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Potential remediation measures for acidic irrigation water derived from the Alamosa River in south-central Colorado

by

George A. Desborough[1] and William H. Raymond[1]

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ABSTRACT

The Alamosa River headwaters have been the focus of attention with regard to acidic and heavy-metal pollution since the abandonment of the Summitville gold mine by the operators in late 1992. The abandoned gold mine lies near the head of Wightman Fork, which contributes less than 20 percent to the volume of the Alamosa River. The water of the Alamosa River is stored in Terrace Reservoir for use in irrigating about 45,000 acres of cropland. During times when the irrigation water is acidic (e.g., pH = <4.5), there is significant corrosion of metal irrigator parts such as well casings, pipes, and metal center-pivots. In addition to the acidity problems, the concentrations of dissolved copper sometimes exceed the agricultural limit of 0.2 mg/L set by the Colorado Department of Health and Environment.

Recent studies of natural zeolite—specifically clinoptilolite-rich rocks (CRRs)—have shown that some have a strong cation-exchange affinity for both H^+ and heavy metals such as copper, lead, and zinc. CRRs from Creede, Colo., and South Dakota were tested for their ability to decrease acidity and capture Cu from two irrigation ditches during one test in June and another in July 1994. Because the Creede, Colo., CRR performed poorly for both H^+ and Cu capture, the South Dakota CRR and Leadville limestone were used for a third test in August.

During the June test, which coincided with near-peak of spring runoff, the pH of the water at both test sites was in the range of about 5.3-6.4 and the Cu content was 0.1 mg/L, which is acceptable for domestic water supply. Both CRRs captured only insignificant amounts of H^+, but neither captured detectable Cu.

In July the pH at both test sites was about 3.7-4.0 and Cu concentrations ranged from about 0.7 to 0.9 mg/L. The South Dakota CRR raised pH to the range of 5.2-7.6 and captured an average of 55 percent of the dissolved Cu using 1 kg of CRR to treat 306 L of water at one site. At the second site, the South Dakota CRR raised pH to the range of 5.7-6.4 and captured an average of 42 percent of the Cu using 1 kg of CRR to treat about 470 L of water. The Creede, Colo., CRR raised pH to 4.1-4.7 and captured only 10 percent of the Cu at the first site using 1 kg of CRR to treat about 240 L of water. At the second site, Creede CRR raised the pH to 4.7 and captured about 10 percent of the Cu using 1 kg of CRR to treat about 520 L of water.

During the August tests, the pH of ditch water ranged from about 3.8 to 4.0 and Cu contents ranged from 0.7 to 0.9 mg/L. The South Dakota CRR raised pH to the range of 5.5-6.8 and captured an average of 44 percent of the Cu at the first site using 1 kg of CRR to treat 263 L of water. At the second site, this CRR raised pH to 5.6-6.4 and captured 60 percent of the Cu using 1 kg of rock to treat about 370 L of water. The Leadville limestone raised pH to a range of only 3.9-4.6 at both sites and the Cu content of the water was not changed. In retrospect, smaller grain sizes of both CRR and limestone would have performed much better in the field tests.

Laboratory studies indicate that very finely ground limestone or powdered lime [$Ca(OH)_2$] might be used effectively for remediation of acidity of Terrace Reservoir irrigation water. Because the Cu content did not exceed the 1 mg/L standard of the Safe Drinking Water Act during these 1994 tests, it is presumed that Cu will not be adversely affecting domestic water supplies in this area unless the concentrations rise. However, this should be examined by field tests.

INTRODUCTION AND PERSPECTIVE

This study was done to better understand the interaction of the natural zeolite-clinoptilolite and limestone with acidic and sometimes metal-polluted irrigation waters derived from the Alamosa River headwaters and stored in the Terrace Reservoir in northern Conejos County, Colo. (fig. 1). The Alamosa River water is used for irrigation of croplands of alfalfa, barley, and potatoes in an area of about 45,000 acres in this area (Stout and others, 1994). Miller and McHugh (1994) measured the pH of the Alamosa River headwaters in July 1993 and found that the most acidic streams were on the north side of the river. These streams, which include Iron Creek, Alum Creek, Bitter Creek, and Wightman Fork, are mostly natural acidic drainages caused by extensive iron sulfide alteration associated with the Platoro caldera complex. These streams also have relatively low concentrations of dissolved calcium (<30 mg/L), thus providing irrigation water with only small amounts of dissolved calcium for plant growth. Low calcium concentrations have adverse effects on trout tolerance of heavy metals, and water pH less than about 6.5 also has adverse effects on trout viability.

Since the abandonment of the Summitville gold mine at the headwaters of Wightman Fork in late 1992, considerable attention has been focused on manmade pollution of the upper Alamosa River. Drainage from Wightman Fork appears to contribute less than about 20 percent of the total flow of the Alamosa River at the confluence of these two drainages, based on observations at low flow in August 1993 and 1994 and at high flow in June 1994. The adverse publicity concerning the abandoned Summitville mine and its contribution of cyanide and heavy-metal pollution to the Alamosa River caused alarm to the public sector, to environmental activists, and to those using Alamosa River water for irrigation and drinking water. As a consequence, both Federal and State agencies responded by conducting geological and geochemical investigations of the Alamosa River watershed in 1993 and 1994. Preliminary results of some of these investigations were published in 1994 (Erdman and Smith, 1994; Plumlee and others, 1994; Plumlee and Severson, 1994; Stout and others, 1994), and more recently results were published in Colorado Geological Survey Special Publication 38, entitled "Proceedings: Summitville Forum '95."

The main drainage basins in the headwaters of the Alamosa River above Wightman Fork have not had mineral production efforts during the past century, although exploration pits at the turn of the last century produced some small waste dumps that have since been naturally leached for at least 100 years. Natural metal-rich acid drainage has been occurring in upper Alamosa River headwaters above Wightman Fork for many thousands of years prior to mining in the area (Miller and McHugh, 1994; Bove and others, 1995; Kirkham and others, 1995).

Since the spring of 1993, the Environmental Protection Agency (EPA) has made a sustained effort to remediate the acidity, cyanide, and heavy-metal-pollution problems at the abandoned Summitville mine site at the headwaters of Wightman Fork. Thus, the chemical composition and pH of Wightman Fork waters have been influenced greatly by these EPA pollution remediation measures which will continue for several years or longer.

During the last two years, field experiments with clinoptilolite-rich rocks (CRRs) in heavy-metal-polluted drainages in Colorado have shown that some CRRs have a high affinity for capture of copper, lead, and zinc by ion-exchange reactions (Desborough, 1993; 1994). In addition, some CRRs selectively capture H^+ and thus increase the pH of acidic water (Desborough, 1993). Considering these aspects, it was decided to test some CRRs in the irrigation drainage waters from the Terrace Reservoir to determine if some remediation measures could solve both the acidity and heavy-metal-pollution problems of these waters.

ANALYTICAL METHODS

All water samples were analyzed using atomic absorption methods by Skyline Laboratories, Inc., Wheat Ridge, Colo. Sodium and magnesium analyses were determined by Skyline Laboratories, Inc., using inductively coupled plasma methods. All of the other elements in clinoptilolite-rich rocks were analyzed using energy dispersive X-ray methods (Yeager and Quick, 1992).

PRELIMINARY STUDIES IN 1993

During the latter part of June 1993, Larry Gough (USGS) placed 20 g of sized (2.0-4.7 mm) raw CRR samples in nylon mesh bags in the flowing water of the Terrace headgate and in an irrigation ditch about 10 km downstream (Widger pond inlet) from the Terrace headgate (fig. 1). The largest amounts of copper and zinc were captured by the South Dakota CRRs (table 1).

Table 1.—Concentrations of copper and zinc captured from irrigation water by five samples of raw clinoptilolite-rich rock (2.0-4.7 mm) at the Terrace headgate and at the Paul Widger pond inlet ditch in northern Conejos and southern Rio Grande Counties, Colo., in late June 1993

Raw CRR	Cu	Zn
	parts per million	
Terrace headgate		
6/24/93-6/29/93 (5 days)		
Fort LaClede, Wyo. (FLW)	625	140
South Dakota, 1 (SD1)	980	165
South Dakota, A (SDA)	925	165
South Dakota, C (SDC)	925	125
Tilden, Tex. (TT)	330	50
Widger pond inlet		
6/29/93 (<24 hours)		
FLW	535	155
SD1	1,065	150
SDA	1,020	155
SDC	715	170
TT	325	40

Three similar tests were done with seven CRRs in August 1993 during very small flow volumes in the Alamosa River. One test was done in the Alamosa River just above the confluence with Wightman Fork, another was done at the mouth of Wightman Fork, and a third was done at the Terrace site (fig. 1). Data for concentrations of selected elements in the water, plus certain elements gained or lost by the CRRs, are given in table 2. For the Alamosa River above the confluence with Wightman Fork, analyses for Cd, Cu, Zn, and Pb in the water showed less than 0.1 mg/L; Ca was 13.1 mg/L and the pH was 6.6 on 8/10/93 and 7.0 on 8/11/93.

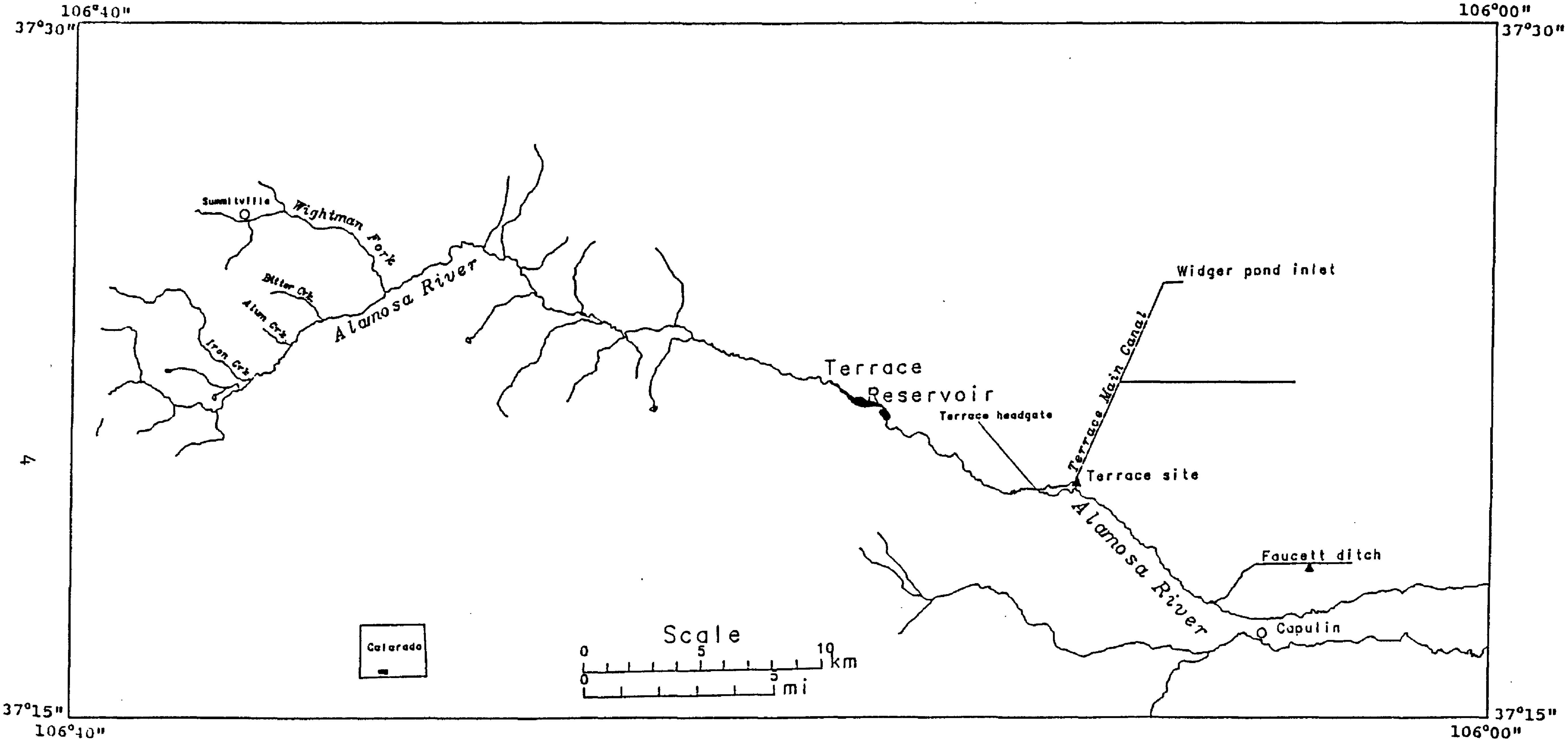

Figure 1. Terrace irrigation study area and part of the Alamosa River drainage basin.

Data in table 2 show that for 1993 tests in Wightman Fork the South Dakota (SDA) CRR captured 380 ppm of Cu in less than two days, which is more than three times the Cu captured by the other CRRs. The Fort LaClede, Wyo. (FLW), CRR captured large amounts of Ca, which is detrimental to the nutrient value of crop irrigation water and also adversely affects trout habitat. In addition, the SDA CRR contributed significant potassium to the water that would be beneficial to croplands.

Data in table 2 for 1993 tests in Terrace irrigation water show that the SDA CRR captured 3.6 times more copper from the irrigation water than did any other CRR. The amount of dissolved Cu (0.2 mg/L) in this water is at the upper limit of the agricultural (irrigation and stock watering) standard, according to the "Colorado Basic Stream Standards for Metals" (Colorado Department of Health, 1989, table 5), and it exceeds the "Class 1 Aquatic Life" standards by a factor of 20, partly because of the low Ca content (< 100 mg/L of $CaCO_3$). Both the FLW and SDA CRRs captured much more Ca from the irrigation water than did the other CRRs.

Table 2.—Concentrations of elements in the water and elements gained or lost (-) by raw clinoptilolite-rich rocks (2.0-4.7 mm) at Wightman Fork and the Terrace site in August 1993

[n.d. = not determined]

Raw	K	Ca	Cu	Zn	Rb	Sr	Cd	Ba	Pb
CRR*	ppm × 10^3		parts per million						
Wightman Fork									
8/10/93-8/11/93 (1.7 days) pH = 6.5-7.2, T = 9-16°C									
water (8/10/93) (mg/L)	4.8	108	2.8	1.2	n.d.	n.d.	0.01	0.03	<0.01
FLW	0.3	0.8	115	10	0	45	<1	295	0
FCEN	-0.3	0.3	40	0	-30	25	<1	50	0
HLO	-0.5	0	105	0	0	20	<1	15	0
PBC	-0.2	0.1	0	0	-25	-20	<1	150	0
SDA	-0.6	0.1	380	50	-10	0	<1	120	0
SVO	-1.1	0	25	0	-20	25	<1	25	0
Terrace site									
8/10/93-8/11/93 (1.8 days) pH = 7.8, T = 14.5-20°C									
water (8/11/93) (mg/L)	1.1	26.8	0.2	0.2	n.d.	n.d.	<0.01	0.07	<0.01
CCO	-0.3	0	35	0	-25	125	<1	15	0
FLW	0	1.1	20	55	-15	210	<1	-190	0
FCEN	0.1	0.2	60	0	-30	75	<1	20	0
HLO	-0.2	0.1	60	55	10	60	<1	50	0
PBC	0.2	0.1	0	0	-15	55	<1	-110	0
SDA	-0.1	0.9	220	55	-25	130	<1	185	0
SVO	-0.4	0.1	50	10	-45	90	<1	105	0

CRR* abbreviations: CCO, Creede, Colo.; FLW, Fort LaClede, Wyo.; FCEN, Fish Creek, Nev.; HLO, Harney Lake, Ore.; PBC, Princeton, B.C.; SDA, South Dakota A; SVO, Sheaville, Ore.

Because of the results of preliminary tests of CRRs in the Alamosa River drainage described above, it was decided that the best acidic-remediation CRR from South Dakota would be tested against the Creede, Colo., CRR. The Creede CRR (CCO) occurs in significant amounts (>1 million tons) less than 70 road miles from the Terrace irrigation area. Prior tests of the CCO CRR (table 2) for copper capture showed it to be six times less effective than the South Dakota CRRs; however, the high cost of transportation of the South Dakota CRR justified the testing of the Creede CRR.

Table 3 gives the concentrations of major and trace cations in the Creede, Colo., and the South Dakota CRRs.

Table 3.—Concentrations of major and trace cations in raw Creede, Colorado (CCO), and raw South Dakota (SDA) clinoptilolite-rich rocks (CRR) used for irrigation water studies in the Alamosa River drainage

CRR	Na	Mg	K	Ca	Fe	Cu	Zn	Rb	Sr	Pb	Ba
	weight percent					parts per million					
CCO	0.49	0.40	3.2	1.8	0.8	0	25	190	510	<40	590
SDA	1.30	0.54	3.3	1.9	1.4	0	70	95	315	<40	760

INVESTIGATIONS IN 1994

Experimental Methods

All collection, treatment, and treated-water containment vessels were nonmetallic (e.g., Tygon, PVC, or plastic). All tests were done under siphon conditions requiring a water head of about 1 m. Principal elements of field studies were (1) collection of raw water, (2) routing of irrigation water through experimental treatment, (3) containment and retention of clinoptilolite-rich rock (CRR) used for water treatment, and (4) collection of treated irrigation water. Raw irrigation water for analysis was collected in Nalgene bottles within 0.5 m of the ditch-water treatment intake; these water samples were acidified (1 drop conc. HNO_3/10 mL) and refrigerated to <10°C prior to analysis. Irrigation ditch water was routed by pipe to a ball or gate valve ahead of the 1.4-L CRR container. The water entered the inside of the container at the bottom and exited from the highest point, enabling the system to purge itself of entrapped air. Water routed through the CRR was then collected in approximately 115-L covered containers and retained until the treated water could be sampled using Nalgene bottles. After each sampling, the water was disposed of prior to the next treatment. After the first two days of the first test period, it was determined that an additional 75- or 115-L covered container would be required for longer test intervals. The second container was coupled to an outlet on the original 115-L container.

The first experiment conducted in June 1994 was largely a test of field setup and sampling. The South Dakota (SDA) and the Creede, Colo. (CCO), CRRs were used at two sites. Each 1.4-L CRR container was filled with 1 kg of sized CRR, consisting of 30 weight percent of 2.0-4.7-mm material and 70 weight percent of 4.7-12.7-mm material. The CRR was mixed in the laboratory prior to placement in the containers. The dry CRR in each of four containers was flushed with tap water (pH = 7) 2-24 hours prior to exposure to irrigation water to remove adhering particles and air and to promote rapid ditch water-CRR interaction. Values reported for pH are the mean of measurements of four separate water samples using a digital temperature-corrected meter.

Contrary to our expectations, almost 40 percent of the 44 tests in June, July, and August failed to have the continuous flow rates during the period of testing. The measured initial valve settings of each CRR container were between 4.5 L/h (75 mL/min) to 8.4 L/h (140 mL/min). Departures from values less than 4.5 L/h were chiefly due to accumulation of Fe-hydroxides that plugged the valve openings during the June and July test periods. For the August tests, a filtering system was placed ahead of the flow-control valves in order to eliminate the Fe-hydroxide accumulation problem. However, low flow rates occurred for other reasons—one was a break in the siphon at the Faucett site during the last test due to a curious passerby; at the Terrace site, very low flow in the ditch limited the water siphon head so that only a low flow rate (<6 L/h) could be maintained for each sample container. During the last Terrace site test, the siphon was inadvertently broken due to an unknown reason.

June 1994 Tests

In near-peak runoff in the first week of June, the irrigation ditch water ranged in pH from 5.35 to 6.4 at the Terrace site. At the same time, the ditch water at the Faucett site ranged in pH from 6.1 to 6.4. The Faucett ditch water is derived from a site about 7 km downstream from the point of origin of the Terrace headgate water (fig. 1). The Terrace site is about 2 km from the point of river origin, whereas at the Faucett site the ditch water has traveled about 7 km from its origin at the river.

Results of the June 1994 side-by-side tests of the SDA CRR for ditch water at the Faucett site are given in table 4. In both tests (SDA 1 and SDA 2) the pH of the ditch water (pHi, table 4) was raised to near-neutral or above through the fourth test. This amounted to treatment of 318 to 347 bed volumes, bringing the pH to near neutral. The raw ditch water contained only 6.6-7.5 mg/L of calcium (mean = 6.8) and 0.1 mg/L of copper. Quantitative analysis of the SDA CRR for Mg, Na, Ca, and K, before and after exposure to ditch water, only showed a loss of 0.2 weight percent of Na; concentrations of the other elements were unchanged. The captured H^+ apparently displaced Na. The concentration of Cu (0.1 mg/L) does not exceed the amount allowed by the Colorado Department of Health (1989) for the agricultural standard of 0.2 mg/L (stock watering and irrigation). Table 4 shows that for both of the fifth tests the capture of H^+ was significantly less than that of previous tests. This may indicate that the CRR was H^+-saturated for a pH of about 6.

Results of the June 1994 side-by-side tests of the CCO CRR for the Terrace site are given in table 5. In both tests (CCO1 and CCO2) the pH of the ditch water was raised slightly, but in none of the eight tests was it raised as high as 6.5, even though 5.4×10^{-4} moles of H^+ were captured during a total of four tests, and 433 to 496 bed volumes of water were treated. The raw ditch water (see Appendix) contained only 6.2-6.9 mg/L of calcium (mean = 6.5). For the first three tests (days) the Cu concentration was 0.1 mg/L; for the next two tests it was 0.2 mg/L (see Appendix), which is the upper limit allowed for agricultural water. Analysis of the CCO1 and CCO2 before and after exposure to the water showed a loss of only 0.02 (CCO1) and 0.07 (CCO2) weight percent of Na. No detectable Cu was captured by the CCO CRR.

Table 4.—Results of June, 1994 tests of South Dakota (SDA) clinoptilolite-rich rocks showing interactions with irrigation water at the Faucett site

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour = L/h; mol H^+ = moles of hydrogen; L = liters of water treated; pHi = initial ditch water pH; pHf = final pH of treated water; values in parentheses are calculated and are not actual values]

	Faucett site side-by-side tests of SDA1 and SDA2					
Serial tests	**time, h**	**bv/h**	**mol H^+ $\times 10^{-4}$**	**L (measured)**	**pHi**	**phf**
SDA1						
First	15.3	4.5	0.27	68	6.3	6.9
Second	21.5	(3.1)	0.27	68	6.4	8.1
Third	25.3	4.5	0.79	113.3	6.2	8.0
Fourth	29.3	(2.4)	0.55	69	6.1	6.85
Fifth	23.3	(3.0)	0.0	71	6.1	6.1
Totals			1.9	318		
SDA2						
First	15.3	4.9	0.38	75.5	6.3	7.4
Second	21.5	(4.1)	0.35	88.7	6.4	8.0
Third	25.3	4.5	0.79	113.3	6.2	7.3
Fourth	29.3	(2.8)	0.42	70.0	6.1	6.7
Fifth	23.3	(2.0)	0.14	45.4	6.1	6.3
Totals			2.1	393		

Table 5.—Results of June 1994 tests of Creede, Colo. (CCO), clinoptilolite-rich rocks showing interactions with Terrace irrigation water

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour = L/h; mol H^+ = moles of hydrogen; L = liters of water treated; pHi = initial ditch water pH; pHf = final pH of treated water; values in parentheses are calculated, not actual]

	Terrace side-by-side tests of CCO1 and CCO2					
Serial tests	**time, h**	**bv/h**	**mol H^+ $\times 10^{-4}$**	**L (measured)**	**pHi**	**pHf**
CCO1						
First	18.3	4.5	0.04	81.1	6.35	6.4
Second	25.5	4.5	1.15	115.3	5.9	6.4
Third	28.5	(4.3)	2.58	123	5.35	5.6
Fourth	24.3	4.7	1.70	113	5.55	5.8
Totals			5.44	433		
CCO2						
First	18.3	5.8	0.04	106.3	6.35	6.4
Second	25.5	4.8	2.32	121.9	5.9	6.4
Third	28.5	(3.1)	2.3	88.4	5.35	5.55
Fourth	24.3	7.4	4.7	179.6	5.55	6.1
Totals			9.4	496		

In summary, for the 18 field tests done in June with the SDA and CCO CRRs, each at two different sites, the results seem consistent within analytical uncertainty and the inhomogeneity factors involved in analyzing the CRR.

July 1994 Tests

The equipment and procedures used in the July test were similar to those of the June tests, except that at the two sites the SDA and CCO CRRs were tested side-by-side. The ditch water pH at the two sites varied from 3.7 to 3.95 (tables 6 and 9) and was much more acidic than in early June near-peak runoff (tables 4 and 5).

Table 6 gives the results for the ditch water treatment at the Faucett site, using the CCO and SDA samples (1 kg in each CRR container). The CCO CRR treated about 238 L of acidic water and raised the pH to a maximum of 4.7 for flow rates of 2.2-5.1 bv/h and captured 240 × 10^{-4} moles of H^+ (table 6). The SDA CRR treated more water and captured more than twice the amount of H^+ than did the CCO CRR. The capture of H^+ with regard to final pH (pHf, table 6) is more than a 10 factor less for the CCO CRR than that of the SDA CRR. This difference may be due, in part, to the lower flow rates for the CCO CRR (table 6) relative to those of the SDA CRR.

Table 6.—Results of July 1994 tests of Creede, Colo. (CCO), and South Dakota (SDA) clinoptilolite-rich rocks showing interactions with ditch water at the Faucett site

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour = L/h; mol H^+ = moles of hydrogen; L = liters of water treated; pHi = initial ditch water pH; pHf = final pH of treated water; values in parentheses are calculated, not actual]

	Faucett site side-by-side tests of CCO and SDA					
Serial tests	**time, h**	**bv/h**	**mol H^+ × 10^{-4}**	**L**	**pHi**	**pHf**
CCO						
First	21.5	(2.2)	104	47.5	3.7	4.7
Second	21.0	(3.6)	27	75.7	3.95	4.1
Third	22.8	5.1	109	115.4	3.9	4.4
Totals	65.3		240	238.6		
SDA						
First	21.5	4.9	254	106.0	3.7	7.6
Second	21.0	(3.0)	76	63.4	3.95	6.7
Third	22.8	6.0	181	136.3	3.9	5.2
Totals	65.3		511	305.7		

Table 7 shows the abundances of calcium and copper in the Faucett irrigation water and the amounts of these two elements removed in the treated water, based on analysis of raw and treated water, respectively. The SDA CRR removed about seven times the amount of calcium than did the CCO CRR, which would prove detrimental to the already Ca-deficient soils under irrigation in the area. However, the SDA CRR captured 53 percent of the copper from the water and removed about eight times more copper than the CCO CRR (table 7).

Table 7.—Calcium and copper in ditch water at the Faucett site and the amounts of calcium and copper removed during each of three tests for the Creede, Colo., and South Dakota clinoptilolite-rich rocks, based on analysis of raw and treated water

[vol. = water volume; L = liters; Ca = calcium; Cu = copper]

Date	**Vol.**	**Raw water**		**Treated water**	**Raw water**		**Treated water**
	L	**Ca**		**Ca, removed**	**Cu**		**Cu, removed**
		mg/L	**mg**	**mg**	**mg/L**	**mg**	**mg**
		Creede, Colo., clinoptilolite-rich rock					
7/6/94	47.9	17.4	833	0	0.71	34	2
7/8/94	75.7	18.0	1,362	114	0.77	58	8
7/9/94	115.4	18.7	2,158	173	0.82	95	5
Totals	239		4,353	282		187	15
		South Dakota clinoptilolite-rich rock					
7/6/94	106.0	17.4	1,844	116	0.71	75	35
7/8/94	63.4	18.0	1,141	799	0.77	49	30
7/9/94	136.3	18.7	2,549	1,172	0.82	112	61
Totals	305.7		5,534	2,087		236	126

Table 8 gives analytical results for the two CRRs exposed to ditch water after the third test. Sample inhomogeneity (50 g CRR samples from 1 kg) is probably the cause of the discrepancy in Ca and Cu values between tables 7 and 8. In contrast, small water samples from a large volume of water are more homogeneous. Even so, the values for copper capture differ by less than 25 percent (tables 7 and 8) in terms of water and rock concentrations.

Table 8.—Gains and losses (-) of magnesium, sodium, potassium, calcium, copper, zinc, and barium by clinoptilolite-rich rocks (CRR) exposed to ditch water at the Faucett site after the third test

CRR	**Mg**	**Na**	**K**	**Ca**	**Cu**	**Zn**	**Ba**
	weight percent				parts per million		
CCO	-0.04	0.31	0.2	0.06	0	0	115
SDA	0.04	0.30	0.3	0.4	155	30	-85

Table 9 gives analytical results for the Terrace site. The CCO CRR treated about 520 L of ditch water and raised the pH to 4.7 in all three tests at flow rates of 7.7-8.4 bv/h (table 9). The SDA CRR treated about 470 L of water and raised the pH to about 6.1 for three tests at flow rates of 5.5-8.4 bv/h. While the total amounts of H^+ capture are within about five percent of each other, the final pH of SDA-CRR-treated water (pHf, table 9) is an order of magnitude higher than that of the water treated by the CCO CRR.

Table 9.—Results of July 1994 tests of the Creede, Colo., and South Dakota clinoptilolite-rich rocks showing interactions with ditch water at the Terrace site

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour = L/h; mol H^+ = moles of hydrogen; L = liters of water treated; pHi = initial ditch water pH; pHf = final pH of treated water]

		Terrace side-by-side tests of CCO and SDA				
Serial tests	**time, h**	**bv/h**	**mol H^+ $\times 10^{-4}$**	**L**	**pHi**	**pHf**
CCO						
First	21.5	8.4	263	179.8	3.9	4.7
Second	20.5	8.3	197	170.0	3.9	4.7
Third	22.0	7.7	240	170.0	3.85	4.7
Totals	64		700	519.8		
SDA						
First	21.5	8.4	305	179.8	3.9	6.45
Second	20.5	8.3	237	170.0	3.9	6.2
Third	22.0	5.5	197	121.1	3.85	5.7
Totals	64		739	470.9		

Table 10 gives the amounts of calcium and copper in the Terrace site ditch water and the amounts of these two elements removed or gained in the treated water, based on analysis of raw and treated water. The SDA CRR removed almost 0.2 weight percent Ca, whereas the CCO CRR contributed 0.04 weight percent Ca to the water. The SDA CRR removed about 37 percent of the copper, whereas the CCO CRR removed less than 10 percent.

Table 11 shows the results of analysis of the two CRRs exposed to ditch water after the third test. In view of the CRR sample inhomogeneity problems, there is generally good agreement between the Ca data in tables 10 and 11. For Cu in water treated by the CCO CCR (39 mg) the agreement with the rock analysis is good (55 ppm). For Cu in the water treated by the SDA CRR the amount of copper removed from the ditch water (141 mg) is in poor agreement with the results of the CRR analysis (95 ppm).

Table 10.—Calcium and copper in ditch water at the Terrace site and the amounts of calcium and copper gained or lost (-) in each of the three tests for the Creede, Colo., and South Dakota clinoptilolite-rich rocks, all based on analysis of raw and treated water

[vol. = water volume; L = liters; Ca = calcium; Cu = copper]

Date	Vol. L	Raw water Ca mg/L	Raw water Ca mg	Treated water Ca, gained or lost, mg	Raw water Cu mg/L	Raw water Cu mg	Treated water Cu, removed mg
			Creede, Colo., clinoptilolite-rich rock				
7/7/94	179.8	17.4	3,128	72	0.78	140	0
7/8/94	170.0	17.5	2,975	51	0.81	138	8
7/9/94	170.0	18.0	3,060	289	0.86	146	31
Totals	519.8		9,163	412		424	39
			South Dakota clinoptilolite-rich rock				
7/7/94	179.8	17.4	3,128	-360	0.78	140	20
7/8/94	170.0	17.5	2,975	-680	0.81	138	37
7/9/94	121.1	18.0	2,180	-812	0.86	104	84
Totals	470.9		8,283	-1,852		382	141

Table 11.—Gains and losses (-) of magnesium, sodium, potassium, calcium, copper, zinc, and barium by clinoptilolite-rich rocks (CRR) exposed to ditch water at the Terrace site after the third test

CRR	Mg	Na	K	Ca	Cu	Zn	Ba
		weight percent				parts per million	
CCO	-0.1	0.25	-0.1	0.10	55	20	95
SDA	0.04	0.50	0.1	0.20	95	35	65

In order to evaluate the effect of grain size, the CRRs in each of the four tests were sieved to separate the 2-4.7-mm fraction from the 4.7-12.7-mm material. Analysis of these fractions showed that the finer grained material captured about three to five times more copper than the coarser grained material.

August 1994 Tests

Because of the relative inefficiency of the Creede, Colo., CRR for remediation of acidity and Cu capture, it was decided to choose the Leadville limestone from west of Trout Creek Pass in Chaffee County, Colo., for the last field test side-by-side with the South Dakota CRR. One kg of the limestone, using 300 grams of the 2-4.7-mm material and 700 grams of the 4.7-12.7-mm material, occupied only 750 cc, as compared to 1,000 cc occupied by the same weight of the South Dakota CRR. Because of this difference, the calculated water flow rates (bed volume/hour) appear significantly different.

At the Faucett ditch site used in prior tests, the ditch flow was terminated due to ditch construction. We used water from the pond into which the ditch water had flowed.

Table 12 gives test results for the pond water and a wide range in flow rates. The SDA CRR treated 263 L of water and raised the pH to a weighted mean of 6.4. The Leadville limestone treated 271 L of water and raised the pH to about 4.4. The reason the limestone performed poorly was because the grain size was much too coarse, as will be shown later.

Table 12.—Results of August 1994 tests of South Dakota (SDA) clinoptilolite-rich rock (CRR) and Leadville limestone (LL) at the Faucett site

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour; H^+ = moles of hydrogen; L = liters of water treated; pHi = pond water pH; pHf = pH of treated water; numbers in parentheses are calculated, not actual values]

	Faucett site side-by-side tests of SDA and LL					
Serial tests	time, h	bv/h	mol H^+ $\times 10^{-4}$	L (measured)	pHi	pHf
SDA						
First	23	(4.0)	129	92.7	3.9	6.8
Second	19	6.7	236	126.8	3.8	5.5
Third	23.2	(1.9)	82	43.5	3.8	6.7
Totals	65.2		447	263.0		
LL						
First	23	7.0	126	121.0	3.9	4.5
Second	19	6.5	108	93.0	3.8	4.2
Third	23.2	(3.2)	92	57	3.8	4.6
Totals	65.2		326	271		

Table 13 shows the amounts of calcium and copper in Faucett pond water and the amounts of these two elements removed in the treated water, based on analysis of raw and treated water, respectively. The SDA CRR removed about 40 percent of the copper and 28 percent of the calcium. The limestone contributed about 0.2 mg/L of calcium to the water, but the copper content was unchanged.

Table 13.—Calcium and copper in pond water at the Faucett site and the amounts of calcium and copper removed during each of three tests for the South Dakota clinoptilolite-rich rock, based on analysis of raw and treated water

[vol. = water volume; L = liters]

Date	Vol. L	Raw water Ca		Treated water Ca, removed	Raw water Cu		Treated water Cu, removed
		mg/L	mg	mg	mg/L	mg	mg
8/3/94	92.7	26.7	2,475	1,001	0.85	79	42
8/4/94	126.8	26.9	3,411	786	0.85	108	32
8/5/94	43.5	26.9	1,170	213	0.80	35	13
Totals	261.0		7,056	2,000		222	87

Table 14 shows that the SDA CRR captured sodium, calcium, and copper, but the amounts of magnesium, potassium, zinc, and barium were not changed. The amounts of captured calcium are in good agreement with those of the water analyses (table 13), but the amount of copper is only half that shown by the water analyses. The difference in copper is probably due to sample inhomogeneity or sampling error for the small (50 g) sample taken from the 1 kg of CRR.

Table 14.—Gains and losses (-) of magnesium, sodium, potassium, calcium, copper, zinc, and barium by the South Dakota clinoptilolite-rich rock at the Faucett site after the third test

Mg	Na	K	Ca	Cu	Zn	Ba
	weight percent				parts per million	
0	0.7	0	0.15	40	0	0

Table 15 gives the results for tests at the Terrace site. The SDA CRR treated about 370 L of water and raised the pH to a range of 5.6 to 6.4. The limestone treated 389 L of water and raised the pH to a range of 3.85 to 4.3.

Table 16 shows the amounts of calcium and copper in the Terrace ditch water and the amounts of these elements in the treated water. The SDA CRR removed about 26 percent of the calcium and about 60 percent of the copper. No significant change in the calcium or copper content of the raw and treated water was observed for the limestone-treated water.

Table 15.—Results of August 1994 tests of South Dakota (SDA) clinoptilolite-rich rock and Leadville limestone (LL) at the Terrace site

[time, h = elapsed time for each test in hours; bv/h = bed volumes/hour; mol H^+ = moles of hydrogen; L = liters of water treated; pHi = initial ditch water pH; pHf = final pH of treated water; numbers in parentheses are calculated, not actual values]

	Terrace site side-by-side tests of SDA and LL					
Serial tests	**time, h**	**bv/h**	**mol H^+ × 10^{-4}**	**L measured**	**pHi**	**pHf**
SDA						
First	16.8	4.9	140	82.3	3.85	6.4
Second	23.5	5.2	262	121.0	3.75	5.6
Third	26.0	(4.3)	155	111.7	3.9	5.9
Fourth	23.0	(2.5)	79	57.7	3.9	5.6
Totals	89.3		636	372.7		
LL						
First	16.8	9.0	134	118.3	3.85	4.3
Second	23.5	7.5	66	132.5	3.75	3.85
Third	26.0	4.9	64	95.6	3.9	4.2
Fourth	23.0	(2.4)	28	42.6	3.9	4.2
Totals	89.3		292	389.0		

Table 16.—Calcium and copper in ditch water at the Terrace site and the amounts of calcium and copper removed during each of four tests for the South Dakota clinoptilolite-rich rock, based on analysis of raw and treated water

[vol. = water volume; L = liters]

Date	**Vol. L**	**Raw water Ca**		**Treated water Ca removed**	**Raw water Cu**		**Treated water Cu removed**
		mg/L	**mg**	**mg**	**mg/L**	**mg**	**mg**
8/2/94	82.3	27.2	2,238	584	0.9	74	62
8/3/94	121.0	27.6	3,340	1,004	0.9	109	66
8/4/94	111.7	28.0	3,128	715	0.7	78	45
8/5/94	57.7	27.5	1,587	410	0.8	46	17
Totals	372.7		10,293	2,713		307	190

Table 17 shows the gains in sodium, calcium, and copper and the losses of potassium and barium for the SDA CRR. Both the calcium and copper amounts are much less than the data for these indicated by the water analyses (table 16). This difference is attributed to the difficulty in obtaining a representative sample from the 1 kg of CRR used in the field tests. The water analyses are interpreted to be a more accurate representation of the amounts of calcium and copper captured by the CRR.

Table 17.—Gains and losses (-) of magnesium, sodium, potassium, calcium, copper, zinc, and barium by the South Dakota clinoptilolite-rich rock for the Terrace site after the fourth test

Mg	Na	K	Ca	Cu	Zn	Ba
weight percent				parts per million		
0	0.6	-0.2	0.15	35	0	-60

DISCUSSION OF FIELD TESTING

Field testing water flow rates were partly designed on the basis of laboratory studies of metal-rich and acidic mine water by Schultz and Zamzow (1993) and Vos and O'Hearn (1993) who used CRRs for laboratory studies of heavy-metal remediation. Schultz and Zamzow (1993) used a flow rate of 5 bv/h in all tests; Vos and O'Hearn (1993) used flow rates ranging from 1.3 to 5.4 bv/h. We therefore chose an optimum flow rate of about 5 bv/h for our field studies; however, flow rates during actual tests ranged from 4 to 8 bv/h.

The exposure periods of CRR to the Alamosa River irrigation water were insufficient because the SDA CRR was still capturing significant H^+ and Cu during the final tests in the July and August experiments.

LABORATORY TESTS

During the August test, we collected about 12 L of raw ditch water from the Terrace site for simple laboratory study of water-rock interactions with regard to pH. In one experiment, 5 g of SDA CRR (2-4.7 mm) was placed in a 1.4-L CRR container (flow from bottom upward like used in field studies) and 4 L of water was passed across the CRR at a rate of 100 mL/min. The 4 L of water with an initial pH of 3.9 was raised to 7.4 in 24.5 hours. This is not a fair test for field conditions, but it might be inferred that the SDA CRR could neutralize much more acidic water than was demonstrated in our field studies if longer test periods were conducted.

Tests of greater significance were done with finely ground Leadville limestone and ditch water (pH = 3.9) from the Terrace site. The first three tests were done in rock:water ratios of 1:4,000, 1:8,000, and 1:10,000 using limestone of mesh size less than 0.01 mm. These tests were done without agitation of sample and water, and the acidity was neutralized in less than 3 hours. A fourth test was done using <0.0045-mm diameter limestone in a ratio of 1:22,400; 0.1 g of limestone in 2.24 L of water brought the pH of the ditch water to pH of 6.3 in less than 30 minutes without agitation. After 3 days, when the pH of the water was 8, some of the limestone remained undissolved.

Additional laboratory tests were done using H_2SO_4-acidified tap water with finely ground Leadville limestone. These tests measured the pH of the acidified tap water sample each time measurement of the pH of the limestone-treated water was made. Also, the pH meter was calibrated at pH of 7 and 4 prior to measurements of the pH of each solution. The first of these tests used acidified water of 3.8 pH in a limestone:water ratio of 1:60,000 using 0.2 g of

<0.01-mm diameter limestone in 12 L of water; the pH of the water was raised to 5.4 in 10 minutes and to 6.3 in 155 minutes. For a second test, using a limestone:water ratio of 1:106,000 (0.12 g of <0.044-mm diameter limestone in 12.8 L of water) with an initial pH of 3.5, the pH was raised to 4.4 in 10 minutes and to 5.0 in 20 minutes; after 24 h and 48 h the pH was 5.6. Thus, finely ground Leadville limestone from the site sampled near Trout Creek Pass in Chaffee County will neutralize acidic water rapidly.

Other options for neutralization of acidic water include lime [$Ca(OH)_2$] or cement kiln dust that is dominantly lime. Both of these products are considerably less efficient for neutralization than high-calcium limestone (Pincomb and Shapiro, 1994, table 1). Costs versus efficiency will determine which could be best utilized in the subject application after field tests were conducted using acidic irrigation water. In any case, the costs of very efficient acidic neutralization products would be less than about $75 per ton delivered to Monte Vista, Colo. (based on current prices).

ACIDITY OF ALAMOSA RIVER HEADWATERS

The pH of four headwater tributaries of the Alamosa River and the Alamosa River just above the confluence of Wightman Fork were measured on the same day during each of the three testing periods (table 18). The values for 6/4/94 were near-peak spring runoff. The values for 8/3/94 were obtained after 2 days of heavy rainfall in the headwaters. It is clear from the pH data in table 18 that much or most of the acidic pollution in the Alamosa River is contributed from drainages upstream from Wightman Fork (which drains the Summitville mine area). This conclusion was inferred from the data of Miller and McHugh (1994) based on 8/16-20/93 pH measurements of Alamosa River headwater tributaries. Studies of Bove and others (1995) and Kirkham and others (1995) reached the same general conclusion.

Table 18.—Headwater pH of the Alamosa River and four tributaries during the three irrigation water testing periods

[The Alamosa River site is 0.4 km upstream from the confluence of Wightman Fork]

Dates	Iron Creek	Alum Creek	Bitter Creek	Wightman Fork	Alamosa River
6/4/94	6.2	3.0	4.3	3.7	4.8
7/8/94	3.1	2.4	2.7	3.7	4.1
8/3/94	3.2	2.6	3.0	3.7	4.4

Measurements of pH in 1993 by Stout and others (1994) for the Terrace area irrigation waters ranged from 5.3 to 6.7 during 6/10-15/93 and 7/16-19/93. These pH values for 7/93 are significantly higher than those reported here for the 7/94 measurements (tables 6, 9, and Appendix). We measured pH values of 7.8 on 8/11-12/93 (table 2) at the Terrace site. The pH of the Faucett ditch was 8 on 8/11/93, and the pH of the Alamosa River near Capulin was about 8 on 8/11/93. Walton-Day and others (1995) reported a pH range from 4.6 to 7.0 for the

Alamosa River just above Terrace Reservoir for April-September 1993. Smith and others (1995) report pH values of irrigation ditch water from the Terrace Reservoir as low as 4.1 for measurements in 1994. This appears to indicate that local climatic factors, such as heavy rainfall in the Alamosa River headwaters, may cause substantial increases in the acidity of the irrigation water. Ortiz and others (1995) have documented the effect of a localized rainstorm on the pH of the Alamosa River in August 1993 when the pre-storm pH upstream from Terrace Reservoir was 6.1, but the post-storm pH at the same site was 4.6.

SUMMARY

Based on our field and laboratory studies in 1993 and 1994, the following conclusions or inferences are provided:

1. The South Dakota clinoptilolite-rich rocks (CRR) are significantly more effective in reducing acidity and removing copper from Terrace Reservoir irrigation water than the Creede, Colo., clinoptilolite-rich rocks. The cost of using CRR to remediate acidity for such large volumes (1,000-2,000 gpm) of water is prohibitive for this case.
2. The use of a smaller grain size of both CRR and limestone would have been more effective in reducing acidity. Removal of copper by CRR is enhanced by using a smaller grain size.
3. Finely ground limestone is the most effective and most economical method of reducing the acidity of Terrace Reservoir irrigation water.
4. Remediation of acidity of Wightman Fork, a tributary of the Alamosa River, will have little influence on the acidity of the Terrace Reservoir water. However, removal of copper from the headwaters of Wightman Fork (at the Summitville mine) may eventually reduce the amounts of dissolved copper in the Terrace Reservoir.
5. The Alamosa River irrigation water was much more acidic in July and August 1994 than it was during the same months in 1993.

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APPENDIX

Data given here are for the pH and concentrations of Ca and Cu in raw ditch water and for Ca and Cu concentrations in water treated by or exposed to the rocks tested during the three 1994 test periods.

Table 19.—Copper and calcium in raw and treated water at the Faucett and Terrace sites for June, July, and August 1994 tests

[SDA = South Dakota clinoptilolite-rich rock treated water; CCO = Creede, Colo., clinoptilolite-rich rock treated water; LL = Leadville limestone treated water]

Date and sample	pH	Ca	Cu
		mg/L	
	June 1994 Raw water		
Faucett site			
6/2/94	6.3	7.5	0.1
6/3/94	6.4	6.7	0.1
6/4/94	6.2	6.6	0.1
6/5/94	6.1	6.6	0.1
6/6/94	6.1	6.7	0.1
Terrace site			
6/2/94	6.2	6.9	0.1
6/3/94	6.35	6.6	0.1
6/4/94	5.9	6.4	0.1
6/5/94	5.35	6.2	0.2
6/6/94	5.55	6.2	0.2
	Treated water		
Faucett site			
6/3/94-SDA1		5.1	0.1
6/3/94-SDA2		5.5	0.1
Terrace site			
6/4/94-CCO1		5.8	0.1
6/4/94-CCO2		6.1	0.1
	July 1994 Raw water		
Faucett site			
7/5/94	3.7	17.1	0.68
7/6/94	3.7	17.6	0.73
7/8/94	3.95	18.5	0.81
7/9/94	3.9	18.9	0.84
Terrace site			
7/6/94	3.85	17.4	0.77
7/7/94	3.9	17.5	0.80
7/8/94	3.9	17.4	0.82
7/9/94	3.85	18.6	0.90

Table 19.—Copper and calcium in raw and treated water at the Faucett and Terrace sites for June, July, and August 1994 tests—Continued

Date and sample	pH	Ca	Cu
		mg/L	
	Treated water		
Faucett site			
7/6/94-SDA		6.5	0.38
7/8/94-SDA		5.4	0.29
7/9/94-SDA		10.1	0.37
7/6/94-CCO		17.3	0.67
7/8/94-CCO		16.5	0.67
7/9/94-CCO		17.2	0.73
Terrace site			
7/7/94-SDA		15.4	0.67
7/8/94-SDA		13.5	0.56
7/9/94-SDA		11.3	0.17
7/7/94-CCO		17.8	0.81
7/8/94-CCO		17.2	0.76
7/9/94-CCO		16.3	0.68
	August 1994 Raw water		
Faucett site			
8/2/94	4.0	26.5	0.8
8/3/94	3.8	26.9	0.9
8/4/94	3.8	26.9	0.8
8/5/94	3.8	26.9	0.8
Terrace site			
8/2/94	3.85	27.2	0.9
8/3/94	3.75	27.6	0.9
8/4/94	3.9	28.0	0.7
8/5/94	3.9	27.5	0.8
	Treated water		
Faucett site			
8/3/94-SDA		15.9	0.4
8/4/94-SDA		20.7	0.6
8/5/94-SDA		22.7	0.5
8/3/94-LS		26.7	0.8
8/4/94-LS		26.3	0.8
8/5/94-LS		26.3	0.8
Terrace site			
8/2/94-SDA		20.3	0.1
8/3/94-SDA		19.5	0.3
8/4/94-SDA		21.4	0.4
8/5/94-SDA		20.7	0.5
8/2/94-LS		26.7	0.9
8/3/94-LS		27.2	0.9
8/4/94-LS		28.0	0.8
8/5/94-LS		28.3	0.8