

USE OF PASSIVE ANOXIC LIMESTONE DRAINS TO ENHANCE PERFORMANCE OF ACID DRAINAGE TREATMENT WETLANDS¹

by

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Abstract. Constructed wetlands for treating acid drainage are preferred, low-cost, alternatives to conventional treatment. Drainage with high Fe (e.g., >50 mg/l) and zero alkalinity has not been amenable to treatment with wetlands alone, primarily due to Fe hydrolysis and resultant low pH which requires chemical treatment. Anoxic limestone drains (ALDs) increase the alkalinity of seepage that is then routed to a constructed wetlands. Increased alkalinity buffers the wetlands system from pH decreases, and enhances the effectiveness of wetlands treatment. The Tennessee Valley Authority has modified two low-pH constructed wetlands with ALDs. Results indicate retrofitted wetlands function at efficiencies meeting effluent limitations without chemical treatment. Designs of ALDs are site-specific, but generically consist of an excavated seepage-interception trench backfilled with crushed limestone covered with plastic and clay soil. Dissolution rates of limestone in operational and simulated ALDs were experimentally measured in attempts to estimate design parameters and longevity of an ALD. Potential problems with ALDs include structural and hydraulic stability, plugging due to reaction products within the ALD, and inadequate design and installation.

Additional Key Words: Tennessee Valley Authority, constructed wetlands, Alabama, Tennessee, alkalinity

¹Paper presented at the 1991 National Meeting of the American Society for Surface Mining and Reclamation, Durango, CO, May 14-17, 1991.

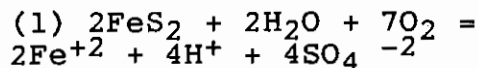
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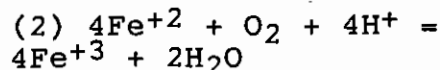
Introduction

Staged constructed wetlands are an effective alternative to conventional means of treating acid mine drainage (AMD) (Brodie 1990a,b,c; Hedin 1989). Wetlands efficiencies are related primarily to flow rates, pH, and concentrations of dissolved oxygen, iron, manganese, acidity, and alkalinity. Foremost are the relationships among influent dissolved oxygen, iron, and alkalinity concentrations, and influent/effluent pH.

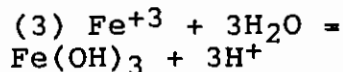
Metals removal mechanisms in wetlands are variable and numerous, but it is probable that microbially-catalyzed Fe oxidation and hydrolysis are major reactions responsible for iron removal in acidic waters in aerobic wetlands systems (Faulkner and Richardson 1990). These reactions may dictate the ultimate success or failure of a wetlands system because of their acidity-producing natures, summarized below.



(Pyrite oxidation)



(Ferrous oxidation)



(Ferric hydrolysis)

Reactions 1-3 occur all in the backfill, but only reaction 3 acts to increase acidity in a wetlands. If sufficient buffering capacity is not present in the acid drainage, pH will decrease. Several aerobic wetlands systems, although removing substantial amounts of iron and manganese have had influent pH near 6.0 and effluent pHs of less than 3.0 (Brodie 1990a,c, 1991a,b; Kepler 1989). Table 1 shows typical pH decreases in TVA's 006 Kingston Fossil Plant constructed wetlands in Roane County, TN. This 2.3-acre system receives, on average, 379 gpm of acid seepage from an ash disposal area with an influent pH of 5.5 and total Fe of 170 mg/L. Effluent from the wetlands system has a pH of 2.9 and total Fe of 83 mg/L; thus the hydrolysis of about 87 mg/L of Fe is responsible for the decreased pH.

Numerous methods for treating acid drainage have been used to meet effluent standards; however these methods are expensive and require constant attention to maintain compliance with effluent limitations. Also, many chemical additives have an adverse impact on aquatic biota in receiving streams. Limestone treatment has many advantages over conventional chemical treatment including lower sludge production, lower cost, and less potential for overdosing. However, the use of limestone rock as a

buffering agent was abandoned because, in an oxidizing environment, iron hydroxides coat the limestone surface and inhibit or eliminate the dissolution of the limestone, thus preventing effective buffering.

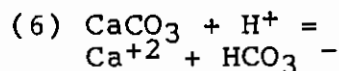
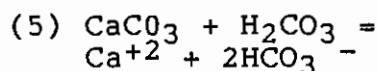
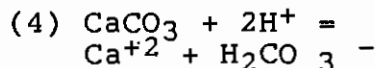
In 1988, the Tennessee Division of Water Pollution Control (TWPC) constructed and evaluated prototype passive anoxic limestone drains (ALDs) at 2 mine sites in Tennessee (Turner and McCoy 1990). At the same time, Tennessee Valley Authority (TVA) recognized a correlation between alkalinity and dissolved oxygen of wetlands influent, and the relative success of 11 TVA constructed wetlands, and conducted several relevant bench-scale studies.

Results of a TVA study identified a correlation between wetlands influent alkalinity and effluent pH, Fe, and Mn. A possible "accidental" roadbed originated anoxic drain (AROAD) was identified at the TVA Impoundment 1 constructed wetlands, which treats high-Fe (100 mg/l) acid drainage emanating from a fine-coal refuse disposal area at the Fabius Coal Preparation Plant site in northeastern Alabama (Brodie 1990a,c). Air photo investigations showed that a 1400-ft long, 25-foot high earth dam was constructed in 1974 over an existing coal haul road built of crushed rock that was presumably obtained from the local Monteagle formation, a high CaCO_3 , oolitic limestone (see Figure 1). Although this limestone roadbed was not intended for water quality

enhancement, it is hypothesized that it is pretreating acid drainage seeping through the dike by adding alkalinity (300 mg/l) and buffering capacity so that the wetlands is producing compliance-quality discharges. TVA is currently drilling and sampling the dike, and conducting monitoring well sampling to determine the nature and condition of the AROAD (Brodie and Garvich 1991).

Several constructed wetlands in Tennessee and Alabama have now been enhanced by the addition of anoxic limestone drains (ALDs) as the initial component of a staged treatment system. The ALD, which consists of a shallow, limestone-filled trench excavated into the spoil and sealed from the atmosphere, passively introduces buffering capacity, as alkalinity, into the acid drainage. Changes in pH due to acid production from Fe hydrolysis in the wetlands are buffered due to the high alkalinity in the influent.

It is assumed that the following reactions are applicable within the ALD:



Equation 4 reacts limestone with acidity (at pH < 6.4) present in the mine drainage to form free calcium and dissolved carbon dioxide (carbonic acid). The carbonic acid further reacts with

limestone in equation 5 to produce alkalinity. As reactions 4 and 5 act to increase pH above 6.4, equation 6 becomes the major reaction where bicarbonate is the dominant dissolved CO₂ species (Cravotta et al 1990).

Systems observed to date indicate that if the ALD is properly constructed, limestone in the ALD will not armour with Fe or Al precipitates and alkalinity can be significantly increased in the ALD effluent. This paper discusses the design, construction, and operational results of a few ALDs and some preliminary research on the utility and longevity of this potentially inexpensive component of a long-term acid drainage treatment system.

Discussion

Preliminary Considerations

The preliminary question is whether an ALD is necessary or beneficial to an existing or proposed wetlands treatment system. Hydrology and water-quality characteristics primarily determine the need for and feasibility of an ALD. Tables 2-5 show the relationships among influent/effluent pH, alkalinity, Fe, and Mn in TVA's 13 constructed wetlands. Table 2 shows that effluent pH of constructed wetlands is directly related to influent alkalinity. Only the 4 systems with zero alkalinity in the inflow fail to meet the NPDES permit limitation of 6.0. TVA's wetlands systems having influent alkalinity loading

greater than 4 g/day/m² (GDM) (corresponding to 73-113 mg/L alkalinity, as CaCO₃) always produce complying pH (i.e., > 6.0) discharges, even with Fe contents as high as 110 mg/L. However, where influent Fe concentrations exceed 18 mg/L, low-pH effluent has been observed in cases of zero influent alkalinity (Brodie 1990a). Table 3 supports the idea that high-Fe concentrations are causative of low wetlands effluent pH. Table 4 suggests that Fe removal is not related to alkalinity concentrations. Table 5 shows a positive relationship between Mn removal and influent alkalinity. However, Fe removal occurs more rapidly at high pH (e.g., > 5.5) and is very slow at low pH (< 3.0).

Previously suggested guidelines have been revised to determine the utility for an ALD (Brodie et al 1990).

Case 1: Alkalinity > 80 mg/L, Fe > 20 mg/L; an ALD may be beneficial, but a wetlands system based on previously reported chemical loading rates (Brodie 1990b; Hedin et al 1990) is probably adequate.

Case 2: Alkalinity > 80 mg/L, Fe < 20 mg/L; an ALD is not necessary, only an adequately designed wetlands.

Case 3: Alkalinity < 80 mg/L, Fe > 20 mg/L; an ALD is recommended and probably necessary as the initial stage in a constructed wetlands system.

Case 4: Alkalinity < 80 mg/L, Fe < 20 mg/L; an ALD is recommended, but not necessary.

Case 5: Alkalinity ≤ 0 , Fe < 20 ; an ALD will likely be necessary as Fe approaches 20 mg/L.

Case 6: Dissolved oxygen > 2.0 mg/L or Fe oxidizing conditions exist (e.g., pH > 6.0 , eH > 100 mv. An ALD should not be installed because of probable limestone armoring.

Case 7: Ferric iron present in appreciable concentrations in the unaerated seep sample; an ALD may not be appropriate due to potential limestone armoring.

Additionally, because the ALD is generally associated with a staged wetlands treatment system, preliminary considerations for constructed wetlands, such as available area, geology and topography, and the availability of soil and limestone must be regarded (Brodie 1988).

In order for an ALD to perform successfully, unaerated mine drainage must come into contact with the buried limestone. Discrete seeps or boils from embankments are generally good starting points for excavation of the ALD into the backfill. Non-point seep areas may require more innovative means of collection, such as specialized rock drains or the construction of an embankment to contain the ALD. Underground mine adits could be sealed and flooded, routing the drainage via a pipe to an ALD, or a mine adit could be backfilled with limestone and thereby delineate the boundaries of the ALD.

Maximum expected flow through the ALD should be determined to prevent hydraulic failures and disruption of the sealed nature of the ALD. Several methods exist for designing underdrains and French drains, which are similar in construction to ALDs; but ideally, maximum flow should be determined or estimated from actual flow measurements over an extended time period. Alternatively, flow can be predicted from various hydrologic methodologies, although this may be difficult and expensive due to the need for contributing area delineation and characterization of the geologic materials. Because of the relative low cost of an ALD compared to conventional treatment systems, an oversized ALD is recommended to ensure hydraulic stability and to maximize longevity. In fact, desired longevity of the ALD will likely dictate a design size far in excess of the hydraulic design requirements. A suggested crude, but conservative, method for hydraulically designing the ALD is to calculate infiltration from the contributing drainage basin at the point source of the seepage and assume an attenuation factor based on the mine fill characteristics. As an example, annual infiltration over a 100-acre watershed might be estimated at 12 inches, giving an average flow from groundwater of about 60 gpm. Assuming all flow is from the seep and high flow is 3 times the average flow, an

ALD might be designed to handle 180 gpm.

Another important preliminary consideration for ALD construction is water quality. The success of a wetlands system to meet effluent limitations is directly and primarily related to influent pH, acidity, alkalinity, Fe, and Mn concentrations.

Alternatively, dissolved oxygen and oxidation-reduction potential (ORP) are critical for the proper operation of an ALD. All waters that would be routed through an ALD should be sampled and analyzed for the above mentioned parameters. Samples should be collected directly from the seep via methodologies precluding sample aeration (e.g., peristaltic pumps, large syringes, etc.). Dissolved oxygen (DO) and ORP must be field measurements. Fe^{+2} and Fe^{+3} are good indicators of ORP (and possible armouring). If samples are immediately acidified with HCl (or another non-oxidizing acid) Fe^{+2} can be determined accurately in the lab.

Design and Construction

The basic design of an ALD is shown in Figure 2. The ALD consists of an open, unlined trench or excavation backfilled with gravel-sized, crushed, high- $CaCO_3$ limestone. The limestone is covered with plastic to preclude oxygen infiltration and CO_2 exsolution. It is desirable to place geotechnical fabric over the plastic for protection of the plastic from puncture by

equipment or manpower. A clay soil is then placed over the fabric.

As discussed above, adequate hydraulic design and subsequent oversizing is critical for the proper operation and long-term stability of an ALD. Maximum and average flow should be estimated and the dimensions of the ALD then determined in conjunction with consideration of other site restraints (e.g., steep topography, shallow bedrock, multiple seeps). Because of the infinite number of potential shapes and configurations of an ALD, it is beyond the scope of this paper to suggest guidelines for shapes and dimensional ratios of an ALD.

Side slopes of the excavation are not critical to the operation of the ALD and are best made near vertical to facilitate construction. High-calcium limestone (i.e., $\geq 90\%$ available $CaCO_3$) should be used. TVA and TWPC have used 3/4 to 1-1/2 in crushed rock in existing ALDs because of its high hydraulic conductivity, large surface area, and ready availability; however, a different gradation, a well-graded mixture, or a layering of several grades of rock may be suitable. Larger rock size or more dolomitic (i.e., $CaMgCO_3$) stone is not recommended due to the loss of surface area in the ALD with the large rock and the lower reactivity of dolomite. The depth of limestone backfill should be determined by (1) the need to accommodate the maximum probable flow, (2) the desired longevity of the ALD,

and (3) a comfortable safety factor for both (1) and (2). Desired longevity is discussed below. TVA's and TWPC's ALDs have contained limestone depths of 2 to 5 ft. Because the very lowest portion of the backfill may be rendered ineffective due to its tendency to embed in the bottom of the excavation, about 6 inches additional depth of limestone should be included to account for this loss.

A minimum of 2 ft of soil cover should be placed and compacted over the plastic and fabric covered limestone backfill. Soil should be sufficiently impermeable to oxygen migration. If available and practicable, clay soil is recommended. Alternatively, a clay, clayey-silt, or silty-clay loam should be adequate. The final cover should be slightly crowned and protected with erosion control fabric or adequately waterbarred to prevent erosion. Crowning will allow for subsidence of the ALD over the years due to limestone dissolution. Ideally, the crowned ALD should be riprapped or revegetated with species such as sericea (Lespedeza cuneata) or crownvetch (Coronilla varia), which will discourage the establishment of trees whose roots could penetrate the ALD and render it ineffective.

Prototype installations of ALDs have used 5-mil plastic and/or filter fabric covered with clay soil to seal the limestone from the atmosphere (Turner and McCoy 1990). More recent ALDs have used 20-mil plastic or double layers of

10-mil plastic, which is significantly less expensive than 20-mil and is more readily available. Geotechnical fabric should be of quality sufficient to protect the integrity of the plastic from puncture under loads from equipment and workers.

An additional design recommendation is the use of an oxidation basin immediately after the seep discharges from the ALD and prior to routing that flow into a constructed wetlands. The purpose of an oxidation basin is aimed at high Fe influents (e.g., >50 mg/L). The basin allows the newly aerated and highly alkaline ALD discharge water to react and precipitate the majority of its Fe load, which can be achieved physiochemically and without the use of a constructed wetlands. The U.S. Bureau of Mines has suggested a general empirical guideline which states that about 50 mg/L Fe can be removed in a wetlands cell before reaeration is required (Kleinmann 1990). A basin will greatly enhance the efficiency and lifespan of a downstream constructed wetlands as the basin can be dredged, thus preventing excessive precipitate buildup in the wetlands. Such a pond needs simply to be designed according to existing guidelines with considerations of regulatory requirements, desired capacity, and maintenance. Alternatively, a modified marsh-pond cell with a major portion devoted to deep (3-6 ft) water might be more applicable to acid drainage with low to moderate Fe concentrations.

Results

To date 17 ALDs have been installed in Alabama and Tennessee with 3 more planned for 1991 installation (Brodie 1991a; Turner 1991). At least 1 ALD has been installed at a wetlands system in Pennsylvania (Meehan 1991). Tables 6-8 show typical pre-ALD and post-ALD water quality for TVA's Impoundment 4 constructed wetlands ALD system, which was installed in May 1990. As shown in Table 6, the ALD has successfully increased alkalinity and enabled the wetlands to decrease Fe and Mn concentrations to compliance levels; wetlands effluent pH rose from 3.1 to 6.3, and Fe decreased to <2.0 mg/l and Mn <2.0 mg/l. The Impoundment 4 wetlands influent (i.e., the ALD effluent) had an average pH of 6.3 and concentrations of 85 mg/L Fe and 17 mg/L Mn (see Table 7) before the ALD. After the ALD was installed, ALD effluent Fe and Mn concentrations have appeared to decrease. This phenomenon could indicate precipitation of Fe and Mn within the ALD, but is more likely a reflection of the general improvement of the seep-water quality over the past several years as a result of land reclamation.

ALD costs are associated with the design and engineering, manpower, equipment, materials and any operational or maintenance requirements. Table 9 shows the costs of an ALD installed at TVA's Fabius Coal Preparation Plant site in Jackson County, AL to treat up to 100 gpm of acid drainage flowing into the Impoundment 4

constructed wetlands (Brodie 1990a,c, 1991a).

The total installation cost of the ALD was about \$19,000. This compares to annual costs of about \$20,000 for NaOH and \$10,000 for operation and maintenance associated with conventional treatment of the Impoundment 4 flow from October 1985 to May 1990. Thus, the payback period for the costs associated with the ALD was about 7 months. The TWPC reported total costs for two ALDs at mine sites in TN of \$8000 and \$6000 for limestone, labor, and heavy equipment (Turner and McCoy 1990). The significant differences between these costs and TVA's costs lie in the cost of rental equipment, which would generally be available at a mining operation. Thus, TWPC's reported costs may be more representative of what a coal operator might incur for an ALD.

Longevity of an ALD has been estimated by TVA and TWPC laboratory and field investigations into the dissolution rate of limestone. These studies include small-scale pilot studies and evaluations of existing ALDs. The TWPC suspended preweighed nylon bags of limestone in an access well within the ALD at the Prototype 2 site in Scott County, TN (Turner and McCoy 1990). After 6 months, the bags were retrieved and weighed. Average loss of the test limestone rocks was 0.24 g/kg/day. Average expected longevity of the ALD would be 23 years, at which time total dissolution will have occurred (Brodie et al 1991).

Table 9

Final Costs of TVA's IMP4 ALD

Design & Engineering	\$3500
Labor & Supervision	3000
Equipment (dozer, hydraulic excavator, loader)	8000
Limestone (400T @ \$7/T)	2800
Plastic	600
Geofabric	800
Seed, Mulch, Fertilizer	<u>250</u>
 Total	 \$18,950

A study by TVA at its Kingston Fossil Plant in Roane County, Tennessee suggested that increased flow rates increased the rate of limestone dissolution and the amount of alkalinity in the effluent. Flow rates varied from 0 to 3.12×10^{-3} L/min/kg limestone. Anoxic seep water was routed through 4 55-gallon polypropylene barrels filled with high-CaCO₃ (about 92%), 3/4 in. crushed limestone. The anoxic seep water entered the bottom of the barrels and flowed upward, exiting at the top of the barrels; thus ensuring anoxic conditions prevailed in the limestone when seep flow was present. Flows were monitored and adjusted weekly, and effluent was sampled on a weekly basis. Table 10 shows the results of this sampling effort. Barrel 3 was removed from the experiment after 56 days because of difficulties in maintaining flow to it from the seep. A larger diameter inlet line was installed on barrel at the same time.

The limestone dissolution rate for the first 56 days of the study, as calculated from weight lost by an approximately 20 kg sample in a PVC mesh bag in Barrel 3, was 4.19 g/kg or 0.0748 g/kg/day; implying a longevity of 37 years for a scaled-up equivalent system. Preliminary estimates for dissolution rates for barrels 1, 2, and 4 for the entire 90-day period range between 2.96 and 5.80 g/kg, implying daily rates of between 0.0329 and 0.0644 g/kg/day. At these daily rates, an ALD experiencing similar loading conditions would be expected to last between 42 and 83 years.

Another method of estimating longevity of an ALD is to compare pre-ALD and post-ALD data in the ALD effluent. For example, Ca⁺² at TVA's Impoundment 4 seep increased from an initial 14 mg/L to about 56 mg/l after ALD construction. Thus, the ALD is adding 41 mg/l of Ca⁺², which corresponds to

Table 10

Preliminary Results TVA Limestone Dissolution Study
at Kingston Fossil Plant

	Inflow	Bbl 1	Bbl 2	Bbl 3	Bbl 4
pH	5.9	6.8	6.5	6.5	6.3
Alkalinity (mg/l)	62.8	143.1	97.0	94.5	95.3
Acidity (mg/l)	20.2	-111.0	-68.2	-64.5	-49.7
Total Al (mg/l)	0.04	0.02	0.00	0.01	0.01
Total Fe (mg/l)	60.8	28.0	43.7	55.5	52.7
Total Mn (mg/l)	2.1	1.4	1.6	1.5	1.7
Total Cu (mg/l)	0.00	0.00	0.00	0.01	0.01
Total Ca (mg/l)	108.3	170.0	127.8	143.8	131.0
Total Mg (mg/l)	23.2	24.7	23.6	23.9	22.6
Total Zn (mg/l)	0.01	0.06	0.02	0.03	0.01
SO ₄ (mg/l)	413.3	403.3	427.8	368.5	381.8
Average Flow (l/min/kg)		2.0E-04	4.9E-04	8.6E-04	2.7E-03
Duration of Test (days)		90	90	56	90
Limestone Dissolved (g/kg)		5.3	3.0	4.2	5.8
Dissolution Rate(g/kg/day)		0.06	0.03	0.07	0.06
Expected Longevity (years)		46	83	37	42

105 mg/l CaCO₃. Average flow is 15 gpm and the ALD contains about 400 tons of 92% CaCO₃ limestone with 2% MgCO₃. Assuming that all the calcium carbonate is consumed and that the 2% Mg can bind with available Ca⁺² rendering it nonreactive (i.e., only 90% of the tonnage should be assumed available for bicarbonate production), the lifespan of the ALD can be calculated at 102 years.

It is noted that longevity of an ALD will be affected by changes in the hydraulic characteristics of the ALD. Limestone dissolution and sediment clogging over several decades will reduce the hydraulic conductivity of an ALD and potentially result in hydraulic failure in the form of surface boils or blowouts.

Overdesign of an ALD on the order of 2 to 3 times is the best insurance for a long-term, stable system. Thus, the effective lifespan of the above-referenced Impoundment 4 ALD is more likely about 50 years.

Conclusions

Passive anoxic limestone drains have been installed at sites in Tennessee and Alabama to increase buffering capacity and alkalinity in acid mine drainage and prevent pH decreases in effluent due to Fe hydrolysis. Results to date are optimistic and in most instances alkalinity has been significantly increased to levels sufficient to buffer the flow from pH decreases. Long-term results must be evaluated to establish the

stability of an ALD over the duration of acid drainage at a particular site.

Design, utility, and longevity of an ALD require further examination to develop guidelines and expectations. Particularly, stone gradation and composition, dissolution rates, reaction mechanisms and products, and the nature of ALD-enhanced Mn removal need to be researched. However, the relative simplicity and low cost of the ALD and preliminary studies indicating lifespans of several decades suggest that the system may be an effective means of renovating inefficient, existing aerobic wetlands and an important component of staged wetlands system designs, providing the capability of treating poorer quality waters with less area. Further evaluation is needed to identify the benefits of using an anoxic drain as a staged component of a sulfate reducing wetlands system.

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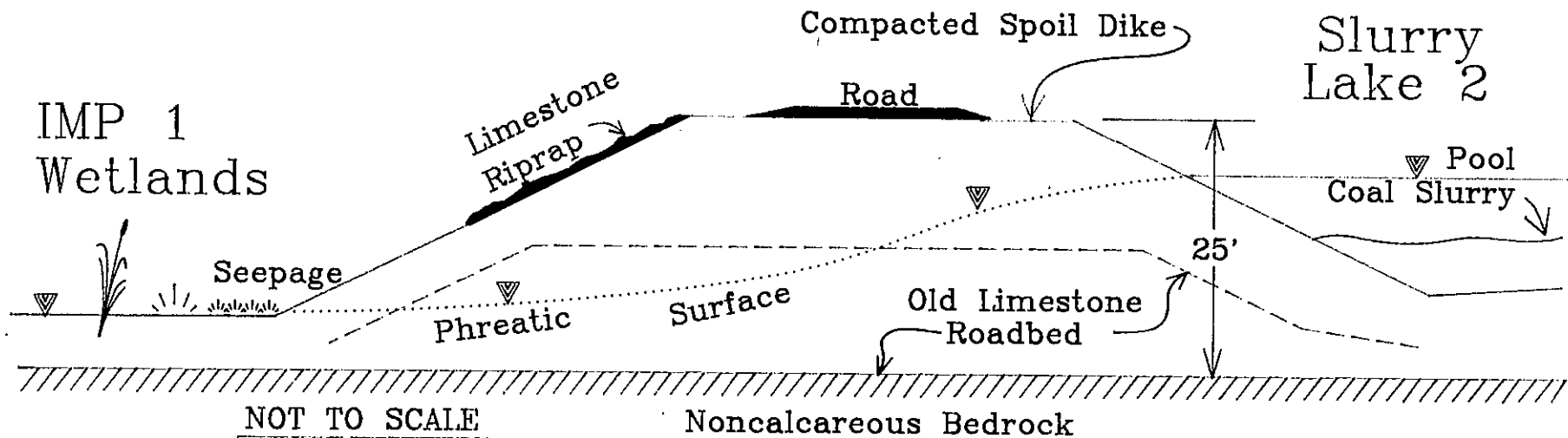


Figure 1. Schematic of Roadbed Originated Anoxic Drain at TVA's Fabius IMP1 Constructed Wetlands

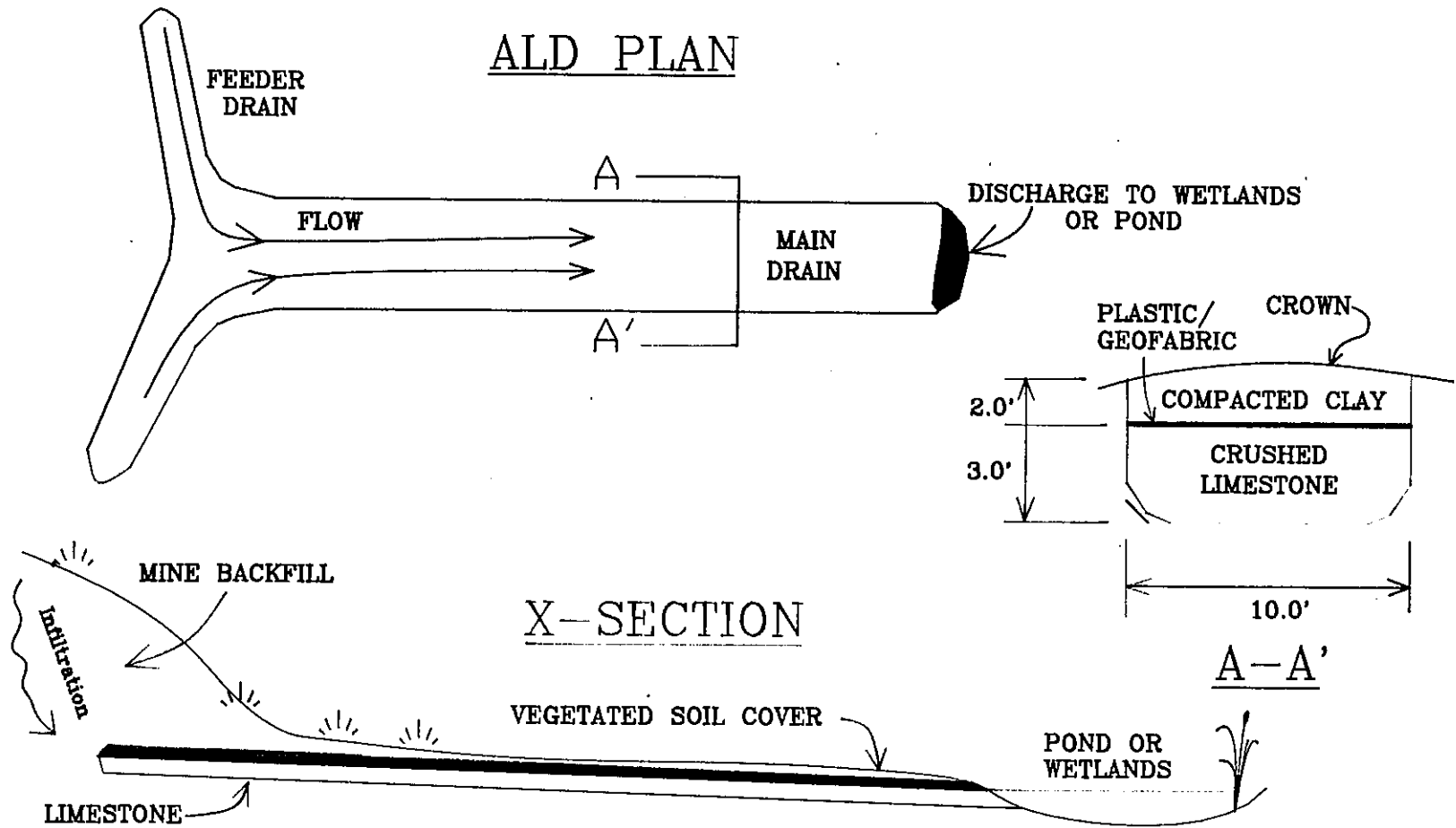


Figure 2. Generalized Schematic of Anoxic Limestone Drain

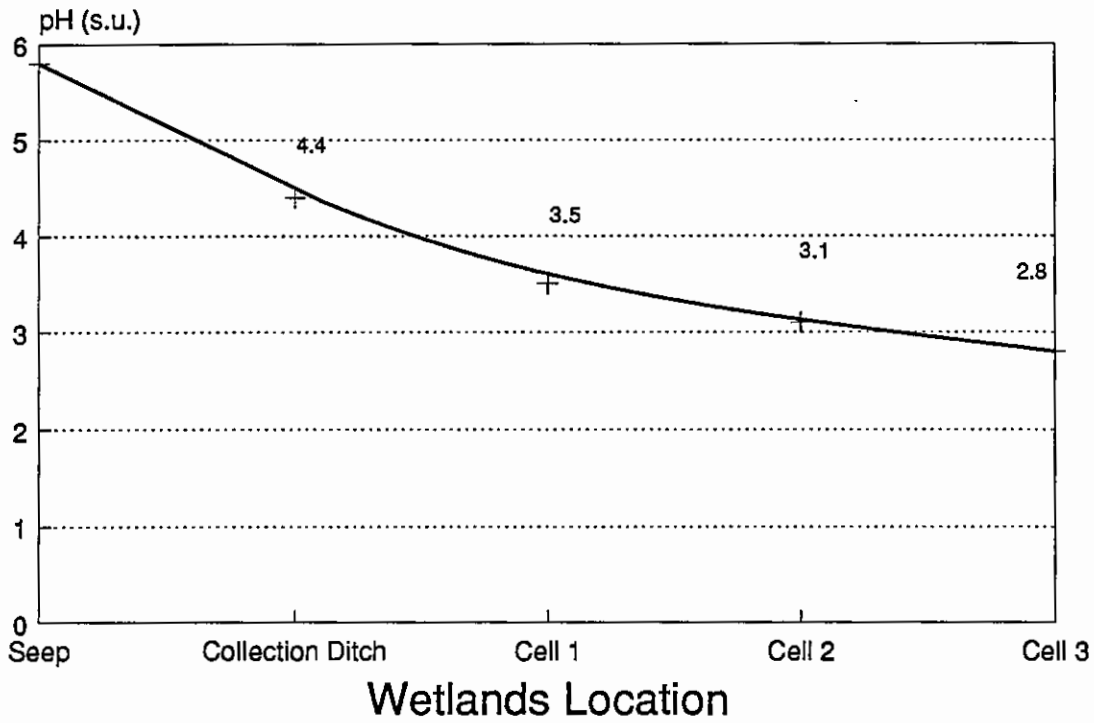


Table 1. pH Decrease at TVA Kingston Constructed Wetlands

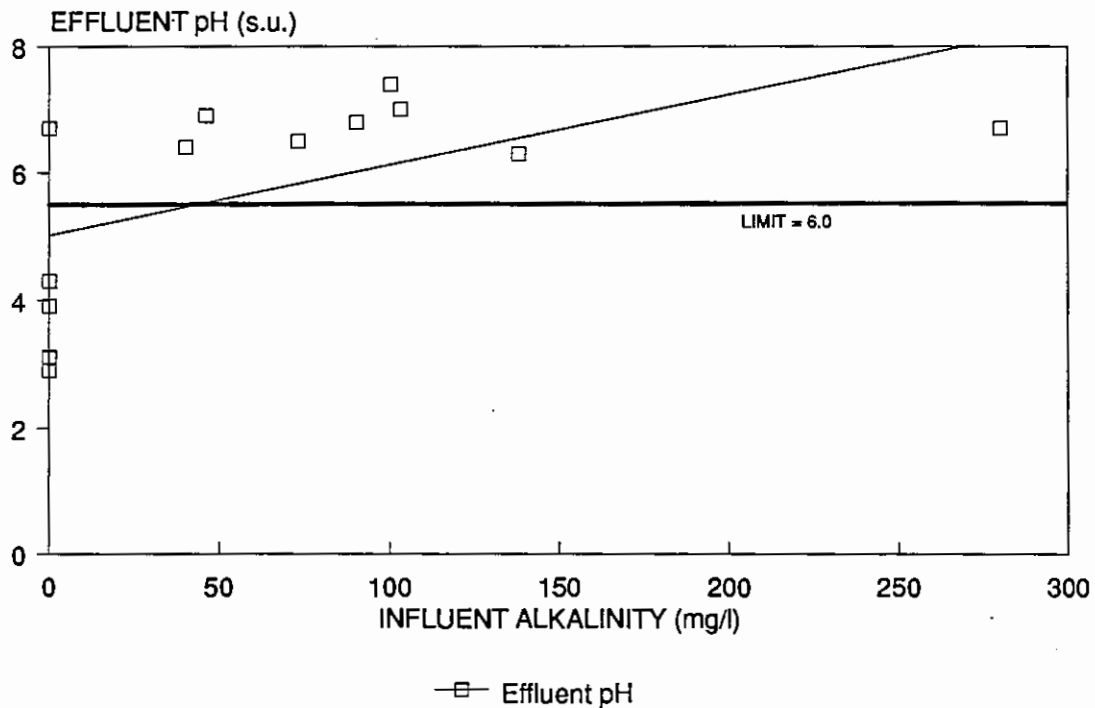


Table 2. Effluent pH vs. Influent Alkalinity (TVA Wetlands)

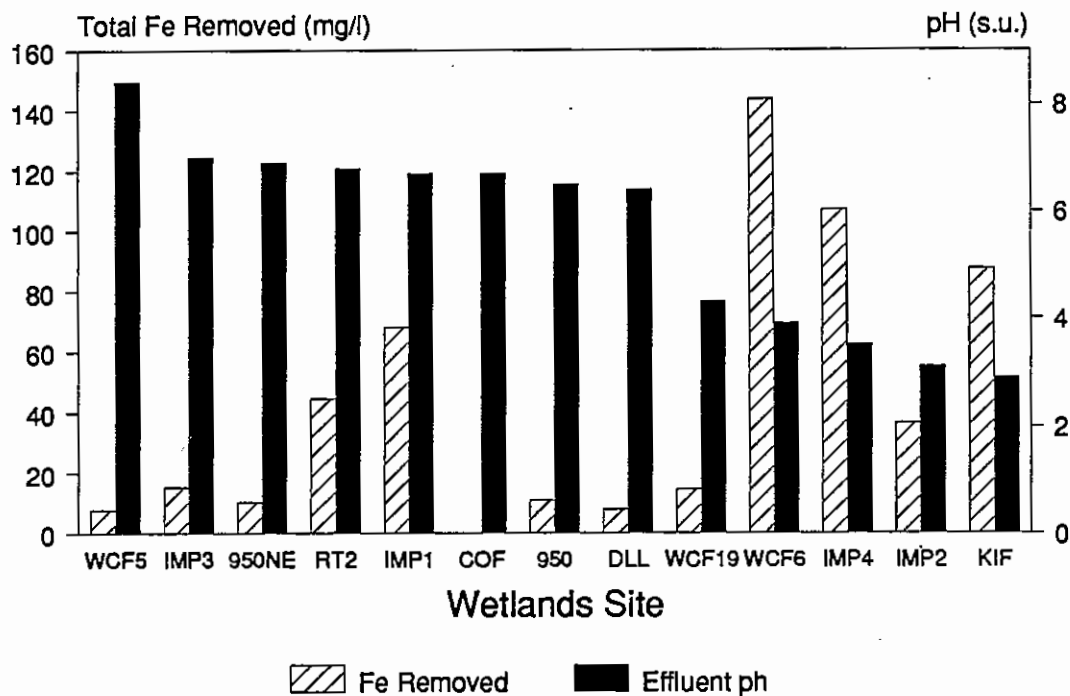


Table 3. Fe Removed vs. Effluent pH (TVA Wetlands)

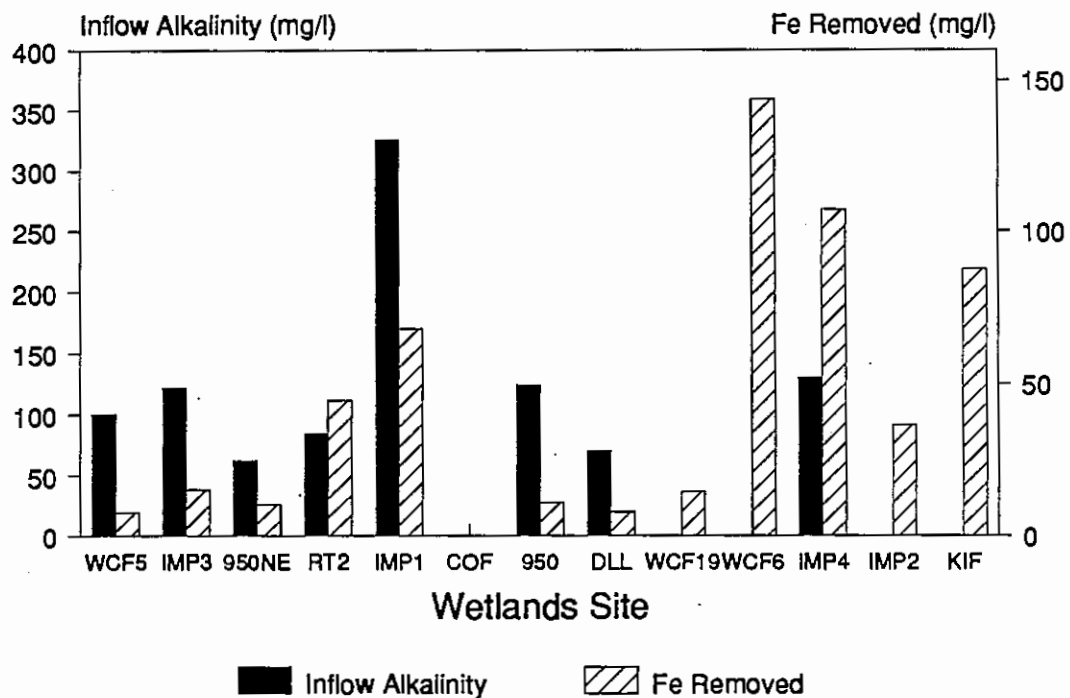


Table 4. Inflow Alkalinity vs. Fe Removed (TVA Wetlands)

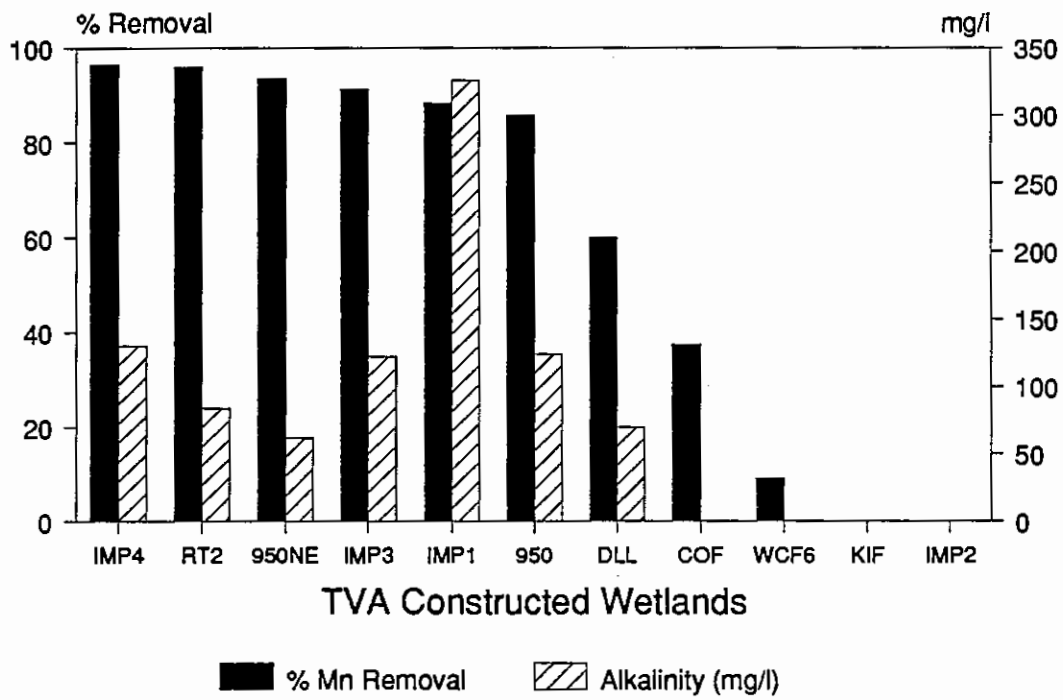


Table 5. Mn Removal Related to Alkalinity

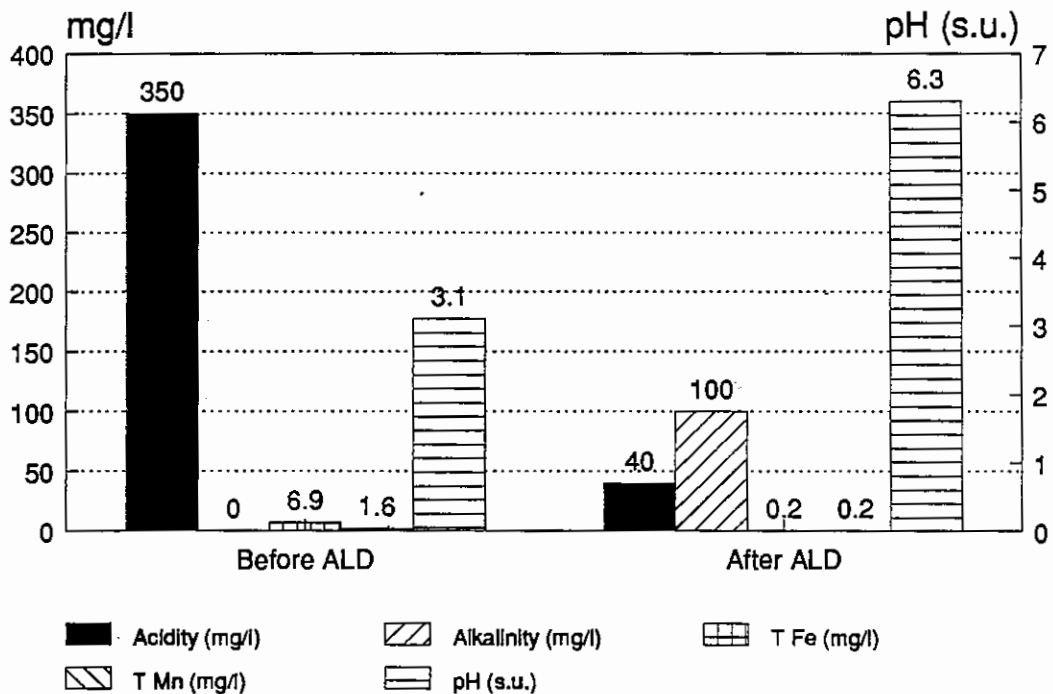


Table 6. IMP4 Wetlands Effluent Data Before and After ALD

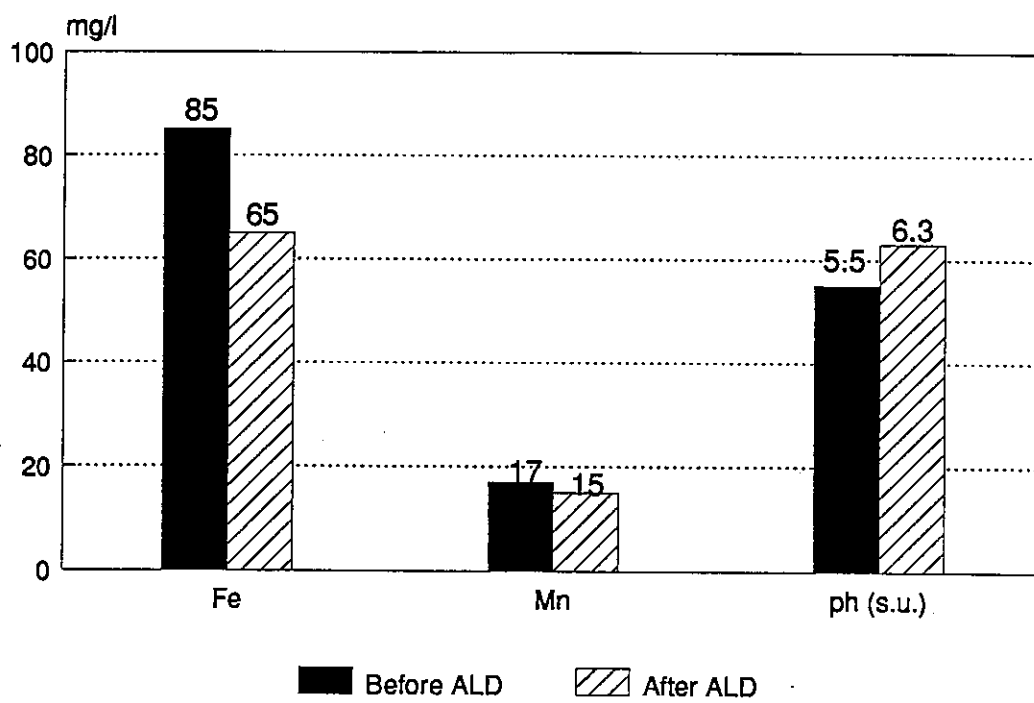


Table 7. IMP4 Seep Data Before and After ALD