to light grey clay, sandy clay and fine to medium grained sandstones. Parting between No IV and V seam is very little consisting mainly of very light grey to grey coloured clay. The roof of V seam (which is only 0.60 m thick at any point of sampling) is grey shale and clay. Chaudhuri (1978) reported that the floor of VI seam is light grey coloured clay, which contains white calcite patches. This clay is marly in nature and effervesees strongly with HV1. The roof of VI seam consists of grey shale and clay and contains yellow coloured pyrite specks and patches and there is complete absence of carbonate minerals in this upper part.

Small quantities of clay ironstone in nodules and layers as well as minute particles of iron pyrite often accompany the argillaceous sediments with the coal measures in general.

HYDROLOGY

Water enters a mine as ground-, surface-, mine process-, backfill- and fresh-water, and the surplus water is collected in sumps and pumped to the surface. Mine process-water is used for the control of dust in drilling, blasting and other operations. Water also enters from quarries which are directly connected to underground workings, in addition to direct entry of surface water through inclines. Water may seep from upper to the lower or from the lower to the upper seam or laterally through the adjacent rock strata, depending on the position of working. Faults, fissures, cracks, joints etc are the most dangerous occurrences in a mine which may connect with a water source at the surface of the working seams and when the workings are advanced at that point, water may start rushing in and may inundate the mine.

The rock formations in the NEC, are unconsolidated and highly permeable, causing water leakage problem in the coal mines of the region. Before mining, rocks above the coal are saturated with rain and groundwater. As mining progresses, the overlying rocks get fractured and ground water starts to drain through the rocks. The unusual wetness of the mines (relative humidity sometimes approaches 100 percent) is due to local geological structure. The coal measures are folded into an asymetrical basin cut by faults and the dip of the coal seams allow water easy passage into the underground workings. Table 1 illustrates the typical discharge data from the coalines of the area.

Table 1 Typical Discharge Data

	Mine	Flow (gpm) Rainy season	Winter season	pH
Ledo (Tirap)	(i) New Mine	800	200	2.7
	(ii) M & P Dip	200	50-100	3.1
	Baragolai	300-400	200	4.2
	Tipong	200	50-100	4.0
	Jeypore	400-500	100-200	2.4

gpm = gallons per minute

This shows that the function of the water is a transport media which flushes the oxiddation products of sulphide minerals in coal and associated strata and carried them into the aqueous environment. The final discharges flow to regulated nearby revines and rivers.

CLIMATE

The area experiences a heavy rainfall, ranging from 250 to 300 cm and 400 cm has also been recorded during the period from April to December. During this period the temperature goes upto 32°C (90°F) in June/July and a high humidity of 91 percent is experienced. During winter, the temperature comes down to 10°C (50°F) and humidity reduces to about 85 percent. In the mines, however, the humidity in the depillaring areas vary from 95 percent to 97 percent.

BACKGROUND OF AMD FORMATION

Occurrence of AMD in coal mines of Assam is due to the association of sulphur with coal and associated strata. Gurdeep Singh (1983) has found that organic sulphur constitutes 50-90 percent of total sulphur present in coal, is structurally bound in coal and does not produce any significant acidity in the mine drainage. Pyritic sulphur is the main cause of acid production. This type is further subdivided into stable and reactive type. The latter occurs with particle size 0.25 μm - 5 μm (fraboidal type) and is extremely reactive and under vigorous climatic conditions, exposed to oxygen and moisure producing sulphuric acid and iron sulphate. This permits rapid growth and activity of certain acidophilic (bacteria). The bacteria Thiobacillus ferrooxidans enhance acid producing reactions at a much faster rate than chemical oxidation (Gurdeep Singh and Bhatnagar, 1985). Strata water from permeable and porous rocks gains access to colliery workings through a number of channels and carries the acid produced by chemical and biochemical oxidation of pyrites. This underground water containing dissolved acids is called AMD.

WATER QUALITY DETERIORATION

The chemical analysis of mine water samples listed in Table 2 show that the waters are severally polluted. The mineralogy of the geological regime controls, to a larger extent, the chemical quality of water permeating through the system.

In general, pyrite and calcareous material present in the system have the greatest chemical impact on the aqueous regime. The waters from NEC mines are generally acidic and toxic in nature. The various physico-chemical parameters of water quality, affected by mining operations are described below.

pH, acidity, iron and sulphate

The pH, although not a quantitative measure of acidity, reflects the degree of acidity or alkalinity in the drainage. In general, the pH of the mine waters is affected as a result of mining. Considerable changes of pH from neutral to highly acidic has taken place when these waters flowed over mined areas. The pH survey in conjunction with acidity and sulphate data show that severe acidic conditions exist at the Ledo (Tirap) and Jeypore colliery areas, while mild acidic conditions prevail in the Baragolai and Tipong colliery areas. This reflects the predominance of reactive pyrite in the seams of these areas and difference of acidity and sulphate contents may be due to variation of occurrence of reactive pyrite minerals in these areas. The less acidity and more sulphate contents indicate that alkalinity produced from calcareous materials subsequently neutralised some acid in the drainage. In some sections, however, drainage waters are found to be in near neutral to alkaline range, showing that there is a paucity in the particular section of the seam and predominance of calcareous materials.

The acidity in these mine drainage arises principally from the formation of free sulphuric acid and the hydrolysis of oxidation products (iron sulphates) of pyrite. Sulphate content represents the amount of available reactive pyrite in the particular seam. The relationship between acidity and sulphate can be governed by the following chemical reactions as reported by Caruccio et al (1977).

$$Fes_2 + H_2O + 3\tfrac{1}{2}\ O_2 = FeSO_4 + H_2SO_4 \qquad (1)$$

Ferrous sulphate thus formed from the oxidation of pyrite, may be oxidised further to ferric sulphate in presence of bacteria and dissolved in the acidic water (Gurdeep Singh and Bhatnagar, 1985).

$$Fe_2\ (SO_4)_3 = 2\ Fe^{+++} + 3SO_4^{-2} \qquad (2)$$

Fe^{3+} can hydrolyse on

$$Fe^{+++} + 3H_2O = Fe(OH)_3 + 3H^+ \qquad (3)$$

to form acidity and iron hydroxide. The Fe^{3+} iron formed during the acid-generating reactions further reacts with the available pyrite to form additional acidity, ferrous and sulphate ions. These reactions indicate that sulphate is in part stoichiometrically related to acidity as reported by Caruccio (1968). Although, sulphate and iron arise from a common source, the acidity generated is a function of oxidation state of iron. Acidity increases as the iron is oxidised. However, this oxidised iron usually precipitates out at the base of drainage due to less solubility unless the pH is very low. Sulphate content remains constant and can be used to aproximate the degree of acidity present or that was present before neutralisation took place.

When the neutralizing capacity of the environment is exceeded, acid begins to accumulate and the pH decreases. As the pH decreases, the rate of iron oxidation by oxygen also decreases. But at lower pH, the rate of iron oxidation by T.ferrooxidans increases. The action of the bacteria causes increased acid production, which serves to further lower pH. As the pH falls to less than 3, there is increased solubility of iron and the decreased rate of $Fe(OH)_3$ precipitation and overall rate of acid production enhanced. This is supported by the pH, acidity, sulphate data of mine waters when related to iron contents in Table 3.

Hardness

Hardness is a measure of the occurrence and abundance of divalent cations ie, Ca^{2+}, Mg^{2+}. Very high hardness values in these mine water samples indicate that these waters differ from the more common type of hard waters in that sulphate rather than bicarbonate is the dominent anion. Corbett (1977) and Caruccio et al (1977) indicated that the high hardness values coupled with high acidity and low pH values indicate that the primary cation contributing to it is soluble iron apart from Ca^{2+} and Mg^{2+} ions. The validity of these high values of iron (upto 760 ppm) in mine waters is supported by calculation of electro-neutrality, which achieves a balance only upon inclusion of iron data and either approach or become iron-sulphate type waters in some cases (usually in the Ledo and Bagagolai waters). In other cases these are Ca-Fe-Mg sulphate type of waters.

Conductivity

The specific conductivity values can be related to the total dissolved solids and this approximate the chemical activity taking place within the aquifer or coal seam through which water flows. Specific conductivity values further confirm the concentration of the total chemical species in water.

In general, these waters have low conductivity in conjunction with low pH reflect the nonavailability of calcareous material and are unable to afford the natural buffering capacity in the geological regime.

Table 2

Trace Metals Analysis of Mine Water Samples

Parameter (mg/l)	Ledo (Tirap)			Mine Water Samples				Jeypore	
	1	2	3	4	1	2	3	4	5
pH	2.7	2.7	2.5	3.1	1.9	2.8	2.45	2.3	2.5
As	1.6	0.4	-	-	0.4	0.4	-	1.0	0.6
Cd	1.2	-	0.9	4.8	-	-	0.3	4.0	1.6
Cr	6.6	0.8	10.0	6.5	11.0	1.0	0.9	4.6	8.5
Cu	0.4	0.7	0.7	-	-	0.6	0.9	-	0.3
Hg	0.3	-	-	-	0.5	-	-	0.2	0.9
Pb	1.1	0.7	1.6	0.9	0.5	1.2	1.0	0.9	0.9
Zn	2.5	1.0	2.4	3.0	1.2	4.6	0.8	1.5	0.8
Mn	2.4	17.0	12.0	8.2	8.2	8.2	1.6	1.2	3.4
Al	2.0	1.9	49.0	3.6	10.5	44.0	29.0	56.0	13.0
Ni	3.0	3.6	3.1	3.5	3.1	4.0	N.D	4.5	6.0

Table 3 Chemical Analyses of Mine Water Samples

Mine water sample	pH	Acidity (ppm)	Sulphate (ppm)	Hardness (ppm)	Total Fe (ppm)	Ca (ppm)	Mg (ppm)	Dissolved solids (ppm)	Specific conductivity mhos at 25°C x 10 ^{2}	F^- (ppm)
Ledo (Tirap)										
Sample 1	2.7	2040	3050	2300	350	104	108	2518	0.8	1.10
Sample 2	2.7	2100	2500	1800	358	98	78	3180	0.5	0.60
Sample 3	5.3	95	1662	990	120	125	63	1662	0.15	0.60
Sample 4	3.1	1230	2880	3300	670	245	74	4200	0.4	1.10
Sample 5	2.5	980	3210	1130	136	116	75	3060	0.2	1.56
Jeypore										
Sample 1	2.9	1090	2420	2900	545	65	66	4210	0.5	1.36
Sample 2	2.8	1185	3100	3170	760	60	100	3816	0.4	1.27
Sample 3	2.45	2350	3110	2120	388	42	84	2680	0.5	1.22
Sample 4	2.5	2110	2956	1930	385	58	49	4066	0.5	1.20
Sample 5	2.3	2480	3030	1490	192	50	58	2600	0.2	1.56
Sample 6	5.9	88	1156	590	100	328	104	1190	0.16	0.80
Baragolai										
Sample 1	8.4	-	607	415	22	250	112	750	0.15	0.60
Sample 2	3.98	190	984	900	60	222	96	1200	0.21	0.96
Sample 3	4.45	144	1010	790	120	126	59	874	0.14	0.90
Sample 4	4.1	160	529	595	56	288	78	1180	0.32	1.10
Sample 5	4.3	124	1235	840	50	530	120	1530	0.14	1.22
Tipong										
Sample 1	3.87	210	611	570	39	98	68	814	0.24	1.10
Sample 2	8.15	-	500	605	16	504	110	780	0.16	0.70
Sample 3	4.1	100	984	890	45	400	112	936	0.22	1.50

1. Samples are light yellow - yellow - yellow reddish in appearance.

2. Mean values are given in table.

Table 4

Spectrographic Analysis of Coal Samples

Sample	Major elements about 1.0 percent	Major elements 0.1 - 1.0 percent	Trace 0.1 percent	Not present
1. Coal powder sample	Fe, Al, Mg, Si, Ti, Ca	Ba	C, Cr, Cu, Ni, Ga	Na, K, Sr, Mn, Ni, Pb, Co, Mo, Zn, Sn, Ge, Sb, Bi, In and rare earths
2. Coal lump sample	Fe, Al, Mg, Si, Ti, Ca	Ba	Cu, Ga	Na, K, Sr, V, Mn, Cr, Pb, Ni, Co, Mo, Zn, Sr, Ge, Sb, Bi, In, rare earths

Concentration of F^- ion seldom exceeds 2 mg/1 in these mine water. Apart from rare occasions where F^- may occur as a matrix mineral, a geological source of F^- in area is unlikely and most of the F^- input is probably from rainfall. This further supported by the fact that in winter season F^- concentration in mine water become negligible.

Trace metals contamination

At the low pH levels, the toxic tract (heavy) metals are more soluble and enter into solution as a result of secondary reactions between iron sulphates, sulphuric acid and the compounds in nearby clays, sandstones, limestones, sulphides and various organic substances present in mine drainages or streams and explains the existence of Ca, Mg, Al, Ne, K, Mn, Zn, Pb, Cu, Cr, Cd etc in mine drainages (Rawat and Gurdeep, 1983).

Clays (Kaolinite and Illite) by virtue of their ion exchange process can preferentially adsorb heavy elements and release lighter ones, thus can change the concentration of various ions in the mine drainage (Smith, 1974, Barnshisel and Rotremel, 1974). However, ion exchange process is favourable only in the pH range between 4-10 and in AMD waters where pH is less than 4, the ion exchange process may not be marked.

Trace metals as quantitatively determined by atomic absorption spectrophotometer are given in Table 3 show that these are present in significant amounts and are injurious to human health. The various physico-chemical characteristics and especially trace metals contained in these acidic mine drainage exceed the limiting drinking water standards (given in Table 4), as laid down by various international organisations (Scott and Smith, 1981).

The various major (above 1.0 percent, minor (0.1 - 1.0 percent) and trace (0.1 percent) elements as analysed by Emission Spectrograph, in the coal samples, are listed in Table 5. However, some trace metals (only those could be analysed by AAS) which are absent in coal samples are found to present in significant quantities in AMD samples show that some additional sources of these trace metals occur in the rock sequence.

Hill (1972) reported that the most coal bodies are associated with sulphides and/or sulphosalts. The most common sulphides are the iron, ir, Pyrite (FeS_2), pyrrhotite and marcasite. In addition, there are numerous other sulphides and sulphosalts. Hawley (1972) cited the general formula for sulphides as $A_m X_n$ where A consists of a metallic elements or sometimes As, Sb and Bi. The general formula for sulphosalts is $A_m B_n X_p$. The diversity of the combinations of the chemical elements is tremendous, and over 125 occur, naturally. Thus a potential for trace metals in discharges from mines is always present.

The acid produced in mine drainage as a result of pyrite oxidation causes the other associated sulphides and sulphosalts either to oxidation or exposing to conditions conductive to their breakdown with the release of diverse toxic elements to the aqueous environment. Under certain proper physio-chemical conditions, any of the ions shown in Table 5 may be present in the drainage. For example, the breakdown of chalcopyrite ($Cu Fe S_2$) would release Cu and Fe. However, Au, Ag and Se associated with chalcopyrite may also be present. The metal ions in the drainage will depend on the sulphide and sulphosalts present, the materials associated with the strata (clays and minerals) and the chemical characteristics of the metal ion and water. The solubility of the mineral phase is a primary factor.

Further work is in the progress in the water pollution abatement due to acid mine drainage with particular reference to trace metals removal. Health statistics are also being obtained.

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