SAND-SIZED LIMESTONE TREATMENT OF STREAMS IMPACTED BY ACID MINE DRAINAGE

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Abstract. Direct single point, instream applications of limestone sand were made in two streams severely acidified by acid mine drainage. After high flows distributed the sand downstream, dissolution of the limestone significantly reduced the acid loads in both streams. Data from the treated stream sections and laboratory experiments showed that the efficiency of treatment was more dependent on limestone particle size than $CaCO_3$ content. Results from the stream treated with a narrow range of limestone sand-sized particles showed a high utilization efficiency with nearly complete dissolution of the limestone. The data indicated that monthly addition of limestone sand may be sufficient for complete treatment of the streams studied. An analysis of such a treatment scheme indicated it to be highly cost efficient when compared to other active and passive treatment systems.

Keywords: acid mine drainage, limestone, neutralization, treatment

1. Introduction

Studies on the use of limestone to neutralize streams acidified by acid mine drainage (AMD) or acid deposition (AD) (Clayton *et al.*, 1998; Zurbuch *et al.*, 1997) have been encouraged by its availability and relatively low cost. Research into the neutralization of AMD has generally concluded that limestone aggregate is of limited use because of low utilization efficiencies ($<$50%), and the tendency of larger size aggregates to become non-reactive due to coatings of iron and aluminum oxyhydroxide precipitates that form during the reaction process (Lovell, 1973; U.S. Environmental Protection Agency, 1983). Recent AMD research has centered on the use of limestone in passive systems such as anoxic limestone drains (Hedin *et al.*, 1994) and open limestone channels (Ziemkiewicz *et al.*, 1996). The results from these studies have not only been mixed as to their immediate and long term success, but their application appears to be limited to small flows with low or moderate acid loads and where suitable land area is available for their construction.

Researchers treating streams acidified by AD have largely relied upon mechanical systems that dispense finely ground limestone powders or limestone slurries (Olem, 1991). The direct instream application of limestone aggregate to AD-acidified streams has been reported on by a number of workers (Arnold *et al.*,

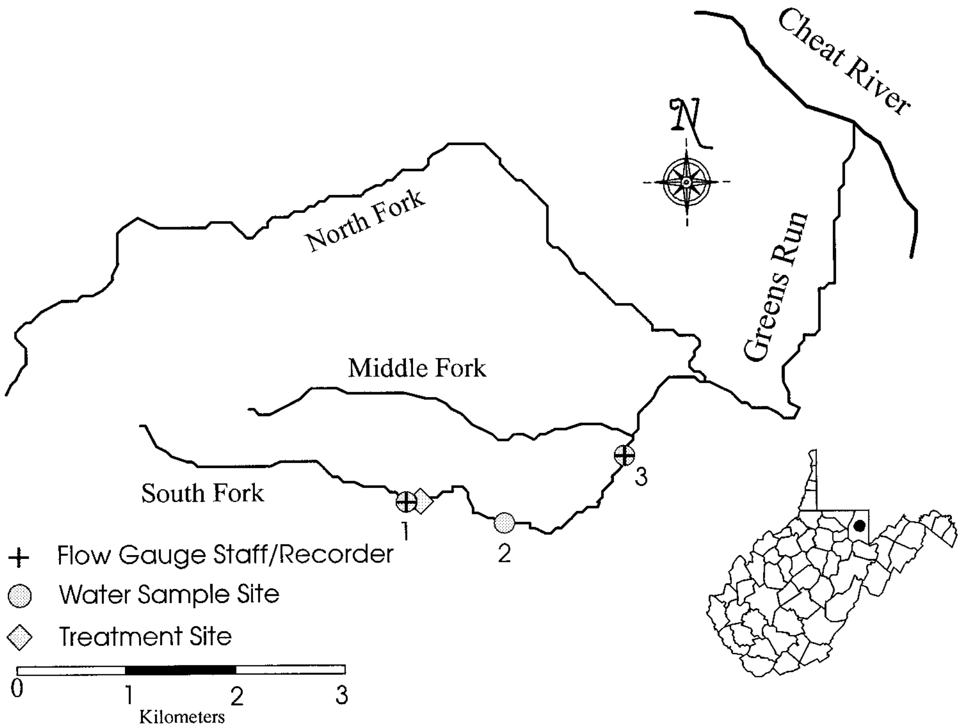

Figure 1. Map of Greens Run showing the South Fork station locations.

1988; Baalsrud, 1985; Gunn and Keller, 1984). The conclusion drawn from most of these studies was that limestone does not provide prolonged neutralization due to the particle surfaces becoming coated and non-reactive by the same precipitates occurring in AMD situations (Porcella *et al.*, 1990). Recently, however, research has shown that the limestone sand introduced into AD-impacted streams remained reactive and effectively treated the acidic water and restored fish populations (Ivahnenko *et al.*, 1988; Downey *et al.*, 1994; Clayton *et al.*, 1998).

A major West Virginia river, acidified by both AMD and AD, was biologically restored largely by treating its tributaries with instream applications of limestone sand (Zurbuch *et al.*, 1997). The apparent effectiveness of the limestone sand in those tributaries containing AMD led to the study presented herein. The objectives of the study were: (1) to verify the effectiveness of sand-sized limestone aggregate to treat streams heavily impacted by AMD; (2) to optimize treatment effectiveness; and (3) to determine the neutralization efficiency of limestone sands with different $CaCO_3$ contents.

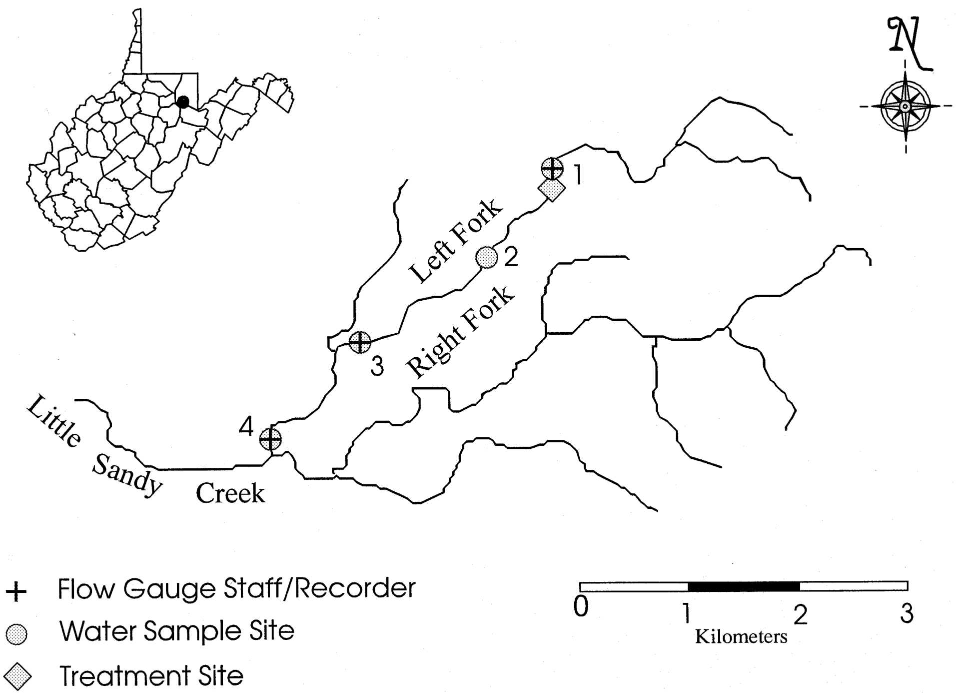

Figure 2. Map of Little Sandy Creek showing the Left Fork station locations.

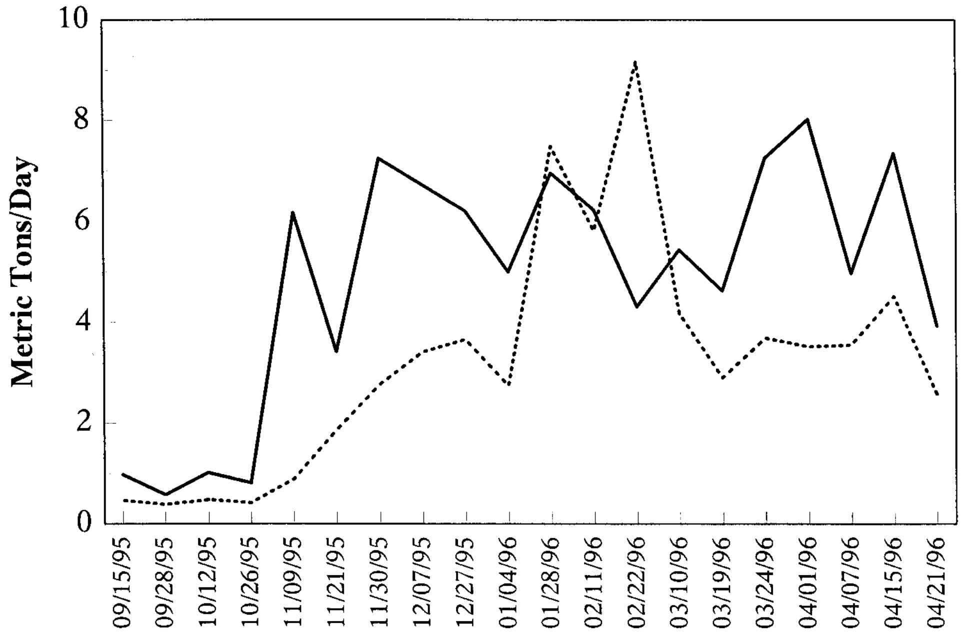

Figure 3. Pre treatment acid loads of South Fork Greens Run (- - -) and Left Fork of Little Sandy Creek (—).

2. Study Streams

Two AMD-impacted streams were selected for the study in Preston County, WV: the South Fork of Greens Run near Kingwood, and the Left Fork of Little Sandy Creek near Fellowsville. In both cases, samples for water chemistry and flow measurements could be obtained above and below treatment with little or no additional inputs from tributaries. The limestone treatment sites and water sampling stations are shown for the South Fork of Greens Run in Figure 1 and for the Left Fork of Little Sandy Creek in Figure 2.

The headwaters of Greens Run is underlain by the Pennsylvania age Upper Freeport coal bed and associated rocks. Extensive deep and surface coal mining have taken place within the South Fork's drainage basin with a number of acid seeps entering the stream above the limestone treatment site. Pre treatment water sampling showed the South Fork's acid load to range from <1 to 9.2 metric tons per day, averaging 3.2 metric tons per day (Figure 3).

The headwaters of the Left Fork of Little Sandy Creek are also underlain by the Pennsylvanian age Upper Freeport coal bed. The entire stream is affected by AMD, most of which enters the stream above the limestone treatment site. Pre treatment acid loads ranged from <1 to 8.0 metric tons per day and averaged 4.8 metric tons per day (Figure 3).

3. Methods

3.1. Treatment

The limestone was added to the two streams by dump truck at a single point. The streams' flow then distributed the particles downstream where they mixed with the natural streambed sediments. Previous AD research (Downey *et al.*, 1994; Zurbuch *et al.*, 1996; Clayton *et al.*, 1998) showed that particles >70 μm in size remained, for the most part, within the streambed for a few hundred meters below the treatment site where they slowly dissolved in the acid flow. Thus in this study it was believed that the added limestone either dissolved or remained in the sediments within the treated stream section.

Two grades of limestone were used in the study streams. The South Fork of Greens Run was treated with a high grade ($>$97% $CaCO_3$) sand-sized Ordovician Black River limestone aggregate from the Germany Valley quarry, Pendleton County, WV. The Left Fork of Little Sandy Creek was treated with a lower grade (83% $CaCO_3$) Greenbrier limestone aggregate from the Greer quarry, Monongalia County, WV.

The application rate for both streams was based on approximately twice the projected highest monthly acid load of 182 metric tons. This was determined from both pre treatment and historical data available for both streams. The South Fork

TABLE I

Metric tons of limestone and its $CaCO_3$ content used to treat the South Fork of Greens Run and the Left Fork of Little Sandy Creek and application dates. Limestone used to treat the South Fork of Greens Run was 97% $CaCO_3$. That used to treat the Left Fork of Little Sandy was 83% $CaCO_3$ for the first treatment and 69% the second treatment

South Fork Greens Run			Left Fork Little Sandy Creek		
Date	Tons		Date	Tons	
	Limestone	$CaCO_3$		Limestone	$CaCO_3$
04/24/96	35.6	34.5	04/25/96	53.1	44.1
04/25/96	16.9	16.4	04/26/96	61.9	51.4
04/26/96	51.9	50.3	04/29/96	56.9	47.2
04/30/96	17.2	16.7	04/30/96	82.9	68.8
05/01/96	132.3	128.4	05/01/99	83.2	69.1
05/02/96	36.2	35.1	05/02/96	27.1	22.5
05/07/96	75.5	73.2			
Sub total	**365.6**	**354.6**	**Sub total**	**365.1**	**303.1**
05/21/96	17.1	16.8	05/22/96	81.0	55.9
05/22/96	17.4	16.9	05/23/96	57.9	39.9
05/29/96	57.2	55.5	05/24/96	58.2	40.2
05/30/96	57.5	55.8	05/28/96	26.7	18.4
05/31/96	60.4	58.6	05/29/96	84.3	58.2
06/03/96	100.1	97.1	05/30/96	27.1	18.7
06/04/96	55.3	53.6	06/03/96	29.9	20.6
Sub total	**365.2**	**354.3**	**Sub total**	**365.1**	**251.9**
Total	**730.8**	**708.9**	**Total**	**730.2**	**555.00**

was initially treated with 366 metric tons of limestone (355 metric tons $CaCO_3$) from April 24 to May 7, 1996 and the Left Fork was treated with 365 metric tons (303 metric tons $CaCO_3$) from April 25, to May 2, 1996 (Table I). During the first month following treatment, the acid loads were higher than expected depleting significant quantities of the first treatment, and a second application was made to both streams. The South Fork received an additional 365 metric tons from May 21 to June 4, 1996 and the Left Fork of Little Sandy received an additional 365 metric tons from May 22 to June 3, 1996 (Table I). After the second treatment was made on the Left Fork, it was discovered that the limestone from the Greer quarry

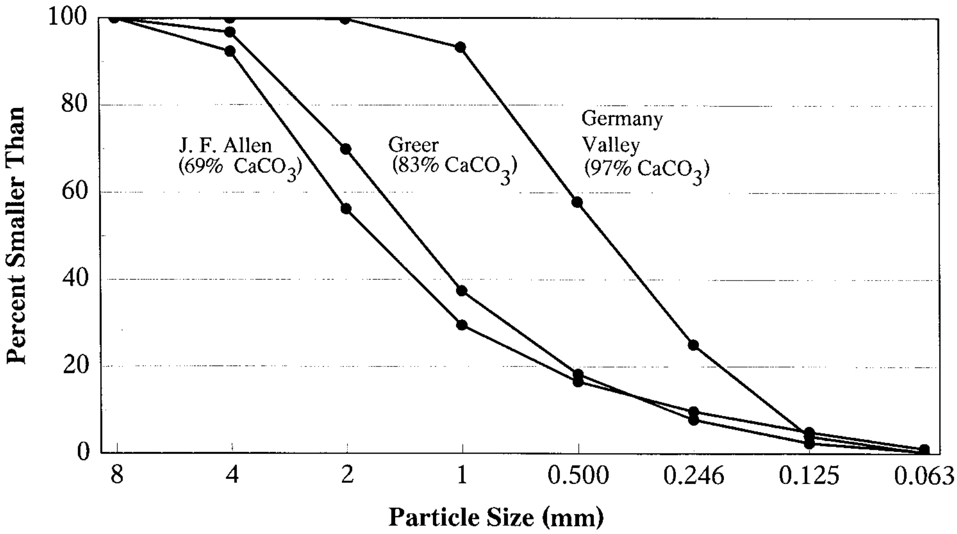

Figure 4. Particle size distribution of three limestone grades tested.

varied greatly in quality and that used for the second application contained only 69% $CaCO_3$ or 252 metric tons of $CaCO_3$.

Laboratory tests were conducted on the two grades (97 and 83% $CaCO_3$) of limestone used in the study plus an additional Greenbrier limestone (69% $CaCO_3$) obtained from the J.F. Allen quarry near Elkins, Randolph County, WV. Particle size distributions of the three grades of limestone are shown in Figure 4. Dissolution experiments consisted of subjecting a 10 g sample of each aggregate to 500 mL of water acidified to pH 2 with HCl while stirring at 20 °C for 10 min. The pH of the solution was monitored at 15 sec intervals for the first minute, 30 sec intervals for the next five minutes and at one minute intervals for the remainder of the experiment. The two lower grades of limestone were then size adjusted by sieving to eliminate the larger fraction ($>$1 mm), the experiments were repeated and the results compared to that from the high grade limestone. Dissolution rates were determined from the slope (pH change per minute) of the dissolution reaction curve for pH plotted against time (Sherlock, 1997).

3.2. CHEMISTRY

Pre treatment water quality data were collected from September 15, 1995 to April 21, 1996 and treatment data were collected from April 29, 1996 to February 19, 1997. During the pre treatment period, samples were collected on a bi-weekly basis with weekly samples being taken during the treatment period. Field measurements were made for stream flow, pH, temperature and specific conductivity. Laboratory analyses were made for pH, specific conductivity, acidity, alkalinity, aluminum, calcium, iron, magnesium, manganese, potassium, sodium and sulfate.

Each sampling included an unfiltered sample for determining pH; a filtered sample for specific conductivity, acidity, alkalinity and sulfates; an unfiltered acidified sample for total calcium; and a filtered acidified sample for dissolved metal analyses. Determination of field pH was made with an Orion Model 250A meter with a 91–57 triode electrode. Conductivity was obtained with a Labcraft Model 264–774 meter. Laboratory measurements for pH and conductivity were done with a Fisher Model 910 and Model VWR 2052 meter, respectively. Cations were analyzed by graphite furnace atomic absorption spectrometry and sulfates by turbidimetric analysis. All analyses were done in accordance with Standard Methods (American Public Health Association, 1989).

3.3. Flow

Continuous recording gauges (Stevens Model 71 type A) were operated on South Fork of Greens Run (Station 3) and the Left Fork of Little Sandy Creek (Station 4). Staff gauges were installed on the South Fork at Stations 1 and 3 and on the Left Fork at Stations 1, 3 and 4. Instantaneous measurements of stream flow were obtained by means of a Marsh-McBirney flow meter (Model 201). The mid-section method of the U.S. Geological Survey was used for discharge measurements (Buchanan and Somers, 1969).

3.4. Data analysis

Limestone depletion was calculated based on the percentage of $CaCO_3$ content as described by Clifford and Snavely (1954). For the South Fork of Greens Run, 1.0 metric ton of the 97% $CaCO_3$ limestone was required to neutralize 1.0 metric ton of acid. Neutralization in the Left Fork of Little Sandy Creek required 1.2 metric tons of the 83% $CaCO_3$ limestone per metric ton of acid and 1.4 metric tons of the 69% $CaCO_3$ limestone per metric ton of acid. The amount of limestone required in the Left Fork during the second treatment period was 1.4 metric tons per metric ton of acid and was based on the proportions of the 83% $CaCO_3$ limestone remaining after the first treatment and the amount of the 69% $CaCO_3$ limestone added during the second treatment.

Data were analyzed to determine neutralization efficiency (based on the percent reduction in acid load) of the two grades of limestone used and the depletion rates of the limestone. On both study streams, pre treatment data showed a natural reduction in acidity levels from Station 1 (upstream control) downstream to the lower most sampling stations. To more accurately estimate the reductions in acid loads due to treatment, the untreated acidity levels at Station 3 of the South Fork and Stations 3 and 4 of the Left Fork were calculated using regression analyses of the pre treatment acidity data at Station 1 versus the pre treatment acidity data at Station 3 of the South Fork and Stations 3 and 4 of the Left Fork. The regression equations used to calculate the untreated acidity are as follows:

South Fork of Greens Run:
Untreated Acidity Station 3 = 0.72 × Acidity Station 1 - 96.41 (r^2 = 0.84)
Left Fork of Little Sandy Creek:
Untreated Acidity Station 3 = 0.50 × Acidity Station 1 + 37.28 (r^2 = 0.95)
Untreated Acidity Station 4 = 0.42 × Acidity Station 1 + 16.48 (r^2 = 0.97)

Similar analyses were used to calculate daily flows; untreated and treated acid loads on a daily basis, and changes in metal loading. Statistical differences in metal, sulfate and acid loadings were determined using paired *t* tests (alpha $\leq$0.05). All statistical analyses were done with Statistical Analysis Systems Institute's JMP software version 3.1.5 (SAS Institute, 1995).

4. Results

4.1. Limestone

Laboratory tests of limestone dissolution showed that the Greer quarry limestone (with 83% $CaCO_3$) initially used in the Left Fork of Little Sandy Creek dissolved at 44% of the dissolution rate of the Germany Valley limestone (with 97% $CaCO_3$) used in the South Fork of Greens Run. The J.F. Allen quarry limestone (with 69% $CaCO_3$) dissolved at only 15% of the dissolution rate of the Germany Valley limestone; the J.F. Allen limestone is similar to the Greer quarry limestone used for the second treatment of the Left Fork. These differences in dissolution proved to be mostly a function of limestone particle size, with the coarser (sand to gravel) Greer quarry and J.F. Allen quarry limestones dissolving slower than the sand-sized Germany Valley limestone. Further laboratory tests with screened fine-sand-sized ($>$1 mm) limestone revealed that the Greer quarry and J.F. Allen quarry limestones dissolved at 82 and 87%, respectively, of the rate (pH change per minute) of the Germany Valley limestone. For a more detailed discussion, see Sherlock (1997).

4.2. Chemistry

4.2.1. *South Fork of Greens Run*

Pre treatment water quality data collected on the South Fork of Greens Run at Station 3 (Table II) showed a mean acidity of 380 mg L^{-1} with a range of 178 to 676 mg L^{-1}. Following treatment, the acid loads were reduced significantly. The treated acid loads compared to the estimated untreated acid loads summarized by week are shown in Figure 5, while the percent of the acid load neutralized is shown in Figure 6.

During the first treatment period (April 24 to May 20, 1996), the acid load at Station 3 was estimated at 256 metric tons, and the amount of acid neutralized was estimated at 194 metric tons or 76%. During the second treatment period,

TABLE II

Means and (ranges) of various chemical parameters from South Fork Greens Run stations. Pre treatment data from September 15, 1995 to April 21, 1996. Treatment data from April 29 1996 to February 19 1997. Values in mg L^{-1} except pH and conductivity. Acid loads in metric tons day^{-1}

Station	pH	Acidity	Conductivity	SO_4	Al	Fe	Mn	Mg	Ca	Acid load
1 – Pre	2.85	679	1893	813	40.9	126.9	5.1	28.4	77.4	4.82
	(2.69–3.01)	(488–938)	(277–2400)	(604–1128)	(24.5–60.8)	(82.0–192.7)	(3.3–8.1)	(16.6–57.8)	(44.7–130.3)	(0.42–16.8)
1 – Treat	2.88	590	1577	725	28.5	86.7	3.9	23.7	66.1	5.02
	(2.61–3.67)	(221–879)	(951–2072)	(333–995)	(13.1–41.2)	(32.7–127.2)	(2.2–5.8)	(12.1–36.1)	(39.6–95.9)	(0.31–23.5)
2 – Pre	2.99	555	1666	636	33.5	98.4	4.1	22.7	70.5	
	(2.65–3.18)	(2.85–817)	(1211–2170)	(467–853)	(17.1–51.2)	(32.5–153.3)	(2.3–6.5)	(12.3–34.0)	(36.5–127.3)	
2 – Treat	3.75	286	1258	599	20.1	37.5	3.2	20.9	116.2	
	(2.85–7.39)	(2.0–665)	(517–1909)	(192–995)	(1.7–31.3)	(1.7–83.8)	(1.5–4.5)	(8.7–31.7)	(41.9–215.8)	
3 – Pre	3.13	380	1323	457	24.3	65.4	3.2	17.9	63.5	3.23
	(2.70–3.44)	(178–676)	(925–1970)	(313–705)	(11.8–45.2)	(17.0–111.5)	(1.7–5.7)	(9.9–31.8)	(33.1–119.7)	(0.40–9.15)
3 – Treat	4.03	161	1016	473	13.2	15.9	2.2	17.1	112.3	1.60
	(3.09–7.20)	(12–352)	(339–1794)	(125–791)	(1.5–23.4)	(0.4–3.5)	(0.4–3.5)	(6.7–26.2)	(45.2–201.4)	(0.0–6.8)

Station 1 was not treated.

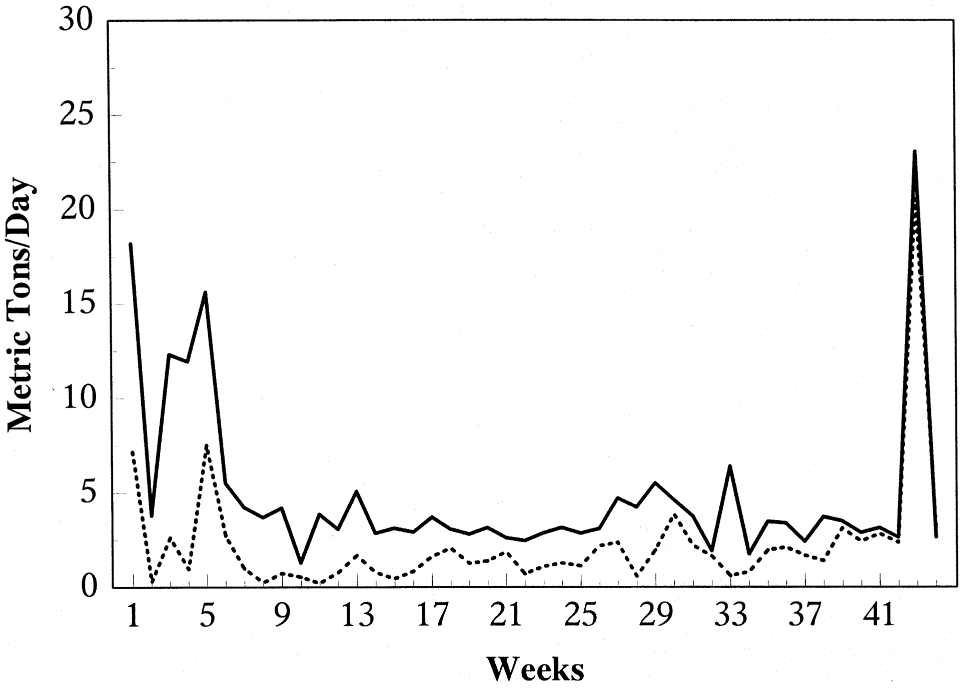

Figure 5. Regression estimated untreated acid load (—) compared to treated acid load (- - -) at Station 3 on South Fork of Greens Run.

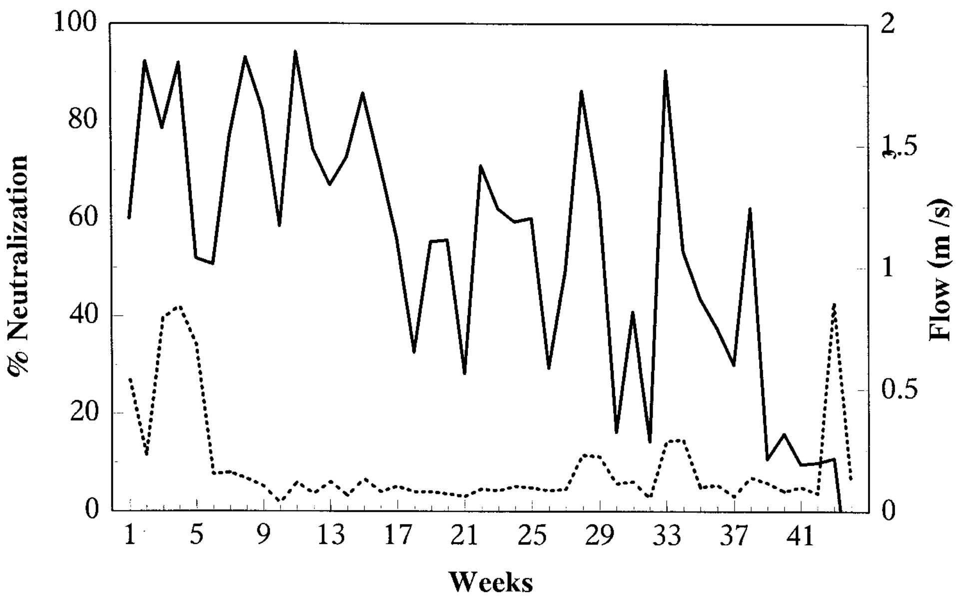

Figure 6. Percent acid load neutralized (—) compared to stream flow (- - -) at Station 3 on South Fork of Greens Run.

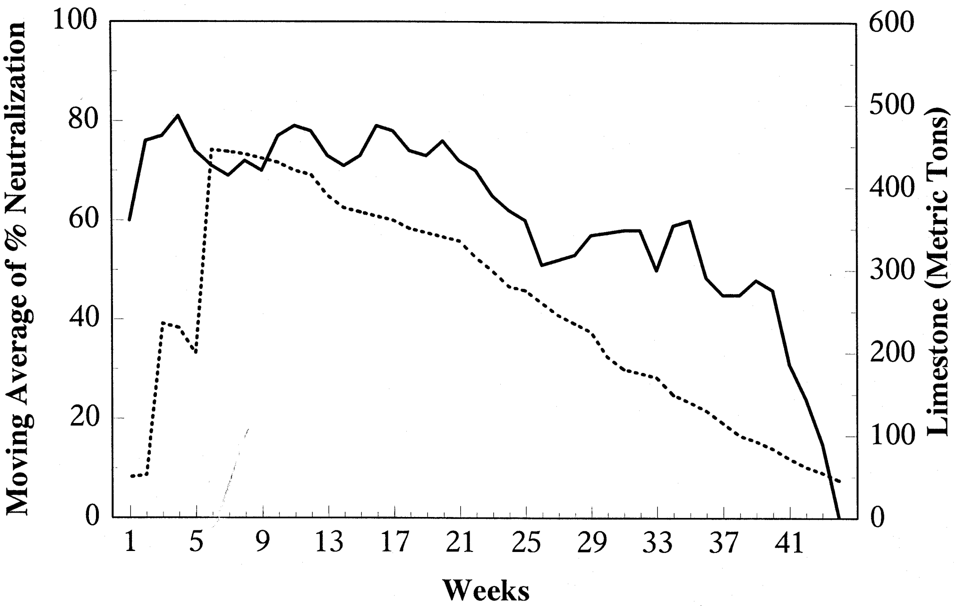

Figure 7. Moving average of percent acid load neutralized (—) at Station 3 on South Fork of Greens Run compared to the limestone remaining in the stream (- - -).

the acid load was estimated at 839 metric tons of which 470 metric tons (56%) were neutralized. For the entire treatment period, the acid load was estimated at 1095 metric tons of which 664 metric tons (61%) were neutralized. Figure 7 shows the weighted moving average of percent neutralization and limestone depletion. Generally, neutralization was highest during the early part of the study when flows were highest. Efficiency decreased during low summer flows but tended to increase whenever flows increased. When data collection ended, an estimated 46 metric tons of limestone remained in the stream.

The mean values and ranges for various chemical and physical parameters are shown in Table II. Mean values at Station 1 were not different during the pre treatment and treatment periods when adjusted for flow differences. Analyses for metals and sulfate loads during the treatment period at Station 3 showed significant reductions in iron and aluminum, significant increases in calcium and no difference in sulfates.

4.2.2. *Left Fork of Little Sandy Creek*

Pre treatment water quality (Table III) collected on the Left Fork of Little Sandy Creek (Station 4) showed a mean acidity of 182 mg L^{-1} and ranged from 48 to 489 mg L^{-1}. Following treatment, the acid loads were reduced significantly. Plots of the treated acid loads compared to the estimated untreated acid loads are shown in Figure 8, while the percent of the acid load neutralized is shown in Figure 9.

TABLE III

Means and ranges of various chemical parameters from Left Fork of Little Sandy Creek. Pre treatment data from September 15, 1995 to April 21, 1996. Treatment data from April 29, 1996 to February 10, 1997. Values in mg L^{-1} except pH and conductivity. Acid loads in metric tons day^{-1}

Station	pH	Acidity	Conductivity	SO_4	Al	Fe	Mn	Mg	Ca	Acid load
1 – Pre	3.06	398	1229	367	29.8	52.9	1.1	15.8	41.1	6.21
	(2.44–3.30)	(104–1123)	(629–2490)	(180–1142)	(8.7–86.7)	(14.0–142.7)	(0.4–2.6)	(6.2–35.5)	(16.0–105.4)	(0.92–29.57)
1 – Treat	3.07	336	1142	482	22.7	45.4	1.0	15.6	37.6	4.84
	(2.75–3.54)	(65–820)	(287–2306)	(98–1156)	(4.5–51.1)	(7.2–98.7)	(0.3–1.7)	(4.1–30.8)	(10.5–67.3)	(1.38–19.29)
2 – Pre	3.13	371	1182	314	26.9	44.7	1.1	14.9	39.2	
	(2.50–3.55)	(91–1004)	(512–2450)	(115–1104)	(6.6–75.0)	(6.9–109.6)	(0.4–2.8)	(5.2–35.6)	(15.2–93.7)	
2 – Treat	3.41	246	925	389	19.3	30.7	0.9	14.8	54.1	
	(2.90–5.06)	(15–697)	(63–2045)	(63–979)	(2.8–43.4)	(1.1–75.1)	(0.2–1.6)	(3.6–30.0)	(20.8–92.4)	
3 – Pre	3.28	246	953	277	15.6	32.5	0.8	14.2	47.5	5.32
	(2.68–3.67)	(76–616)	(485–1740)	(110–929)	(5.4–35.0)	(6.1–76.9)	(0.3–1.8)	(5.4–32.8)	(17.9–118.5)	(0.69–9.03)
3 – Treat	3.53	176	807	364	13.4	23.9	0.8	15.7	60.5	3.36
	(3.03–4.94)	(15–422)	(44–1728)	(77–883)	(2.1–23.1)	(1.2–51.7)	(02–1.2)	(3.7–31.3)	(19.8–99.4)	(0.89–7.58)
4 – Pre	3.39	182	803	215	13.7	21.9	0.7	12.8	45.1	4.77
	(2.76–3.96)	(48–489)	(325–1830)	(95–685)	(4.1–36.8)	(3.2–55.2)	(0.2–1.9)	(4.5–32.2)	(16.7–121.8)	(0.58–8.03)
4 – Treat	3.73	130	756	298	9.8	14.0	0.6	12.3	50.6	2.98
	(3.1–5.66)	(8–332)	(17–1678)	(59–714)	(0.1–18.6)	(0.9–59.0)	(0.2–1.1)	(3.1–24.2)	(17.6–90.8)	(0.96–6.01)

Station 1 was not treated.

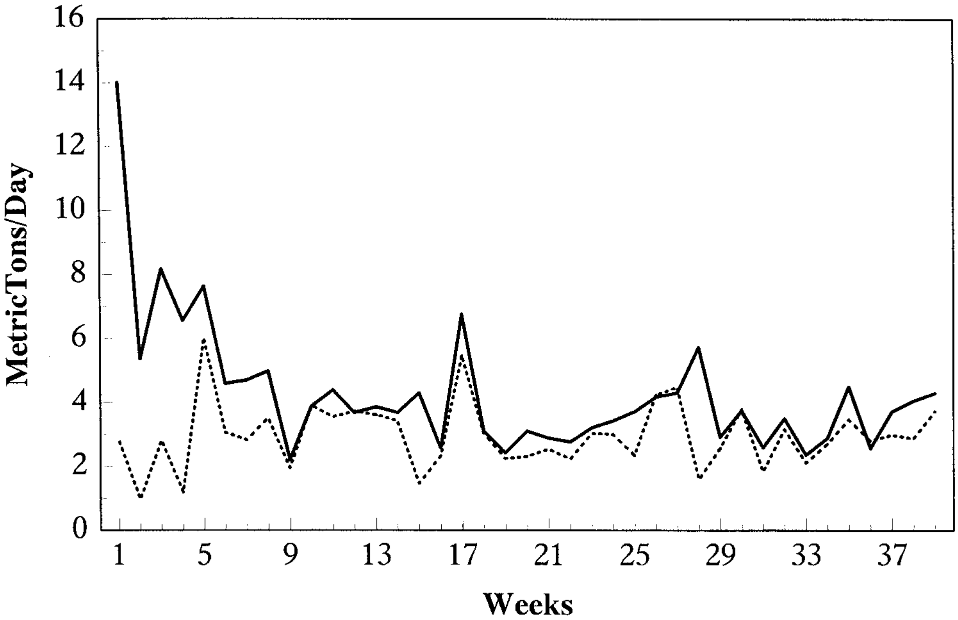

Figure 8. Regression estimated untreated acid load (—) compared to treated acid load (- - -) at Station 4 on Left Fork of Little Sandy Creek.

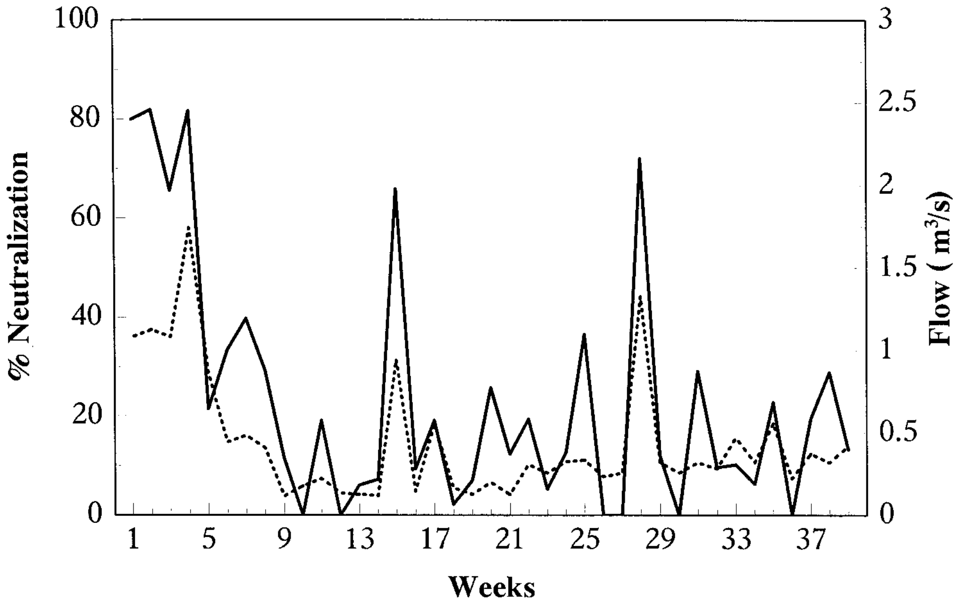

Figure 9. Percent acid load neutralized (—) compared to stream flow (- - -) at Station 4 on Left Fork of Little Sandy Creek.

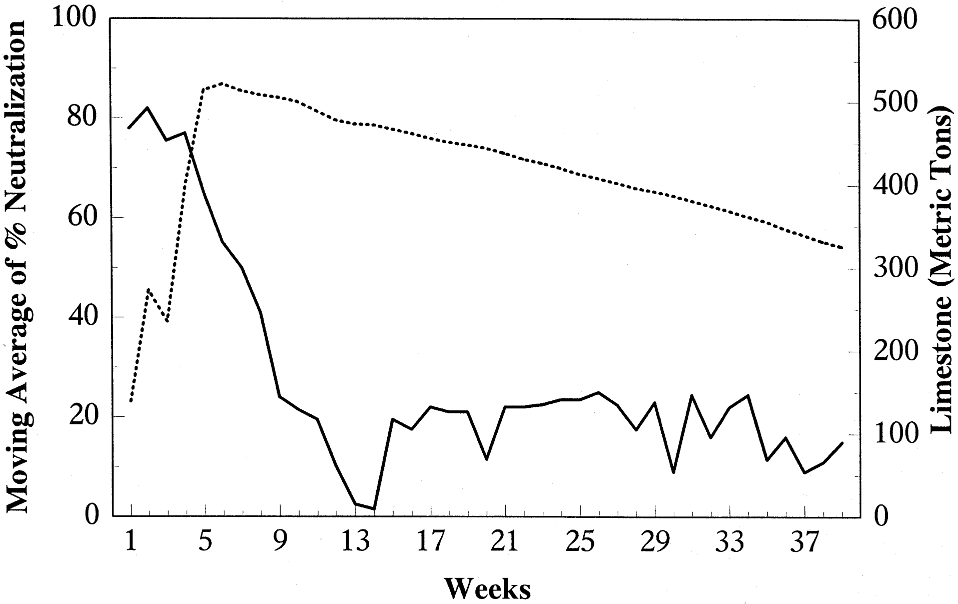

Figure 10. Moving average of percent acid load neutralized (—) at Station 4 on Left Fork of Little Sandy Creek compared to the limestone remaining in the stream (- - -).

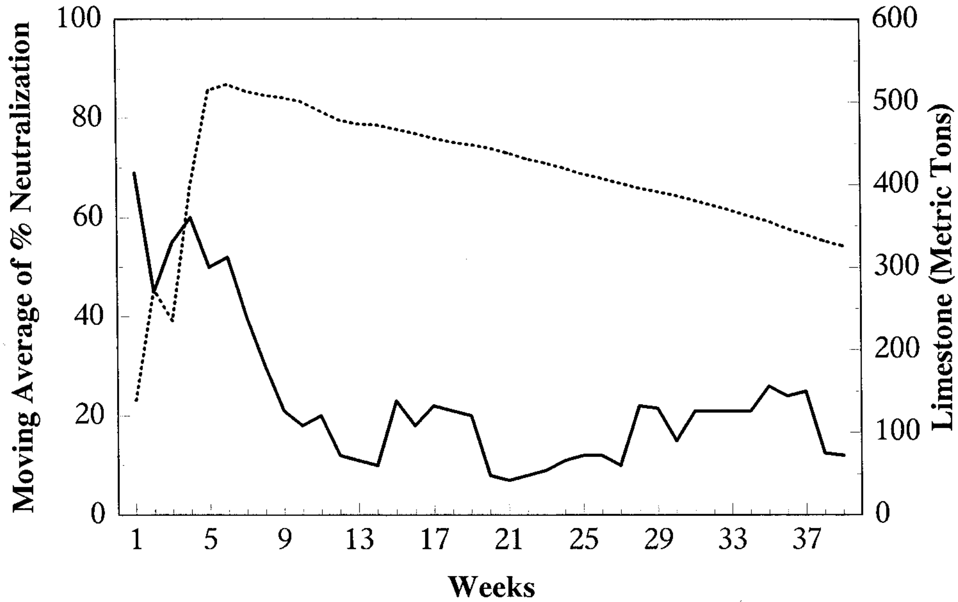

Figure 11. Moving average of percent acid load neutralized (—) at Station 3 on Left Fork of Little Sandy Creek compared to the limestone remaining in the stream (- - -).

During the first treatment period (April 25 to May 21, 1996), the untreated acid load at Station 4 was estimated at 225 metric tons with the amount of acid neutralized estimated at 151 metric tons or 67%. During the second treatment period, the untreated acid load was estimated at 1019 metric tons of which 142 metric tons (14%) were neutralized. For the entire treatment period, the untreated acid load was estimated at 1244 metric tons of which 292 metric tons (24%) were neutralized. Figure 10 shows the weighted moving average of percent neutralization at Station 4. Little additional neutralization occurred between Station 3 and Station 4. At Station 3, 22% of the acid load was neutralized. Figure 11 shows the weighted moving average of percent neutralization at Station 3.

The mean values (and ranges) for various chemical and physical parameters are shown in Table III. An analysis of metal and sulfate loads during the treatment period at Station 4 showed no significant changes except for calcium which increased significantly. Neutralization was highest during the early part of the treatment when flows were highest. Similar to the South Fork of Greens Run, neutralization of the acid load was lower during the low flow period but increased with increased flows. Depletion rates of limestone were significantly less than the South Fork and by the end of data collection, an estimated 404 metric tons of the limestone had been consumed with an estimated 326 metric tons still remaining in the stream.

5. Discussion

The direct instream application of fine-grained limestone significantly reduced the acid loads in two streams impacted by acid mine drainage. Although acid loads were reduced in both streams, overall neutralization was better in the South Fork of Greens Run compared to the Left Fork of Little Sandy Creek, presumably due to the combined effects of differences in $CaCO_3$ content and particle size distribution of the aggregates. Sherlock (1997) found that particle size range is important in determining neutralization effectiveness of the material in laboratory tests whereas differences in $CaCO_3$ content resulted in little difference in neutralization effectiveness when the particle sizes were similar.

Both streams showed similar flow patterns over the study with higher flows recorded during the early part of the treatment period. This corresponded to the period of highest neutralization. Efficiencies declined during summer low flow, but improved with increased flow. A possible cause for lower efficiencies during low flow may be due to the limestone particles being covered with oxyhydroxides of iron and aluminum that reduced contact time. Increased flows would tend to dislodge this coating through bedload transport of the material thereby increasing the neutralization efficiency. Although the percent neutralization in the Left Fork was consistently low, neutralization progressed at a fairly constant rate. Data for the South Fork showed that, although overall higher levels of acid neutralization occurred, neutralization steadily declined over the course of the study. The depletion rate of the limestone in South Fork was more rapid and by the end of the study, little limestone remained. In the Left Fork, it was estimated that nearly half of the limestone remained. The rapidly declining amount of limestone in the South Fork would have resulted in less contact over time, especially if a significant quantity were mixed into the substrate, resulting in declining neutralization efficiencies.

The results from this study confirm the effectiveness of sand-sized limestone aggregate in significantly reducing acid loads. Results were similar to those obtained on several AMD streams in the Middle Fork River drainage (Zurbuch *et al.*, 1997). Sediment analysis and the estimated acid load reduction in Cassity Fork during the Middle Fork River treatment indicated nearly 100% dissolution of the limestone sand. The South Fork of Greens Run showed similar results with nearly 100% of the limestone sand being dissolved by the end of the study. In the Left Fork of Little Sandy Creek, which was treated with a lower grade of limestone and a larger particle size distribution, dissolution rates were significantly less. The greater effectiveness of the high quality sand-sized limestone aggregate was also shown in the treatment of AD-impacted streams (Clayton *et al.*, 1998).

The anticipated coating of the sand-size limestone aggregate by ferric hydroxide precipitates was not a problem. Even though it appeared that the precipitate did cover the limestone sediment as a floc during very low flow periods when the neutralization efficiency was reduced, neutralization efficiencies increased as flows uncovered the limestone particles.

Based on the findings of this study, the treatment of streams severely affected by AMD may prove more effective if the streams are treated at more frequent intervals. Monthly limestone sand treatment with an amount equivalent to that consumed the previous month may be sufficient for complete treatment of the streams studied. Monthly treatments following an initial treatment with limestone equal to two times the individual stream's highest estimated monthly acid load would provide fresh limestone and allow for greater contact time, especially during low flow periods when limestone already in the stream may be buried in the substrate or covered to some extent by iron or aluminum precipitates.

6. Conclusions

The successful use of sand-sized limestone aggregate as a neutralizing agent, coupled with a high dissolution efficiency, gives a decided economic advantage to instream AMD treatment when compared to costs of other active and passive systems (Table IV). An exception to this could be the use of open limestone channels that are under study (Ziemkiewicz *et al.*, 1996). It is estimated that the two streams studied could be treated on a monthly basis for $<$U.S.$50.00 per metric ton of acid neutralized. Capital investment would be limited to development of access sites if needed to place limestone in the streams. The annual cost of treatment would be

TABLE IV

Cost comparison of various methods currently used to treat acid mine drainage

Treatment	U.S. $/Metric ton of acid neutralized	Efficiency
Limestone sand	<$50.00	>70%
Sodium hydroxide[a]	>$700.00	100%
Anhydrous ammonia[a]	>$300.00	100%
Hydrated lime[a]	>$175.00	90%
Open limestone channel[b]	>$20.00	50%
Various passive systems[b]	$500.00	?
Rotary limestone drums[c]	$90.00	80%
Powder slurry doser[d]	$115.00	80%
Pre-mix slurry doser[e]	$153.00	>90%

[a] Skousen *et al*. (1996).
[b] Ziemkiewicz (1995).
[c] Zurbuch *et al*. (1997).
[d] Olem (1991).
[e] Sverdrup (1986).

the cost of limestone sand to replace that consumed by the acid. In terms of utility, the limestone sand treatments are easily modified to match changing water quality conditions. For example, as future reductions are made in acid loading from land reclamation, passive treatments and other technologies, subsequent reductions in the amount of limestone sand could be made.

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