AN "AUTOPSY" OF THE FAILED TANGASKOOTACK #1 VERTICAL FLOW POND, CLINTON CO., PENNSYLVANIA¹

Arthur W. Rose, David Bisko, Amidea Daniel, Mary Ann Bower and Scott Heckman²

<u>Abstract</u>. The Tangascootack #1 passive treatment system was constructed in 1998 to treat an acid discharge from an abandoned surface mine in Clinton Co., PA. The discharge averaged about 155 L/min with pH 4.0, acidity 235 mg/L CaCO₃, Fe 3.7 mg/L, Al 24 mg/L and Mn 68 mg/L. The vertical flow pond had dimensions of 48 x 20 m and was preceded by a wetland and followed by an oxidation/settling pond. The system produced net alkaline effluent for a few months, but by 1 year after construction, the effluent acidity was 120 mg/L, and flows decreased to 20-60 L/min. Plugging by Al precipitate was suspected, despite occasional flushing.

In 2003, investigation showed partial plugging by cattail roots in the effluent pipe, but after removal of the roots, head loss across the VFP was less than 2 cm at flows of about 20 L/min, indicating that plugging was not severe. The VFP was drained and the compost and limestone (2.5 cm dia..) were excavated along several trenches. Compost, originally designed as 30 cm thick, varied markedly from 8 to 25 cm thick. In thick zones, basal compost was coated by black Fe sulfides. The limestone was coated nearly everywhere for at least 25 cm beneath the compost with gelatinous white Al-bearing precipitate mixed with quartz-rich silt. A layer of gypsum beneath the Al precipitate covered the limestone in places. An open joint in the underdrain may have somewhat concentrated flow. The limestone layer retained considerable pore space and permeability. In spite of the flushing and open joint, Al coating was approximately the same over underdrain pipes as elsewhere. Also, the Al coating was present nearly everywhere, under both thick and thin compost, indicating that channeling was minimal. Measurements in 1999 and 2003 show that the system was removing acidity at reasonable rates of 45 to 60 g/m²/day.

Additional Key Words: Acid mine drainage, passive treatment, SAPS

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Introduction

In recent years, vertical flow ponds (VFP), also called SAPS (Kepler and McCleary, 1994), vertical flow systems, and reducing and alkalinity producing systems (RAPS), have become the dominant technology for passively treating acid mine drainage that is net acid. However, experience has shown that some of these units decrease in effectiveness or fail over periods of a few years (Rose, 2003; Ziemkiewicz et al., 2002). In order to minimize future problems of this sort, it is necessary to understand the reasons for failure, and to modify the design or maintenance to avoid problems.

The purpose of this paper is to present information from a VFP that declined markedly in performance within about a year after construction. Though it was still accomplishing some treatment, most of the acid remained in the effluent, and the flow decreased to a few liters/s. Funds were obtained from a Section 319 grant by the State of Pennsylvania for digging up the system and studying the internal characteristics to elucidate the reason or reasons for decreased performance.

Description of the Passive Treatment Facility

The Tangaskootack #1 VFP was constructed in 1998 to treat a discharge of very acid water from the downdip end of a reclaimed surface mine in Clinton County PA (Fig. 1). The site is about 13 km W of Lock Haven, PA at 41°8'37"N, 77°38'47" W.

Coals in the area are primarily Brookville and Lower Kittanning coals, accompanied by fireclay beneath the Brookville (Smith and Skema, 2001). Mining started in the 1800's, with extensive underground mining in the 1900 to 1930 period. Additional surface mining continued up to recent years. The Tangaskootack #1 site lies at the down-dip end of a reclaimed surface mine on the Brookville-Clarion coals. The discharge apparently originated with early mining, and was recognized as pre-existing in a permit granted about 1980. The discharge had not become worse on completion of this surface mining operation in about 1990, so the operator was not held responsible.



Figure 1. Map of discharges, ponds and vertical flow pond at Tangascootack 1 site. Elevations are in meters relative to arbitrary datum. Seep a El. 26.2 is the major discharge to the system. "Leakage" indicates location of major leakage when first visited in 2003.

For the period 7/98 to 8/99, the discharge averaged pH 4.0, acidity 235 mg/L CaCO₃, Fe 3.7 mg/L, Mn 67 mg/L, Al 24 mg/L and SO₄ 1695 mg/L at a flow rate of 155 L/min (Fig. 2). No samples were collected between 12/99 and 5/03.

Two pre-existing sedimentation ponds on the site were modified to a vertical flow pond and an oxidation/settling pond plus connecting ditches (Fig. 1). A cattail wetland preceded the VFP, and by 2003, extensive cattail growth occurred along the channels. The vertical flow pond was designed with water surface dimensions of 48 x 20 m and interior slopes of 1V:2H. A 0.60 m layer of No. 57 limestone (~2.5 cm) was to be overlain by 30 cm of mushroom compost and 1.8 m of water. Six perforated PVC underdrain pipes, 10 cm in diam., were designed to run the length of the pond in the limestone (Fig. 3). Perforations were about 1 cm in diameter at an interval of about 1.5 m. The underdrain pipes were connected by 10 cm diam. header pipes at both ends, and the NE end connected to an upward-sloping 15 cm PVC pipe as an outflow. In addition, at the SW end, a 15 cm PVC draining/flushing line and valve were installed, leading to lower ground near Tangascootack Creek.



Figure 2. Acidity and alkalinity of influent and effluent vs. time. No samples from 1/00 to 5/03.



The acidity-alkalinity of the inflow and outflow are plotted on Figure 2 (Table 1). For

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Table 1. Dat	a on cł	nemistry	and flow	v of Tang	ascoo	tack 1											
Date	Flow I /min	pH (fld)	pH (lab)	Conduct.	Alk. ma/l	Acidity ma/l	Fe ma/l	Mn ma/l	Al ma/l	SO4 ma/l	Ca mg/l	Mg ma/l	K ma/l	Na mg/l	Si ma/l	TSS ma/l	TDS
Inflow				u <i>u</i> , o	<u>9</u> / =	<u>9</u> , _	<u>9</u> , _	<u>g</u> / _	g/ =		<u>g</u> / _		g/ =	<u>9</u> /=	<u>9</u> /=	<u>g</u> / _	g/ =
7/16/1998	152	4.5	3.3	3180	0	334	3.73	80.5	27.8	1500						1	3744
8/5/1998	122	4.0	3.3	3210	0	256	3.21	77.5	27.4	1900						20	4410
9/16/1998	148	4.0	3.3	3150	0	358	3 51	69.2	25	2169						10	3920
10/19/1998	122	4.0	3.4	3000	0	218	2.79	69.3	25.2	1940						1	3154
11/19/1998	80	4.0	3.4	2950	Ő	198	3.28	70.5	25.4	1900						1	3250
12/9/1998	148	4.5	3.3	2930	0	186	3.82	68.8	23.5	1830						14	3058
1/28/1999		4.5	3.5	2560	0	202	7.16	58.9	18.9	1300						34	2568
3/2/1999	80	4.5	3.5	2150	0	142	1.99	43.8	11.6	1210						6	2152
8/4/1999	57	4.5	3.3	3150	0	218	4.01	69.2	27.9	1510						68	3798
5/27/2003	23	4.3		2250	0	175	<0.1	43	13	1593	184	225	6.1	4.9	9.4	Ļ	
7/29/2003	23	3.4		2050	0	204	1.8	40	12	1422	156	197	5.6	4.4	9.5	5	
8/17/2003	232	3.3			0	282	1	50	22	1668	189	237	6.8	5.3	13	3	
8/20/2003	232	4.0	3.6	1100	0	284	1.38	41.7	15.7	1247	160	140				12	
10/30/2003	163	3.2	3.6		0	269	0.98	43.8	18.6	1193						20	
Average	121.6	4.1	3.4	2640.0	0.0	237.6	3.0	59.0	21.0	1598.7	172.3	199.8	6.2	4.9	10.6	5 17.0	3339.3
Outflow from	n final p	pond															
7/16/1998	152	6.5	6.7	3110	58	0	2.9	71.4	0.61	1800						1	3912
8/5/1998	122	6.5	6.6	3210	62	0	1.56	75.3	0.44	2100						22	4330
9/16/1998	148	6.0	6.5	3130	62	0	0.9	64.1	0.3	2269						16	4258
10/19/1998	122		6.5	2920	60	0	0.7	63.5	0.29	1830						6	3395
11/19/1998	80	6.0	6.4	2930	62	44	0.75	72.1	0.4	2010						1	3197
12/9/1998	148	6.5	6.4	2890	56	34	0.38	66.8	0.39	2220						4	3468
1/28/1999		6.0	6.0	2200	44	36	0.67	45.7	3.84	1090						44	2146
3/2/1999	80	6.5	5.7	2130	19	66	0.32	44.2	2.03	1320						20	2124
3/18/1999	285	5.0	4.8	1690	11	104	0.779	40.1	4.86	1004							
3/24/1999	228	5.5	4.7	1719	11	74	0.298	36.5	5.55	1110						1	1392
4/12/1999	350	5.5	5.0	1847	11	46	0.412	37.2	2.84	964						1	1950
4/22/1999	171	5.0	4.8	1975	11	106	0.308	39.7	3.44	1200						26	1952
6/3/1999		5.0	4.7	2500	12	118	0.658	55.5	4.65	1869						22	3186
7/1/1999	76	5.0	4.6	2740	11	126	1.27	62	3.99	899						1	3178
8/4/1999	57	5.0	4.6	3000	11	124	0.98	62.5	2.56	1590						4	3510
8/31/1999	15	5.0	4.7	3030	11	114	0.958	74.2	3.38	1920						36	3628
10/6/1999	42	5.0	4.8	2820	11	118	0.301	73.5	2.66	1930						34	2966
12/9/1999	15	5.2	4.8	2640	13	130	0.605	62.8	4.54	1710						1	3104
5/27/2002	00	64		2170	10	60	0.01	10	<u>م</u>	1991	216	201	5 6	10	5 5		
3/21/2003	20	6.4		2170	40	20	0.91	40	0	1001	210	221	5.0	4.0	0.0) -	
8/17/2002	23	0.3 50		2000	20	3U 1/E	0.0	40	11	1614	344 225	020 000	0	5.7 5.2	0.0	,	
0/11/2003 8/20/2002	232	5.2	55	000	20	140	ו קפיט	49 ⊿∩ 7	6 05 6 05	1014	220	220	0.0	5.5		11	
0/20/2003	202	6.0	0.0 5.2	000	∠ I 17 0	100	0.07	40.7	0.00	1//6	190	139				44 29	
10/30/2003	100	5.0	0.3 17	900.0	17.0	13.0	0.7	41.7	0.0	1440						20 29	
Average	121 0	5.0	4.1 5 4	2/102 2	20 0	70 /	1 0	43.0	12.4	1592	245	207	E	E	0	20	20/11
Filtered sam	101.2	5.7	5.4	2400.Z	23.9	13.4	1.0	- 55	5.0	1000	240	221	0	5	c c	, 17	3041
8/17/2003F	232	5.2					1	49	5	1662	229	231	6.6	5.1	10)	

about 3 months (7/98-10/98), the system generated alkaline effluent, removing all of the 300 mg/L acidity. Then acidity appeared in the effluent, and after 4/99, leveled off at about 120 mg/L acidity, compared with about 200 mg/L influent acidity. This behavior continued until at least 12/99, but flow dropped off markedly in summer and fall 1999. This decrease in flow may be partly due to the very dry spring in 1999. Also, the flow was measured at the outlet of the settling pond, and it is possible that the flow from the discharge was larger but was leaking out from the wetland area. The pattern of increased effluent acidity and decreased flow led to the conclusion that the system was becoming plugged, probably with Al hydroxide because of the

elevated Al in the influent. The system was flushed occasionally in the first year, perhaps 6-10 times.

Observations at the Passive Treatment System

In 2003 a grant was obtained from the State of Pennsylvania to investigate the cause of the degraded behavior. Starting in March 2003, the site was visited and various observations and measurements made.

On a visit in March 2003, the system was partly frozen and flow out of the system was negligible, at most a few liters/min. Most discharge water was leaking from the inflow channel directly to the settling pond. A general lack of relief in the system was noted – only about 1 m difference in elevation from the inflow channel to the final settling pond. In addition, the channel was choked with cattails, impeding flow. When the flushing valve was opened, a large flow was observed, with a gray color. The high flow rate suggested that plugging was not severe.

On 5/1/03, examination of the VFP outflow point showed that a large rock had rolled down across the outflow pipe. After removal of this rock, the outflow pipe was found to be largely blocked by cattail roots. Removal of these roots markedly increased the flow, perhaps up to 20 L/min. Leveling showed that the head difference between the VFP and the outflow was approximately 10 cm.

On 5/27/03, flow from the system was about 20 L/min based on a weir, and the head loss from the VFP to the outflow was only 1+/-0.5 cm, based on more accurate surveying. On opening the flushing/draining valve, outflow water was observed to be orangish for about 1 minute, and gray for at least 10 minutes. The pond was largely drained in a few hours, and draining was completed the following morning.

A series of 5 effluent samples was collected on 5/27/03 during the first 15 minutes of draining the pond. Based on suspended Al in these samples and estimated flow rates, the flush released about 300 g of Al. The system had been previously flushed on 3/19, 68 days earlier. At a flow rate of 3 l/min, about 25,000 g of Al had been accumulated in the VFP during this time. The flush released only about 1% of the accumulation. The inflow and outflow chemistry and the flow rate have been sampled several times starting in May 2003 (Table 1). The unit is removing over 100 mg/L acidity at average flow rates of 240 L/min., and is generating some

alkalinity. Similar acidity removal was observed in the period 3/99 to 12/99. Given the surface area at the water surface of 48 x 20 m, these removal rates correspond to 45 to 60 g/m²/d. This rate is higher than the average rate of 40 g/m²/d found by Rose and Dietz (2002) for a group of 30 vertical flow systems.

Excavation of the Compost and Limestone and Analysis of Samples

On 5/28/03, a set of 12 compost cores was taken on a grid of 4.5 x 10 m (Fig. 3). The cores were collected by pushing a 6.3 cm dia. transparent plastic pipe with a sharpened bottom edge into the compost, removing the pipe and ejecting the compost to a plastic bag. The bags were frozen within a few hours. The thickness of the compost was recorded. Cores for microbial study were also taken by Dr. J. Calabrese of Lock Haven University.

Frozen samples of compost were split approximately in half and air-dried. A 2 g sample of dried compost was then leached with 20 ml of hot 1:1 HCl to dissolve any Al or Fe precipitate. The leachate was filtered and brought up to 50 ml with deionized water for analysis by ICP-ES. Results are listed in Table 2.

Three samples of the basal black compost were leached with water to remove pore fluid, dried at 50°C, ground and analyzed for total S. Results were sample 8A (black), 2.02% S;

Table 2.	Leaches	of compo	ost core	s									
Core	Wt(wet)	Wt(dry)	Water	AI	Ca	Fe	K	Mg	Mn	Na	Si	SO4	Sum
	g	g	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%
1a	102	51.8	49	1.45	1.77	0.30	0.045	0.64	0.04	0.01	0.09	1.58	5.92
2a	197.5	71	64	1.33	0.28	0.24	0.073	0.17	0.04	0.01	0.01	1.34	3.47
3a	110.3	63.8	42	1.62	3.99	0.38	0.06	0.21	0.06	0.01	0.05	1.58	7.95
4a	200	132	34	1.72	0.76	1.65	0.088	0.31	0.12	0.01	0.02	1.34	6.01
5a	136.5	57.4	58	1.90	0.49	0.44	0.093	0.23	0.07	0.01	0.01	2.00	5.24
6a	282	95.1	66	0.91	0.56	0.35	0.043	0.22	0.06	0.01	0.00	1.37	3.53
7a	132.8	31.7	76	1.20	0.28	0.42	0.07	0.27	0.05	0.01	0.01	1.16	3.45
8a													
9a	164.5	94.4	43	1.35	0.30	0.43	0.05	0.14	0.06	0.01	0.01	1.40	3.74
10a	123.5	33.3	73	0.86	0.27	0.47	0.14	0.29	0.06	0.01	0.01	1.03	3.13
11a	115	48	58	2.91	2.45	0.83	0.168	0.72	0.05	0.01	0.02	2.54	9.68
12a	109	49	55	2.15	1.28	0.51	0.12	0.36	0.05	0.01	0.02	2.07	6.56
Average			56	1.58	1.13	0.55	0.09	0.32	0.06	0.01	0.02	1.58	5.33

Values are % of dry compost.

A 2 g sample of dry compost was leached with 20 ml hot 1:1 HCl for about 5 min,

then filtered and analyzed by ICP.

Sample 1B, 1.74% S; and sample 6B, 1.79%S. A sample of brown compost overlying 8A contained 1.49% S, suggesting that only part of the S is sulfide.

On 8/5/03, the pond was redrained, and three trenches were cut through the compost and limestone with a large backhoe supplemented with hand shoveling near underdrain pipes. The compost, limestone and underdrain pipes were examined, sampled and photographed. Limestone was coated with a gelatinous white material for 15 to 30 cm beneath the compost at most locations (Fig. 4 and 5). Limestone deeper than 30 cm was largely free of the white precipitate. In the zones of thicker compost (>15 cm), a firm black layer was observed at the base of the compost. Beneath this black material, the white precipitate appeared somewhat less abundant than elsewhere. Only traces of brown or red precipitate were observed.

The thickness of limestone above the underdrain pipes varied considerably, from 13 cm to 55cm, though at most locations the total thickness of limestone was approximately 60 cm.

Excavation of the upward-sloping outflow pipe disclosed that one pipe joint was open



Figure 4. Well developed white coating on upper part of limestone bed, and black layer at base of compost. Area at left is obscured by loose limestone. Spilled compost covers limestone in a spot near right, and spilled limestone covers compost in center.



Figure 5. Close up view of white coated limestone. Note relatively open pores. Bright white is gypsum coating, and gray is Al coating. Red at bottom is a pocket knife.

about 1 cm, allowing additional flow of water into the underdrain system. Also, underdrain pipes were lacking in the NW corner of the bed, and apparently were connected farther south.

Four samples of limestone (2B, 3B, 4B and 10B) with obvious white coating were agitated with DI water, which washed off the coating. After removal of limestone fragments, the solution was evaporated to dryness. The residue was then acidified to pH 2 with HCl to dissolve Al and Fe precipitates and other soluble materials. Much of the material washed from the limestone was found to be insoluble quartz sand and silt that were apparently trapped in the coating. Analyses of the soluble portion are listed in Table 3. Al is a major component. X-ray diffraction patterns of the dried coating scraped from limestone showed quartz, gypsum and possible calcite, but no Al phase (Fig. 6). The insoluble quartz silt may have come from the overlying compost and been carried into the accumulating Al precipitate.

Examination of the washed limestone fragments showed a bright white coating on some

Table 3. Analyses A. Mass dissolved	s of coatii (mg)	ngs wasł	ned or di	ssolved	from lime	stone
	2B	3B	4B	7B	10B	
AI2O3	140.63	5.1	1.85	37.7	309	
CaO	64.79	153.4	253.1	547	328	
Fe2O3	4.82	0.14	0.03	1.8	140	
K2O	0.23	1.59	0.61	7.23	4.5	
MgO	8.22	9.43	16.4	41.4	32	
MnO	1.04	0.13	0.19	6.1	6	
Na2O	0.45	0.31	0.43	2.5	3.2	
SiO2	3.68			14.5	37	
SrO	0.11			0.9	0.2	
SO4	22.38	1.5	1.88	21.8	41	
B. Wt % of oxides	2B	3B	4B	7B	10B	
AI2O3	58.47	58	37.1	40.70	38.49	
CaO	19.96				30.29	
Fe2O3	1.51	1.6	0.6	1.90	13.19	
K2O	0.06	18.1	12.2	7.80	0.36	
MgO	3.01				3.52	
MnO	0.30	1.4	3.8	6.50	0.51	
Na2O	0.13	3.5	8.6	2.70	0.28	
SiO2	1.74			15.60	5.23	
SrO	0.03			1.00	0.02	
SO4	14.78	17.1	37.6	23.50	8.11	
Basis*	1	2	2	2	1	

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* 1 = Wt % all analyzed oxides, 2 = wt % oxides except CaO, MgO. Analyses 2B, 7B, 10B by Materials Characterization Lab, Penn State Univ. Analyses 3B, 4B by Mahaffey Labs.



Figure 6. X-ray diffraction pattern of white precipitate coating limestone of sample 6a. Q=quartz, G=gypsum, C=calcite

surfaces. X-ray diffraction analysis of this white coating scraped from the limestone surface showed that it is gypsum (CaSO₄· $2H_2O$).

Discussion

The initial high rate of acidity removal is typical of many vertical flow ponds. The high rate apparently represents initial exchange of acid solutes (Fe, Al, H) onto the compost, and reaction with fine limestone powder in the limestone bed or compost. For the later period 3/18/99 to 4/22/99 the average removal rate was about 60 g/m²/d, and for 8/03, the removal rate is 46 g/m²/d, using the water surface area of 960 m². Use of the bed surface area of about 510 m² would give much higher removal rates. This limited data indicates that the system is relatively effective in removing acidity compared to a typical value of about 40 g/m²/d for other systems.

Several lines of evidence indicate that the system was not significantly plugged. The very small head loss of about 1-2 cm at a flow rate of 20 L/min shows high permeability, as does the high flow rate estimated at 2500 L/min from the drain valve. In addition, observation of the limestone bed indicates a high degree of connected pore space, despite some accumulation of coatings. The decreased flow in 1999 and early 2003 is due to some combination of decreased flow from the discharge in the dry summer of 1999, leakage from the channels and wetland, and plugging of the outflow pipe by cattail roots. Low flow rates are promoted by the limited elevation drop and extensive cattail growth along the channels.

Weights of the wet compost and the air-dried compost indicate that about 50% water is present in the compost. From this, a retention time for compost can be estimated. If all the water is assumed to flow, then at 20 L/min across 500 m² of bed that is 20 cm thick, the retention time in the compost is about 1.8 days. If the active pore space is only 25%, the retention time in compost is a little less than a day. At a near maximum flow rate of 230 L/min, the retention time in compost is about 4 hours.

The compost layer varied considerably in thickness, from <10 cm to 25 cm, and was distinctly thinner than the design specification of 30 cm. However, for this low-Fe discharge, the compost was apparently thick enough to reduce ferric Fe and dissolved O_2 , because very little Fe hydroxide precipitate was observed on the limestone. In the areas of compost thicker than about 15 cm, a distinct black layer in the base of the compost contains Fe sulfide at levels of 1 to 2%. Apparently the O_2 and ferric Fe of the influent water were consumed by the top 15 cm of

compost, so that sulfate-reducing bacteria were active in the base of the compost. The redox state is apparently reduced enough for sulfate reduction in retention times of a few hours.

The coating on limestone is composed of a mixture of an amorphous Al species, silt and clay, and gypsum. The Al material and the silt-clay are easily washed off the limestone fragments. At least some of the gypsum is cemented firmly to the limestone.

Table 4 lists saturation indices for various phases in the influent and effluent water. The waters are not saturated with gypsum, though they are within about 30% of saturation. Therefore, the observed gypsum is hypothesized to precipitate because of elevated Ca concentrations beneath the gelatinous Al coating adjacent to the limestone. The elevated Ca results from dissolution of CaCO₃ to generate a diffusion gradient of Ca (and HCO₃) from the calcite surface to the open solution (Fig. 7). Gypsum has also been observed coating limestone

Mineral Sa	aturation index, inflow ¹	Saturation index, outflow ²
$Al(OH)_3$ (am.)	-5.74	-0.75
Alunite (KAl ₃ (SO ₄) ₂ (OH) ₆	-1.35	7.94
Fe(OH) ₃ (am.)	-1.89	1.34
Gibbsite (Al(OH) ₃)	-3.00	1.99
Goethite (FeOOH)	3.82	7.05
Gypsum (CaSO ₄ ⁻ 2H ₂ O)	-0.5	-0.41
Jarosite-K (KFe ₃ (SO ₄) ₂ (Ol	H) ₆ -0.46	3.53
Jurbanite (AlSO ₄ OH)		0.96
Kaolinite $(Al_2Si_2O_5(OH)_4)$	-4.68	5.43
Pyrolusite (MnO ₂)	-8.49	-0.90
Quartz (SiO_2)	0.26	0.32



¹Inflow to VFS 8/17/03

Figure 7. Schematic diagram showing limestone fragment with $Al(OH)_3$ coating underlain by gypsum, and inferred concentration gradient in Ca (and HCO_3) and SO₄ leading to saturation with gypsum. The concentration gradients arise because of diffusion through the $Al(OH)_3$ layer. beneath an organic coating at the Filson 1 vertical flow pond (Rose and Morrow, manuscript) and by Hammerstrom et al. (2003). Thomas and Romanek (2002) also report gypsum replacing and coating limestone in experiments with a limestone-compost mixture treating highly acidic water. Given the gelatinous and water-rich nature of the Al coating at Tangascootack, it is possible that the denser gypsum coating is more effective in inhibiting calcite dissolution than the Al material.

The effluent water is supersaturated with gibbsite $(Al(OH)_3)$ but not with amorphous $Al(OH)_3$. It is possible that the Al phase is related to jurbanite $(AlSO_4OH)$, or (given the low S in the coating) is a phase intermediate between gibbsite and amorphous $Al(OH)_3$ in solubility. Several clay minerals (kaolinite, K-mica) are also supersaturated, as is alunite $(KAl_3(SO_4)_2(OH)_6)$.

A crude estimate of the amount of Al coating can be found from the inflow and outflow concentrations and the flow rates. Taking an average influent Al of 21 mg/L, an average effluent Al of 3 mg/L, and an average flow rate of 120 L/min over 5 years, the mass of Al accumulated in the VFP is about 5000 kg. Compared to the mass of coated limestone (30 cm thick), approximately 200 x 10^3 kg, the Al amounts to 3% of the mass of the limestone bed. For fragments approximately 2 cm in size, the coating is estimated to be about 0.1 mm thick.

At this rate, actual plugging of the bed would take a long time, but the coating undoubtedly inhibits chemical reaction. However, since the Al phase is amorphous and undoubtedly has a high water content and a relatively high diffusion rate, this effect may be subordinate to the precipitation of gypsum next to the limestone. In agreement with the limited effect of the Al precipitate on reaction rate, the bed seems to be removing acidity at a good rate compared to other VFP's. Eventually, however, the effects of coating and precipitate buildup will have a negative effect on treatment.

The limited experiments on Al removal during flushing are in agreement with other tests of this sort (Watzlaf et al., 2003) that show less than 2% removal of Al by flushing. The flushing may remove material from the immediate vicinity of the perforations in the underdrain, but both this data and our observations of the excavated limestone near the underdrain indicate only limited effectiveness of flushing.

The Al precipitate was present at the top in most areas of the limestone bed, both in areas with 20 cm of compost and in areas with almost no compost. This indicates that downward flow

is occurring in nearly all parts of the bed, with little channeling. The Al coating was less obvious in occasional small spots, and appeared thinner beneath some black compost, suggesting local flow variability due to low-permeability spots in the compost.

Based on an expected acidity removal rate of 40 $g/m^2/d$, the VFP is too small to treat the influent load at typical flow rates of 100 to 230 L/min. Addition of one or two additional VFP's is planned for the site. In addition, to handle the problem of Al precipitation, the addition of fine limestone to the compost layer is proposed. Compost layers containing limestone have much higher rates of acidity removal and alkalinity generation (Rose and Dietz, 2002). The ferric Fe and Al precipitate within the compost layer in these systems (Thomas and Romanek, 2002).

Conclusions

The major decline in performance of the Tanga VFP was caused by several factors:

- 1. Normal decline observed in most VFP's during the first few months.
- 2. Plugging of the outflow pipe with roots and a rock.
- 3. Dry weather that decreased the inflow, combined with leakage from the wetland and inflow channel.
- 4. Coating of the limestone with amorphous Al precipitate and gypsum.
- 5. Extensive growth of cattails in the inflow and outflow channels combined with very low gradients in these channels.

Flow decrease caused by the Al precipitates was minor, but armoring of the limestone by Al precipitate and gypsum will potentially decrease the neutralizing ability of the limestone bed.

The white Al-bearing coating was present relatively uniformly at the top of the limestone layer, suggesting that channelized flow was not important. However, thicker compost had a layer of black sulfide-bearing compost at its base, and less Al precipitate was evident at these sites. Possible explanations for this observation are that the sulfide-bearing compost is less permeable, or that the development of the sulfide precipitate somehow affects the Al or gypsum precipitation.

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