MINE-RELATED AND NATURAL ACID SEEPS AT GIBBONS CREEK LIGNITE MINE, TEXAS[1]

by

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Abstract

Natural acid seeps are a common feature in lignite-bearing areas such as Gibbons Creek lignite mine in east-central Texas. The seeps are a surface manifestation of part of the natural sulfur cycle and are reflected in the numerous "sulphur" creeks and springs. Sulfuric acid is generated when iron disulfide (pyrite) oxidizes and there are insufficient bases to neutralize it. Pyrite is common in lignite-bearing rocks because the anaerobic conditions favorable to the formation of lignite were also favorable to the chemical reduction of sulfate to sulfide. The source of the sulfate was sea water, which was part of the depositional environment of the Gibbons Creek lignites 35 million years ago. At that time, the shoreline of the Gulf of Mexico was in the vicinity of the Gibbons Creek deposit. Mine-related acid seeps form in the same way that natural seeps do, except on an accelerated time scale.

Introduction

Maps of lignite-bearing areas abound in "sulphur creeks" and "sulphur springs." The area around Texas Municipal Power Agency's (TMPA) Gibbons Creek lignite mine in east-central Texas is no exception. There is a Sulphur Creek that is a tributary to Gibbons Creek and there are numerous sulphur springs. The most notable of these is Piedmont Springs, which in 1860 was a popular health spa featuring a four-storey hotel with over 100 rooms (*Handbook of Texas Online*, 2007).

The presence of sulfur in surface and ground waters is a reflection of the local geology. The Gibbons Creek lignites belong to the Manning Formation (also known as the Jackson-Yegua) of the Tertiary late Eocene. At that time (approximately 35 million years ago), the Gibbons Creek area was near the ancient shoreline of the Gulf of Mexico (O'Keefe at al., 2005). The Manning sediments were either deposited directly in a marine environment or were submerged soon afterwards in marine transgressions (Yancey, 1997). Either way, these sediments, including the peat and other vegetation that was to become lignite, became saturated with sea water – the source of the sulfur.

The sulfur cycle

Sulfur is one of the major elements that is recycled by natural processes. Sulfur exists mainly in the form of the element (S^0), the sulfide (S^{2-}), or the sulfate (SO_4^{2-}). Under reducing conditions, where oxygen is absent (or more strictly, where there is an environment of electron donors), sulfur exists as sulfide or as the

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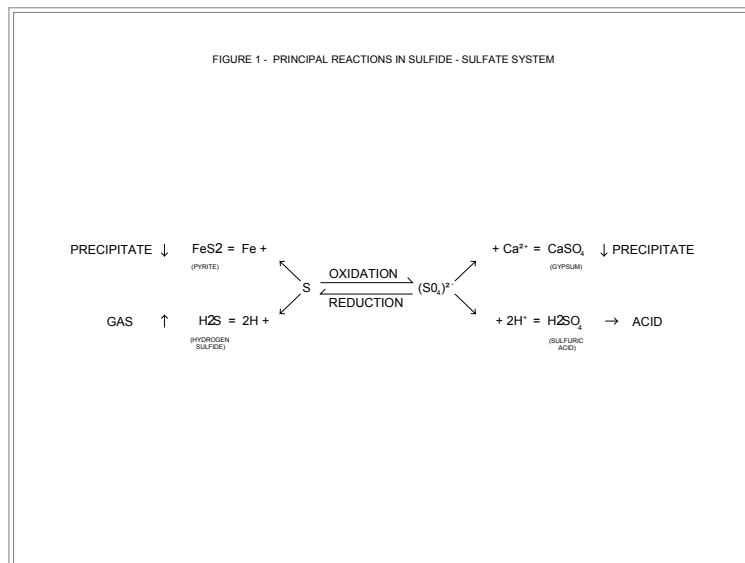
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element itself. Under oxidizing conditions, where oxygen is available (or where there is an environment of electron acceptors), sulfur exists predominantly as sulfate. A good place to start investigating the sulfur cycle is the sea.

The primary constituents of sea water are sodium and chloride ions, together totaling 29.54 g/L (Appendix 1). The chloride ion is very stable and very soluble and therefore remains in solution (unless there are extreme evaporative conditions when it will eventually precipitate in combination with sodium ions as halite – sodium chloride). But, sea water also has an appreciable content (2.65 g/L) of the sulfate ion – $[SO_4]^{2-}$. Sulfate is less stable than chloride because of its susceptibility to be reduced to sulfide. Once reduced, the sulfide ion becomes very insoluble if combined with certain metal cations, such as iron. The resulting compound that is precipitated is iron disulfide (FeS_2), commonly known as pyrite (fool's gold).

The reduction of sulfate to sulfide is particularly favored during decomposition of organic materials in anaerobic conditions (Goldhaber and Kaplan, 1982). Where there is a shortage of atmospheric oxygen, as in water-logged soils and swamps, microbes will in effect scavenge oxygen from sulfates. Such conditions existed in the peat swamps that led to the formation of the Gibbons Creek lignites (O'Keefe et al., 2005). Thus, pyrite tended to precipitate in the ancient accumulations of decaying vegetation and even along root-channels in ancient soils (Horbaczewski, 2007). If there were insufficient metal ions to pair with, the sulfide ions bonded with hydrogen (from water) to form hydrogen sulfide (H_2S). This gas is commonly detected around freshly-exposed lignite seams.

The opposite reaction in which sulfide is oxidized to sulfate, is favored by an abundance of oxygen. Under these conditions, oxygen atoms attach to sulfide to form sulfate ions (this is a highly simplified account; for details of the electron transfer mechanisms involved, the reader is referred to Evangelou, 1995, pp 139-148). Iron disulfide breaks down to release iron, which oxidizes further to "rust" – a complex of hydrated iron oxides and hydroxides. The remaining sulfide becomes oxidized to sulfate ions. These remain in solution unless there is some cation with which they may form an insoluble compound and precipitate out. Sodium, potassium, and magnesium are not good candidates because they form very soluble sulfate salts. But calcium combines with sulfate to form the relatively insoluble calcium sulfate or gypsum (the solubility of gypsum is about 2.6 g/L – U.S. Dept. of Agriculture, 1954). So if there is enough calcium, gypsum will typically form and precipitate out of solution.

For the purposes of this discussion, the main reactions that have been discussed above may be simplified to the following form (Figure 1):

FIGURE 1 - PRINCIPAL REACTIONS IN SULFIDE - SULFATE SYSTEM

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Oxidation of pyrite in geological and mine spoil materials

The natural oxidation of pyrite in geological formations occurs from the surface downwards. Its progress is determined by the rate at which atmospheric oxygen and rain water can advance through the resisting geological formations. A fractionation of sorts develops resulting in a geochemical profile. One of the most striking features of this profile is an oxidation-reduction ("redox") boundary at a depth of about 27 feet (Figure 2). Geological materials above the redox boundary have a characteristic tan, brown, or red coloration indicative of oxidized (ferric) iron (Fe^{3+}) oxy-hydroxides and decomposed organic matter. Geological materials below the redox boundary have dark greenish-gray colors due to the presence of reduced (ferrous) iron (Fe^{2+}) and undecomposed organic matter, including lignite seams.

Figure 2 – Overburden profile in highwall and detail of redox boundary (numbers painted on highwall denote distance in feet above top of lignite seam)

The redox boundary should really be called the redox plane because it is a surface with a lateral extension. It generally parallels the surface topography and in dip-oriented mine pits can be seen to transgress geological units. The depth of the boundary varies according to the permeability of the overlying geological materials but ranges from 15 feet to 40 feet, with an average of about 27 feet.

The fact that the redox plane parallels the surface topography and transgresses lithological units and even displacement faults has deeper significance. It indicates that the plane is a dynamic rather than static feature. It is a feature that is continuing to migrate downwards over geological time. The fact that it occurs at a relatively constant depth (with minor variations for permeability) indicates that it is a feature that is in equilibrium with the surface. It is presumably in equilibrium with the rate of erosion of the surface over geological time – when a foot of material is eroded from the surface, the redox boundary gradually responds by migrating a foot downwards. The implications are geochemically profound. Fresh pyrite, previously protected below the redox boundary, becomes exposed to oxidation as the boundary passes over it. Thus, the natural oxidation process never ceases.

Chemical analysis of the geological materials above the redox boundary reveals additional evidence of ongoing oxidation. Pyrite is almost completely absent above the redox boundary whereas it is completely unoxidized below. The same applies to lignite seams. Above the redox boundary, they are generally degraded through oxidation. Mining does not start at the "outcrop" of lignite seams. It starts at the "subcrop" where the seams intersect the redox boundary (the exact location of subcrops has to be found by "fence drilling" across the geologist's projection of these intersections). Other geochemical indicators of oxidation above the redox boundary are an accumulation of sulfate and a zone (or sometimes several zones) of very low pH indicating active oxidation of pyrite. The oxidized zone above the redox boundary does not necessarily signify that all the pyrite has been oxidized – just that it is an ongoing process. On the other hand, the presence of unoxidized pyrite and lignite, and the absence of sulfate accumulations confirm that oxidation reactions have not started below the redox boundary.

Oxidation in fresh mine spoils follows a similar path but at a faster rate. Geochemical analysis of cores of spoil materials up to 30 years old at another mine in central Texas showed a rapid advance of an "oxidation front" characterized by low pH values (less than 4.0) and high sulfate to pyritic sulfur ratios (Morrison-Knudsen Co., 1987). The data indicated that most of the pyrite in those moderately sandy spoil materials had been oxidized to a depth of 29 feet within 20 years (Horbaczewski and Van Ryn, 1988). The same process appears to be occurring at Gibbons Creek mine. Most of the oxidation occurs in spoil material and is not evident at the surface. However, there are a few instances where some of the groundwater emerges in the form of a seep (Figure 3). A total of nine seeps have been identified at Gibbons Creek (Table 1). The chemical characteristics of these are presented in Appendix 3.

Table 1 - Mine acid seeps and mitigation status

Seep ID	Description	Size (acres)	Start of monthly monitoring	Pre-mitigation		Installation of mitigation structure
				Flow	pH	
1	North of A2 boxcut	1.0	Aug. 2005	< 1 gpm	2.7 - 4.2	July 2008
3	West of Pond 13A	0.50	Aug. 2005	< 1 gpm	3.2 - 7.4	October 2008
5	Above Pond B1P-2	0.25	Aug. 2005	3 gpm	3.1 - 7.4	July 2007
7	Pond SP-13 spillway	0.25	Aug. 2005	25 gpm	3.1 - 7.9	March 2008
8/9	Toes along A3 Drain 2	3.0	Aug. 2005	40 gpm	2.6 - 7.3	December 2008
13	Inlet to Pond B1P-10	0.10	Sept. 2006	30 gpm	5.6 - 7.5	October 2008
14	Below Pond 6A	0.10	Aug. 2006	< 1 gpm	5.9 - 8.0	November 2008
28	From B1 to G1 block	0.25	Aug. 2005	< 1 gpm	3.3 - 6.5	July 2007
29	Near north haul road	0.25	May 2007	< 1 gpm	3.2 - 3.5	To be determined
	Total	**5.7**				

Figure 3 - Map of mine-related and natural acid seep locations at Gibbons Creek lignite mine

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Discussion of two case studies

The following discussion compares the effects of pyrite oxidation in two case studies taken from the Gibbons Creek lignite mine area. The first case study deals with natural oxidation of pyrite in the Peach Creek drainage system, which was never disturbed by mining. The second case study deals with mine-related oxidation of pyrite as seen at Seep 1.

Case study of natural acid drainage: Peach Creek

The Peach Creek drainage system covers an area of 5,162 acres (just over 8 square miles) east of the reclaimed Gibbons Creek mine (Figure 3). Parts of this area were to be mined so it was studied intensively for baseline conditions in the early 1990s. These studies included a water chemistry survey of Peach Creek and its tributaries. In 1995, TMPA decided to switch fuels from the relatively high-sulfur local lignite to low-sulfur "compliance" coal from the Powder River Basin in Wyoming. The Peach Creek area has therefore remained undisturbed.

The results of field and laboratory pH readings taken at various locations in the drainage system are summarized in Table 2. It should be noted that there was little rainfall in this period so that the water flowing in the main channel and its tributaries was not rainfall runoff but base flow fed by groundwater discharge.

Table 2 – pH readings in the Peach Creek system

Sampling site	pH		
	06/10/94	**07/7/94**	**07/29/94**
1	4.6 (f) --------	4.7 (f) 4.4 (l)	4.6 (f) 4.4 (l)
2	6.3 (f) 6.3 (l)	---------	---------
3	3.8 (f) 3.8 (l)	---------	---------
4	6.6 (f) 7.1 (l)	---------	---------
5	6.0 (f) 6.2 (l)	---------	---------
6	6.0 (f) 5.9 (l)		
7	3.9 (f) --------	3.0 (f) 3.7 (l)	4.2 (f) 4.0 (l)
8	4.1 (f) --------	4.0 (f) 4.0 (l)	4.0 (f) 3.7 (l)
9	3.5 (f) --------	3.4 (f) 3.4 (l)	3.5 (f) 3.3 (l)
10	5.2 (f) 5.8 (l)	---------	---------
11	5.8 (f) 6.5 (l)	---------	---------
12	4.0 (f) 4.0 (l)	---------	---------
13	6.0 (f) 6.4 (l)	---------	---------
14	6.4 (f) 6.7 (l)	---------	---------
15	2.1 (f) 3.8 (l)	3.4 (f) 3.6 (l)	2.9 (f) 3.5 (l)
16	4.1 (f) 5.9 (l)	6.1 (f) 6.1 (l)	4.8 (f) 6.4 (l)
17	3.7 (f) 5.1 (l)	4.9 (f) 4.4 (l)	4.4 (f) 4.5 (l)

Notes: (f) – field determination of pH;
(l) – laboratory determination of pH.

The variations in pH reflect variations in base flow (Table 3). Marston Environmental, Inc. (2004) demonstrated a relationship of pH to flow for the nearby Rock Lake Creek North drainage system (just north of the Gibbons Creek mine area – Figure 3). Marston's conclusion was that low pH values at times of base flow were due to acid seepage. The higher pH values at times of higher flow were attributed to rainfall dilution.

Table 3 – Field pH vs. Flow at Rock Lake Creek North

Table 3 – Field pH vs. Flow at Rock Lake Creek North
(April 1979 – January 1982)

Flow	< 0.1 cfs	0.1-1.0 cfs	1.0-33.0 cfs
Average pH*	3.7	4.0	5.2
No. of readings	11	8	22

Source: Marston Environmental, Inc. (2004) – Table 1.1, based on data from
Morrison-Knudsen Company, Inc. (1986)
Note: *Simple arithmetic average.

Analyses of the major cations and anions at the 17 sampling sites in Peach Creek are presented in Appendix 2. In addition to the variations in pH, the data show considerable variation in other parameters that may be due to dilution. One way to remove these effects is to use ratios of parameters instead of the absolute values.

The ratio of SO_4 / Cl indicates the degree of oxidation of pyrite to sulfate. The ratio of these two ions in sea water (as measured in chemical equivalents) is 0.1 (Appendix 1), indicating a preponderance of chloride ions. However, chloride ions are soluble and relatively easily leached out by rainwater from geological materials, resulting in enrichment in sulfate ions. As a result, where there is no evidence of pyrite oxidation, i.e., when pH values are near-neutral (e.g., above pH 6.0 as at sample points 2, 5, 11, 13, and 14), drainage waters show a SO_4 / Cl ratio that is generally below 1.0. Where there is evidence of pyrite oxidation (at pH less than 4.0, as at sample points 3, 7, 9, and 15) the SO_4 / Cl ratio is generally above 2.0).

The ratio of Ca / Mg shows a ten-fold increase in relation to sea water. Most of this is probably due to the release of calcium through weathering of plagioclase and other minerals. There may also have been a decrease of magnesium through adsorption into clay minerals, such as montmorillonite.

The ratio of Na / Cl in sea-water is 0.86. The ratio in the Peach Creek waters is the same or somewhat higher indicating a slight enrichment in sodium, presumably released from the weathering of some sodium-bearing minerals, such as plagioclase feldspars.

The ratio of Ca / SO_4 in sea-water is 0.4. The ratio in the Peach Creek water is somewhat higher than this but is generally below 0.8. This means that there is almost enough calcium to balance the sulfate ions. Sample points 3 and 4 are anomalously high and may have been contaminated. Both are near a highway and may have received limestone from road construction projects.
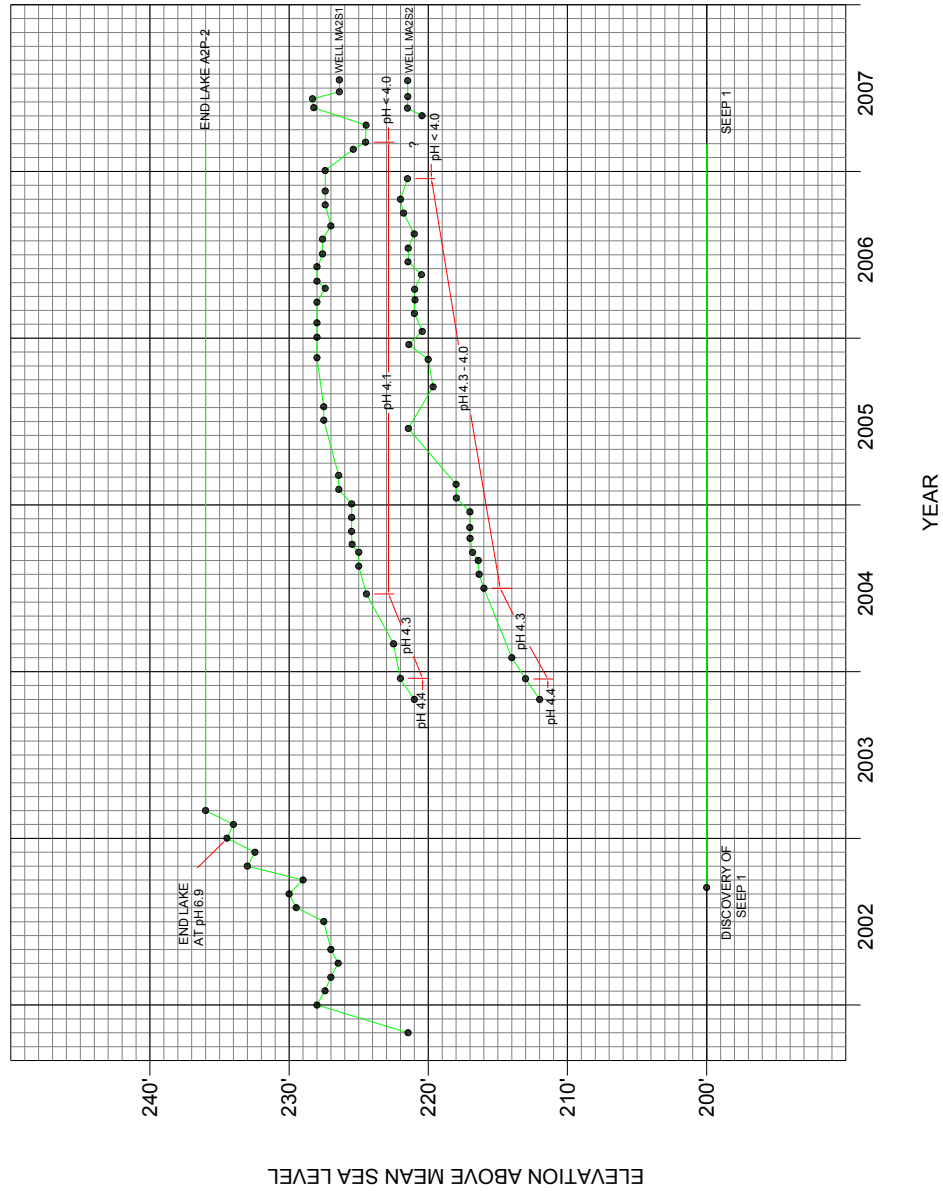
Case study of mine-related acid drainage: Seep 1

Seep 1 is one of the best studied mine-related seeps at Gibbons Creek mine. This seep was discovered on September 5, 2002, and five groundwater monitoring wells were established in the summer of 2003. Monitoring started on a quarterly basis and since June 2004 has been on a monthly basis (data for the most recent sampling are presented in Appendix 4). A remediation plan was prepared by Marston Environmental, Inc. (2007).

The seep appears to be driven by the head of water exerted by end lake A2P-2, which is 2,500 feet away in a straight line. The elevation of the water in the end lake is 236 feet above mean sea level (msl) and the elevation of the seep is approximately 200 feet above msl. The two spoil wells in between the end lake and the seep area show water levels at approximately 227 above msl (well MA2S1, 800 feet from the end lake) and 222 feet above msl (well MA2S2, 1,650 feet from the end lake). The horizontal distance between well MA2S2 and Seep 1 is approximately 850 feet.

At the time Seep 1 was discovered, end lake A2P-2 was still in the process of construction and of filling with rainfall runoff (Figure 4). On September 5, 2002, end lake A2P-2 was at 229.7 feet above msl. By the end of February 2003, the end lake had reached its design elevation of 236 feet above msl. Water levels in the two spoil wells MA2S1 and MA2S2 had stabilized by the end of 2005, indicating a lag of about three years behind the end lake. The fluctuations since the end of 2005 appear to be related to seasonal effects.

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FIGURE 4 - WATER LEVEL ELEVATIONS IN RELATION TO SEEP 1

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Previous studies on other end lakes in the same mine block (Horbaczewski, 2001) showed that there is no evidence of appreciable mass movement of water out of end lakes into the spoil (or vice versa out of the spoil into the end lakes). The resaturation of the spoil appears to occur through the downward migration of rainfall that infiltrates relatively easily into these sandy spoil materials. This is in contrast with pre-mine conditions in which deep percolation of rainfall was impeded by horizontal barriers such as relatively impermeable soil claypans and geological units. Although end lakes do not serve as sources of water to any appreciable extent, it appears that they do control the piezometric surface of the water table in the spoil.

This suggests that the chemistry of the spoil and seep groundwater will be more dependent on the local chemistry than on the chemistry of the water in the end lake. This appears to be borne out by the data. The water in end lake A2P-2 showed a chemical evolution driven by limestone washed in from reclaimed minesoils in the drainage area. The originally very acid pit water (pH 2.9 in September 2000) had been completely neutralized by the end of 2002 (Horbaczewski, 2006). However, the groundwater in the spoil has shown a gradual increase in acidity (decline in pH) since monitoring of the wells started in September 2003. At that time, the pH was 4.4, and by the end of 2006, it was near pH 4.0.

It appears that nature is mimicking in the spoil the same process that occurs naturally. The upper part of the spoil is being oxidized and equilibrium is becoming established. With time, a redox boundary will probably form above, or at, the current water-table. Even now, pyritic materials that had been oxidizing before the rise of the spoil water-table have probably become stable in their new reducing environment and will not oxidize further. At well MA2S1 the water table on July 25, 2007, was at 226.5 feet above msl and the surface of the ground is at 293.7 feet above msl, so the water table was 67.2 feet below the surface of the ground. At well MA2S2 the water table on July 25, 2007, was at 221.6 feet above msl and the surface of the ground is at 298.5 feet above msl, so the water table was 76.9 feet below the surface of the ground.

Pyritic materials in the oxidizing zone above the water-table will proceed rapidly to complete oxidation. The evidence for this is the gradually falling pH of wells MA2S1 and MA2S2 (Figure 4). At the start of monitoring on September 29, 2003, both wells showed a pH of 4.4. The pH of both declined at the same rate, dropping below 4.0 at MA2S2 on December 15, 2006, and at MA2S1on February 5, 2007. Once the supply of pyrite becomes exhausted, the production of acid will cease. In sandy materials, this will occur relatively quickly because the rate of rainfall percolation will be faster and because sandy formations represent high energy geological environments of deposition with little pyrite formation in the first place.

Conclusions

The following conclusions may be drawn about natural and mine-related acid seeps at Gibbons Creek mine:
- The presence of natural "sulphur" seeps in lignite-bearing areas has been recognized for well over a century.
- Undisturbed geological formations show a "redox" boundary representing the depth to which oxidation processes have penetrated from the earth's surface.
- At Gibbons Creek mine, the redox boundary is on average at 27 feet below the surface.
- The redox boundary is not a static feature; it is dynamic and constantly migrating to keep pace with erosion.
- This means that fresh pyritic material is always oxidizing even under natural conditions.
- This explains the presence of the "sulphur" springs and of the very acid base flow in local drainages undisturbed by mining.
- The process of mining and placement of pyritic material that was below the redox boundary in a new location above the redox boundary starts a new oxidation cycle.
- Because of the unconsolidated nature of spoil, rainwater infiltration and percolation is very rapid compared to undisturbed geological formations.
- Oxidation of pyrite placed near the surface therefore occurs very rapidly.
- A related study (Horbaczewski, 2007) shows that within a few decades, pyrite may be completely exhausted from the top 20-30 feet of spoil.

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