THE USE OF WETLANDS TO REMOVE NICKEL FROM MINE DRAINAGE - IS PERPETUAL TREATMENT REALLY POSSIBLE?[1]

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Abstract. Although wetland treatment systems have been shown to be effective for treating both coal and metal mine drainage, the longevity of the treatment has always been a question. Data collected from a wetland in northeastern Minnesota suggests it may be possible to build a wetland that will provide long term treatment.

A 7000 square meter overland flow wetland was built in 1992 to treat a mine drainage with an av798798erage pH of 7.2 and an average nickel concentration of 5.1 mg/L. Nickel removal exceeded 90% for the first three years of operation. In 1995, the stockpile which contributed the major input to the wetland was capped, and both flow and concentrations in the drainage were reduced. An intensive study was conducted on one section of the wetland where a large percentage of the overall removal was occurring. Nickel concentrations in the substrate reached 1.5% by weight and the calculated nickel mass in the substrate was about the same as the overall mass removal calculated from the water quality and flow data.

Based on a model of substrate accumulation in wetlands, the wetland generates 7 kg of nickel removal capacity each year. Since the annual input of nickel has been reduced to around 10 kg, the projected lifetime of the wetland is about 300 years.

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Introduction

Wetlands have been used to treat a variety of water quality problems, including agricultural, municipal and industrial discharges (Hammer, 1989, Moshiri, 1993). Wetlands have also been successful in treating coal and metal mine drainage and can be an attractive alternative to more

conventional treatment methods (Hedin et al., 1994, Eger et al., 1996, Sobolewski, 1997). Wetlands can be less costly to build, use processes which naturally occur to remove metals from the water (e.g.
adsorption, filtration), and offer a system that ideally should operate with little to no maintenance for extended periods of time. Since mine drainage problems can persist for hundreds of years, the longevity of any system is a critical issue. The purpose of this study was to examine metal removal in a wetland and to estimate how long treatment might continue.

Site Description

The Dunka Mine was a large open pit taconite operation which operated from 1962 to 1995. At this location the Duluth Complex, a metalliferous gabbroic intrusion, overlaid the taconite ore and was removed and stockpiled along the east side of the open pit. The Duluth Complex material contains copper, nickel, and iron sulfides, and the stockpiles contained over 50 million metric tons of waste rock and covered about 120 hectares. Seeps appeared at the base of the stockpiles, and flow generally occurred from early April to late November. Average flows from the various seeps ranged from 30 L/min to 840 L/min, but flows exceeding 6000 L/min were observed after periods of heavy precipitation.

Nickel was the major trace metal in the drainage, and annual median concentrations prior to closure were on the order of 3-30 mg/L. Copper, cobalt, and zinc were also present but were generally less than 5% of the nickel values. Median pH ranged from 5.0 to 7.5, but most of the stockpile drainage had a pH greater than 6.5.

Wetlands were located near every stockpile and appeared to offer potential treatment areas for each seep (Eger and Lapakko, 1989). These wetlands were typical of the many small lowland areas in northern Minnesota, and would generally be associated with any mining operation in the area.

In the mid-1980s, LTV Steel Mining Company began an extensive program to evaluate various options for mitigating the problems at this mine. The company's preferred option was a combination of passive alternatives which would reduce flow emanating from the stockpiles, and use wetland treatment to remove metals from the resulting drainage. In 1988, four overland flow test cells were built to investigate methods to optimize metal removal and to provide design data

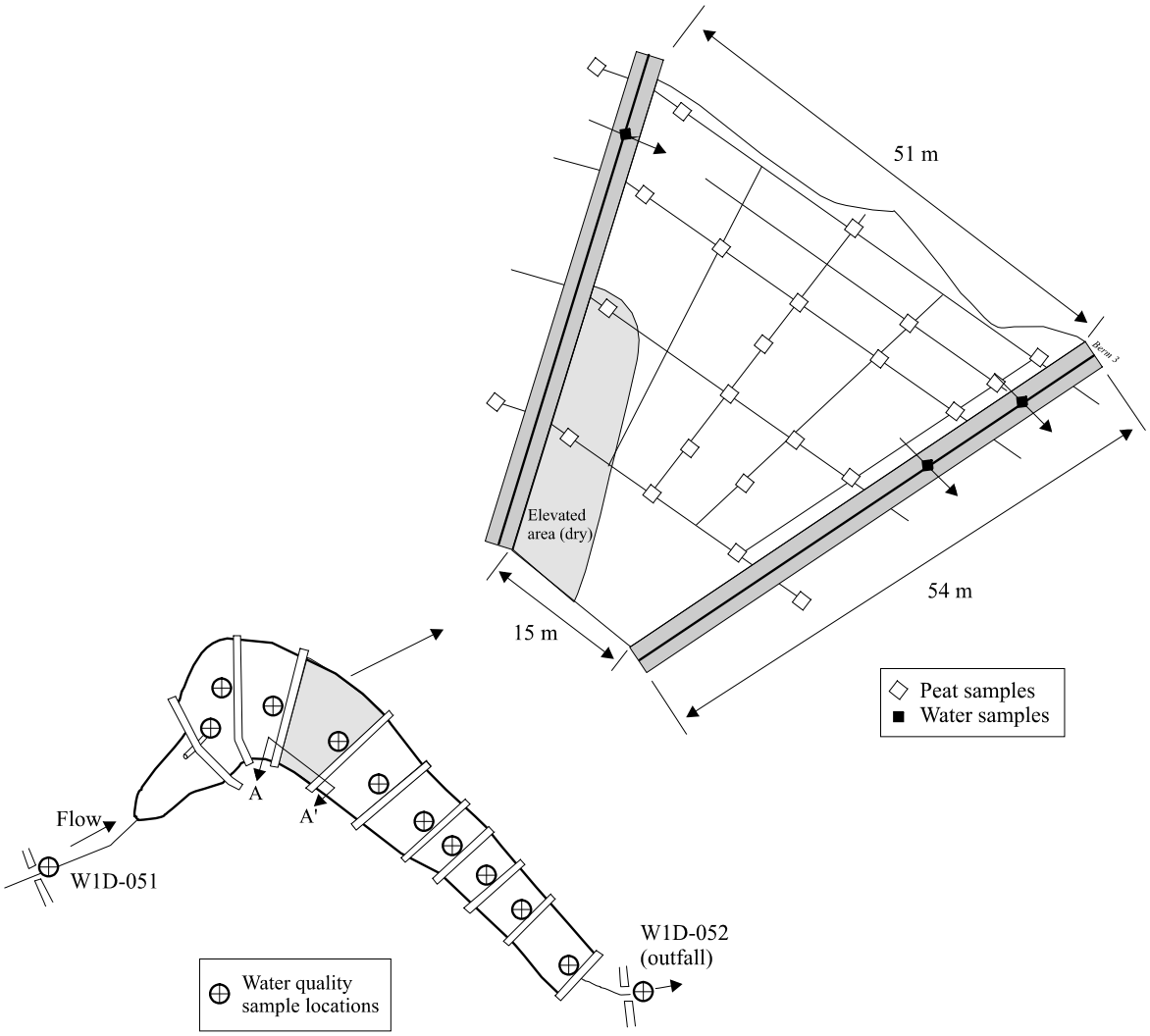
for the ultimate implementation of wetland treatment at this facility (Eger and Lapakko, 1989; Eger et al.,1991, 1993, 1994). Based on the results of this study, two full-scale wetland treatment systems were built in March of 1992 (Eger et al., 1996). By the time the Dunka mine was closed in 1995, the company had already begun to implement a closure plan. The amount of water flowing through the stockpiles was reduced by routing surface and groundwater away from the piles. Infiltration into the waste rock stockpiles was reduced by covering the top portions of the stockpiles and any residual drainage was treated in constructed wetlands (Eger et al., 2000). This study focused on the W1D wetland treatment system, which was constructed in 1992 (Figure 1).

Overland Flow Wetland, W1D

This system was designed by STS Consultants, Ltd., and built by LTV Steel Mining Company in an existing wetland in 1992 (Frostman, 1992). The wetland was originally a combination of emergent (wet meadow) and scrub-shrub type wetlands, and the majority of the woody vegetation, which consisted primarily of alder (*alnus sp.*), was removed from the site. The basic design for the system included the construction of a series of soil berms, which were built to control water levels and to maximize contact between the drainage and the substrate (Figure 1). Soil berms were built with glacial till (sandy silt) available from a surface overburden stockpile on the property. After the berms were constructed, a 30 cm layer of a mixture of local peat and peat screenings was applied to the entire area except the top of the berm. The screenings were a waste material generated during the processing of horticultural peat and consisted mostly of wood fragments and long peat fibers. This material was selected to increase the permeability of the peat to at least 10^{-3} cm/sec and to provide available organic carbon. In the spring of 1992, the berms were hand-seeded with Japanese Millet, while the open water areas were seeded with cattails. To obtain the cattail seeds, cattail heads were placed in a container of water with a small amount of liquid soap and several large metal bolts. The mixture was agitated until the heads broke and the seeds were dispersed. The slurry was then broadcast by hand over the wetland. The majority of the flow to this system originated from the base
of the waste rock stockpile 8018, although additional seepage from another waste rock stockpile also drained to this area.

Study Cell, W1D

Based on the 1992 to 1994 water quality data collected from within the wetland, an individual cell within the original W1D system was selected for intensive study (Figure 1). The study cell was


51 m
Berm 3
Elevated
area (dry)
54 m
15 m
Peat samples
Water samples
A
A'
Flow
W1D-051
W1D-052
(outfall)
Water quality
sample locations

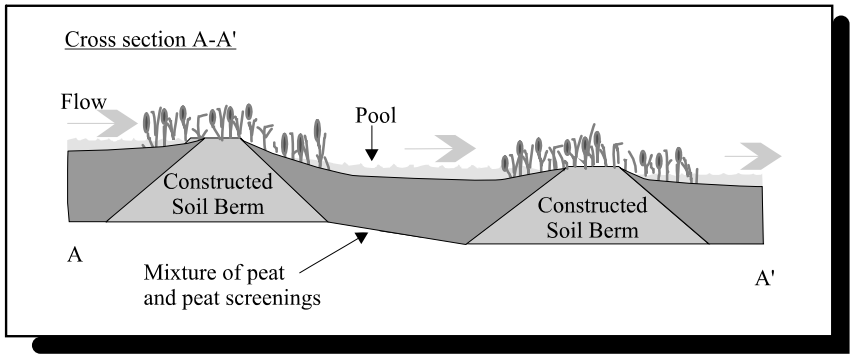

Cross section A-A'
Flow
Pool
Constructed
Soil Berm
Constructed
Soil Berm
A
A'
Mixture of peat
and peat screenings

Figure 1. Schematic of the W1D wetland treatment system study cell.

chosen because it had the largest drop in nickel concentration (26%). Essentially all of the flow

entered the cell at one point and left at two points (Figure 1). Water depth ranged from near 0 at the berm to about 20 cm in the center of the cell.

The total cell area was about 1500 m^2, but about 185 m^2 in the northeast corner was higher than the rest of the cell and was dry for most of the year. Vegetation in the treatment system consisted primarily of cattails (*Typha sp.*). Vegetation was dense around the edges of the study cell, and decreased toward the center. The sparsely vegetated area in the center contained open water, and occupied about 25% of the cell.

Methods

W1D

Water samples of the inflow and outflow of the overall wetland treatment system were collected twice a month by LTV personnel. From 1992 to 1994, LTV also collected samples at each berm in the wetland (Figure 1), and from 1997-1999 input and output samples to the study cell were collected by the DNR. Samples were analyzed for pH, specific conductance, copper, nickel, calcium, magnesium, and sulfate.

Specific conductance and pH were measured in the DNR laboratory with an Orion SA 720 pH meter equipped with a Ross combination pH electrode (model 8165), and a Myron L model (model EP) conductivity meter. Sulfate was analyzed at the Minnesota Department of Agriculture (MDA) laboratory. Prior to October 11, 1998, sulfate was analyzed with the Ion Chromatographic Method (Wastewater Method 4500-SO_4 B) with a Dionex DX300 IC. Subsequently, these parameters were measured with a Lachat QuickChem 8000 using the same methods. Metals samples were analyzed at MDA using a Varian 400 SPECTRAA atomic absorption spectrophotometer in the flame mode or a Zeeman GFAA graphite furnace. This was replaced by ICP/MS (Hewlett Packard HP4500 Series, model #G1820A) on August 22, 1999.

Continuous measurements of inflow and outflow water levels were made with a Steven's Model F recorder, and the flow was calculated from the standard equation for a 60 degree V-notch weir. Due to potential problems with the recording equipment under freezing conditions, continuous flow estimates were generally only available from May through October.

Peat samples were collected in April of 1996 and 1997 (Figure 1). If the surface of the peat was

frozen, samples were collected with a specially designed core tube sampler. A cylindrical cutting head (9.5 cm ID) was fabricated and mounted on the shaft of a power soil auger. With this sampler, samples could be collected while the substrate was frozen, which made identifying and separating layers much easier than with a standard coring device. Deeper samples were collected with a standard Macauley peat sampler.

Samples were generally divided into 10 cm sections: 0-10, 10-20, and 20-30 cm. Samples were dried at 105^0 C for 24 hours. The samples were then processed in a blender, sieved to -80 mesh, and totally digested with a mixture of 5 ml water, 10 ml concentrated nitric acid and 2 ml concentrated hydrochloric acid. The samples were sequentially microwaved for 10 minutes each at 40, 80, 120 and 160 psi. The digested samples were analyzed for trace metals by the MDA laboratory.

Results

Flow

Input flow was measured at the V-notch weir located about 60 meters from the toe of the 8018 stockpile and about 60 meters upstream of the beginning of the wetland (site W1D-051; Figure 1). Additional watershed area contributed flow to the wetland treatment system, but the majority of the nickel load originated at site W1D (Figure 1). Since continuous flow measurements were only available for May through October, the average daily flow calculated over this period has been used to compare the change in flow over time.

Average input flows ranged from 110-136 L/min for 1992 to 1994. In 1995 the top of the stockpile which provided the majority of the flow to the wetland was capped with a 30 mil LLDPE liner. Flows decreased in 1995 and average flows for 1996 to 1999 ranged from 30-83 L/min. Flow in 1999 was the highest of the post-closure flows, the result of a 17.5 cm rainfall in July. Peak flow after this event was on the order of 3400 L/min and the estimated daily flow was 1360 L/min. Annual precipitation for 1999 was 89.3 cm, substantially above the long-term average precipitation of 72.4 cm (Table 1).

Output flow was generally greater than input flow except during hot dry periods when evapotranspiration losses were large. During the summer of 1998 the output flow was 19-23 L/min

less than the input. In July, when the input flow decreased to 15-19 L/min, there was no flow at the outlet. Flow into and out of the study cell was not measured directly. Since there was little contributing watershed between the beginning and end of the wetland, output flow from the entire system (site W1D-052; Figure 1) was used to represent flow through the cell.

Water Quality

There was little variation in pH in the wetland (Figure 2). Both input and output pH generally ranged between 6.7 to 7.6. There was also little difference in pH between the input and output of the study cell; the average value for both sites was around 7.2 (Table 2).

From 1992 to 1994, the input nickel concentration to the wetland treatment system was typically on the order of 1 mg/L in the spring, then increased to approximately 6 mg/L in early summer. Concentrations then remained relatively constant until the seep froze in late fall (Figure 2). The average input nickel concentration to the study cell during this period was 2.9 mg/L. As water flowed through the cell, nickel concentrations decreased by about 26%, to about 1.8 mg/L.

In 1995, input nickel concentrations decreased substantially. The nickel concentrations in the input to the wetland remained low in the spring, but only increased to 2-3 mg/L in the summer. Maximum concentrations gradually decreased to less than 1 mg/L by 1999. As a result of the decrease in the input nickel concentration, the nickel concentration entering the study cell also decreased. Average input concentrations to the cell dropped to 0.32 mg/L for the period 1997 to 1999. Concentrations in the outflow were lower than the inflow and averaged 0.26 mg/L for 1997 to 1999 (Table 2).

Table 1. Flow and precipitation data W1D treatment system, 1992-1999.

Year	Inflow - Average Daily Flow (L/min)		Outflow - Average Daily Flow (gal/min)		Precipitation (cm)	
	May-Oct	Annual	May-Oct	Annual	May-Oct	Annual

		Estimate		Estimate		
1992	125	102	167	132	44	68
1993	110	91	148	117	44	75
1994	136	117	174	144	49	73
Average 1992-1994	125	102	163	132	46	72
1995[1]	102	91	193	163	47	65
1996	83	106	87	110	46	87
1997	30	34	30	49	39	58
1998	30	30	15	15	49	79
1999	83	79	114	91	73	89
Average 1996-1999	57	61	61	68	52	78

[1] The stockpile was capped in 1995.

$$\text{Average Daily Flow (Lmin)} = \frac{\text{total volume (L)}}{\text{\# days x 1440 min/day}}$$

Notes: The number of days used for the annual estimate was 245: April 1 through November 30. Annual average precipitation for Babbitt, from 1961-1990, was 28.49 inches. Annual May-October precipitation for Babbitt (1961-1990) was 21.16 inches (data from Minnesota Climatology Working Group; www.climate.umn.edu).

Figure 2. pH, nickel and sulfate concentrations vs. time for the input and output of the original portion of the W1D treatment system.

Table 2. Mean water quality data, W1D study cell, 1997-99.

Year	n	pH		Ni		Cu		Co	
		In	Out	In	Out	In	Out	In	Out
1997	13	7.02	7.19	0.346	0.240	0.013	0.011	0.003	0.001
1998	3	7.47	7.49	0.216	0.277	0.021	0.017	0.042	0.036
1999	7	7.25	7.28	0.374	0.297	0.013	0.010	0.008	0.006
Ave. 97-99	---	7.16	7.24	0.324	0.259	0.015	0.012	0.017	0.010

Year	n	Zn		Ca		Mg		SO_4	
		In	Out	In	Out	In	Out	In	Out
1997	13	0.033	0.030	180	170	160	160	860	810
1998	3	0.028	0.035	220	220	230	220	1040	910
1999	7	0.054	0.031	210	200	200	200	640	650
Ave. 97-99	---	0.035	0.032	200	180	190	180	880	810

Notes: Nickel concentration in the W1D study cell decreased by 20–30%, a value consistent with the value measured in 1992-1994. The 1998 data is anomalous, and is likely due to the limited number of samples collected and the very low flow conditions that existed in 1998.

n = number of samples for the year

Substrate Samples

The initial trace metal content of the mixture of peat and peat screenings that was added to the wetland was very low; all concentrations were less than 10 mg/kg (Table 3). By 1996, the peat substrate had accumulated measurable amounts of copper, nickel, cobalt and zinc. Nickel concentrations were the highest and varied with depth and location within the cell. The maximum metal concentrations were generally in the top 10 cm of the peat, and nickel concentrations varied from 70 mg/kg to around 14,600 mg/kg (Figure 3). The average nickel concentration for the 0-10 cm segment in the cell was 4960 mg/kg, and decreased to 2110 mg/kg for the 20-30 cm segment (Table 3). Copper, cobalt, and zinc generally followed the same pattern as nickel but concentrations were about an order of magnitude lower. The average copper, cobalt, and zinc concentrations in the top 10 cm ranged from 57 to 136 mg/kg (Table 3).

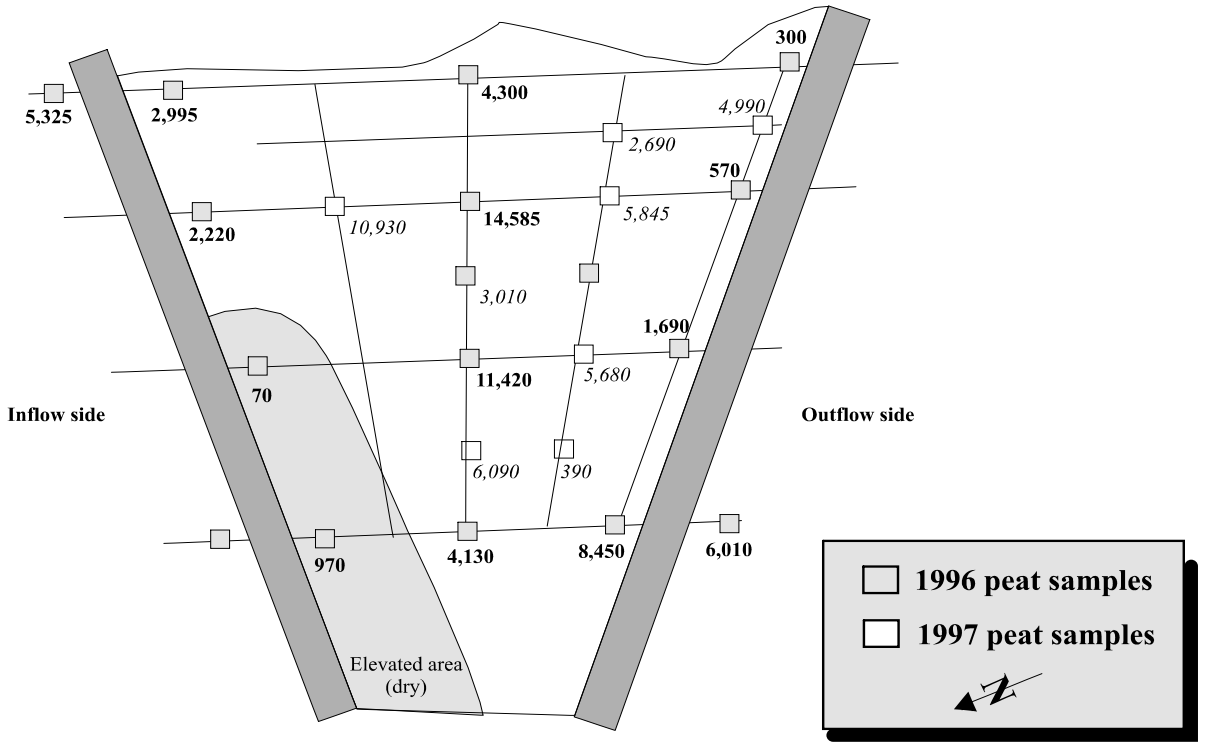
Mass Removal

Overall mass into and out of the wetland was calculated by multiplying the average concentration for the month by the average daily flow for that month. Daily flow data were generally available from May through October, but for April, November and December, there were only a few individual flow readings. Since both flow and precipitation in November and December tended to be low, the average of the limited individual measurements was assumed to be a reasonable estimate of flow. An average value may not provide a reasonable estimate of spring melt flow, since the volume and timing of flow depends on the amount of moisture in the snow pack, temperature and

rainfall. However, metal concentrations during April were about one-half the summer values, so

Table 3. Average metal concentrations in substrate of the study cell (1996-97).

Depth	n	Nickel (mg/kg)	Copper (mg/kg)	Cobalt (mg/kg)	Zinc (mg/kg)
0-10	24	4,959	102	57	136
10-20	24	2,245	51	23	69
20-30	21	2,110	57	29	73



Original substrate	---	5	5	2	7

n = number of samples

Figure 3. Nickel concentrations (mg/kg) in the 0-10 cm layer of peat in the W1D study cell.

the total mass input during April, even with higher flows, would tend to be lower than summer months. From 1992 through 1995, when input load was the highest, the May to October input mass accounted for 86% of the annual load (Eger et al., 2000).

The total mass removed by the wetland was the difference between the input and the output masses. Overall mass removal in the wetland ranged from 171 kg in 1994 to 3 kg in 1997, and corresponded to a percent removal that ranged from 38 to 91 percent (Table 4). Lower mass removal occurred from 1995 to 1999 due to the much lower nickel input to the wetland.

W1D Study Cell

Mass removal in the study cell was calculated from flow and water quality data and from the metal concentrations in the peat. Since there was no significant input of surface and ground water into the cell, there was essentially no change in the concentration of conservative parameters like magnesium (Table 2). As a result, the mass removal of nickel in the study cell could be estimated

Table 4. Nickel mass removal, W1D wetland and study cell, 1992-99.

	Entire wetland					Study cell		
	Mass into wetland (kg)	Mass out of wetland (kg)	Annual mass removal (kg)	Mass removal (%)	Cum. mass removal (kg)	Annual mass removal (kg)	Cum. mass removal (kg)	Calcu-lation method
1992	158	16	142	90	142	37	37	1
1993	162	21	141	87	283	37	74	1
1994	190	19	171	91	454	44	118	1
1995	76	26	50	66	504	13	131	1
1996	36	15	21	58	525	5	136	1
1997	8	5	3	38	528	1	137	2
1998	5	1	4	80	532	1	138	2
1999	16	5	11	69	543	3	141	2

by using the change in concentration. For 1992 to 1994, the average change in nickel concentrations was 26%, so it was assumed that 26% of the total annual mass removal occurred within the cell. Percent removal over the study cell in 1997 and 1999 also averaged 26% (Table 3). As a result, a constant percent removal of 26% was assumed for all years (Table 4). The overall nickel removal in the cell from 1992-1999 was 141 kg, with 84% of the removal occurring between 1992 and 1994. Between 1992 and April 1997, when the final peat samples were collected, 136 kg was removed.

Two methods were used to calculate the mass removal in the study cell from the metal concentrations in the peat. The first method used a computer model which contoured the nickel

values in the peat and assigned concentrations to specific areas within the cell The second method made an overall estimate by multiplying the average nickel concentration in each 10 cm layer of peat by the mass of peat. Since the peat samples were collected in April of 1996 and 1997, the value calculated from the peat would represent the total mass removed through 1996. The total nickel mass in the substrate calculated with these methods was reasonably close to the 136 kg calculated from the water quality data. The computer model calculated the mass to be 112 kg, while the average calculation method gave an estimated value of 126 kg.

Discussion

The overall objective of this study was to determine the lifetime of the wetland treatment system. LTV designed the original W1D system based on average input values for 1990-1991. For this time period, average daily flow was 78 L/min and the average nickel concentration was 5.4 mg/L (Eger et al., 1996). Based on a wetland area of 7000 m^2, an effective removal depth of 20 cm, a peat bulk density of 0.1 gm/cm^3, a maximum removal capacity of 10,000 mg nickel/kg dry peat, and flow from April through November (245 days), the design lifetime was:

$$\text{Lifetime (years)} = \frac{\text{total removal capacity of the wetland (kg nickel)}}{\text{annual load (kg nickel/year)}}$$

$$= \frac{\text{volume of reactive peat x bulk density x removal capacity}}{\text{average daily flow x average nickel concentration x number of days of flow}}$$

$$= \frac{7000\ m^2 \times (100\ cm/m)^2 \times 20\ cm \times 0.1\ gm/cm^3 \times 10{,}000\ mg\ nickel / kg\ peat \times 10^{-3}\ kg/mg \times 10^{-6}\ kg/mg}{20\ gal/min \times 3.785\ L/gal \times 1440\ min/day \times 5.4\ mg\ nickel/L \times 10^{-6}\ kg/mg \times 245\ days\ of\ flow/year}$$

$$= \frac{1400\ kg}{144\ kg} \quad \_ \quad 10\ years$$

In 1995 the entire top of the stockpile was covered with a 30 mil linear low density polyethylene liner (LLDPE) and this cover prevented water from contacting most of the reactive material in the stockpile. Flow at the W1D weir dropped 55%, from an average May to October flow of 125 L/min during 1992-1994, to 57 L/min for the post-closure period (1996-1999).

By preventing precipitation from infiltrating the stockpile and contacting the reactive material, the transport of reactive products was significantly reduced. Nickel concentrations decreased from an average of 3.98 mg/L for 1992-1994 to 0.74 mg/L for 1996-1999. Since both flow and nickel concentrations decreased, the overall load to the wetland decreased by about 90% (Table 4). By reducing the load, the estimated lifetime was increased substantially, from the initial design lifetime of 10 years to about 150 years (Table 5).

The ultimate goal of a passive treatment system is to provide permanent treatment. The major mode of metal removal in this system is assumed to be the same as observed in the initial test cell study (Eger et al., 1994).

Over 90% of the nickel removal in the test cells occurred within the substrate, through a series of reactions (adsorption, ion exchange, chelation) associated with the

Table 5. Effect of stockpile capping in 1995 on load to the W1D wetland and wetland lifetime.

Year	Annual nickel load (kg)	Percent of pre-capping load	Lifetime of wetland[1] (years)
Initial design lifetime	144	---	10
1992-1994	170	---	8
1995	76	45	18
1996	36	21	39
1997	8	5	175
1998	5	3	280 (self sustaining)[2]
1999	16	9	88

[1] Lifetime is calculated by dividing the initial removal capacity of the wetland (1400 kg) by the annual load.

[2] Annual nickel load to the wetland is less than the estimated annual gain in nickel removal capacity (7 kg); the wetland is thus self-sustaining.

organic fraction of the peat. With these types of removal mechanisms, the wetland will have a finite life unless new removal sites can be generated at a rate greater than or equal to the incoming metal load (Eger et al., 1994).

New sites are generated as vegetation dies and new organic substrate accumulates. The average rate of peat accumulation in northern wetlands is

about 1 mm/year (Craft and Richardson, 1993). If the removal capacity of the newly accumulated material is assumed to be 10,000 mg nickel/kg, the wetland would add 7 kg of nickel removal capacity each year:

$$\text{Annual gain in nickel removal capacity} = \text{rate of peat accumulation} \times \text{nickel removal capacity} \times \text{wetland area}$$

$$= \frac{1\ \text{mm/year} \times 0.1\ \text{cm/mm} \times 10{,}000\ \text{mg Ni/kg} \times 7000\ \text{m}^2}{(100\ \text{cm/m})^2 \times 0.1\ \text{gm/cm}^3 \times 10^{-3}\ \text{kg/gm}}$$

= 7 kg Ni/year

In 1998 the annual nickel input load was less than the gain in nickel removal capacity. If the annual input load is less than or equal to the annual increase in removal capacity, the wetland should be self-sustaining. In 1999 the input load was greater than the sustainable load, but the increased load was the result of above normal precipitation, particularly in July when 24.1 cm of rain fell. Using the average of the 1997-1999 input load (10 kg) as representative of the post-closure period, and assuming an annual increase of 7 kg nickel removal capacity, the projected lifetime for the wetland is about 290 years (Figure 4).

If the treatment is to be sustainable and effective, not only must there be new metal removal capacity generated, but the metal must be retained within the wetland. Mass balances calculated on wetland test cells demonstrated that over 99% of the removed metals were associated with the

substrate and less than 1% of the total removal occurred in the vegetation (Eger et al., 1994). These results were consistent with earlier studies on metal removal in a white cedar wetland (Eger and Lapakko, 1988) and with data reported by others (Skousen et al., 1992, Wildeman et al., 1993). Nickel contained within the substrate of the study cell accounted for essentially all of the total nickel removal that was calculated from the change in water quality data. Sequential extraction tests, conducted on a series of substrate samples collected from test cells constructed at the Dunka Mine, demonstrated that only 1-2% of the nickel was water soluble and could, therefore, be easily removed from the substrate (Eger et al., 1994).

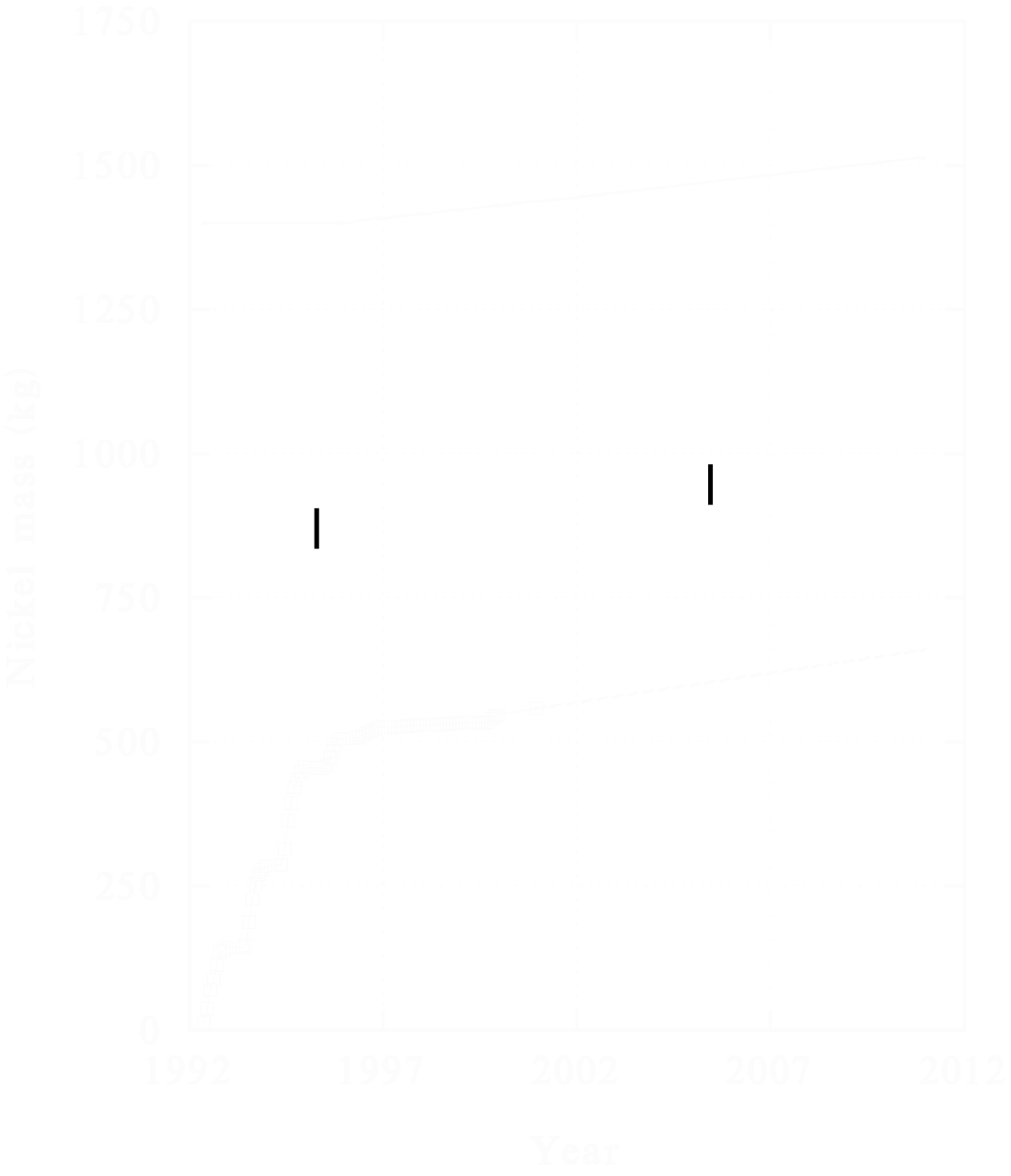


Figure 4. Nickel mass removal in the W1D system, and projected nickel removal capacity.

Additional evidence for the permanent nature of the removal in the wetland is that nickel removal continued despite a decrease in the input concentration of almost an order of magnitude. If the nickel was weakly bound to the substrate, nickel would be

released from the substrate as nickel concentrations in the water decreased, and no removal would occur. Although continuous flow data is only collected from May through October, water quality samples are collected whenever there is water flowing into or out of the wetland. Over the seven years of operation, output concentrations have rarely exceeded input values, and there has always been nickel removal in the wetland (Figure 2).

Conclusions

Since 1995, when the Dunka mine was closed and the stockpiles capped, nickel loads into the W1D wetland have dropped by an order of magnitude. Nickel has been removed every year and there has been no evidence of nickel release from the wetland. The nickel load into the wetland is now about the same as the estimated annual production of new removal sites. If conditions remain unchanged, treatment could continue indefinitely.

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