

# Performance of a 16 ha Engineered Wetland for the Treatment of Neutral-pH Gold Mine Effluent (Ontario, Canada)

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## ABSTRACT

The Campbell Complex (Goldcorp Inc.), located in northwestern Ontario, Canada, has been the site of gold-ore milling operations since 1949. As part of water management measures for the site, an engineered surface-flow wetland was commissioned in 2002 to facilitate final polishing of neutral-pH effluents prior to discharge. The wetland is 16 ha in area, accommodates flows of ~10,000 m<sup>3</sup>/day, and affords a mean hydraulic retention time (HRT) of 6-7 days. Vegetation consists predominantly of transplanted cattails (*Typha spp.*) with substrates comprised of inert mining waste-rock overlaid by organic topsoil containing *Typha* rootstock and native flora. The system operates during the ice-free period only (May through October), and serves as the final point of discharge for the mine. The wetland serves as a net sink for ammonia (NH<sub>3</sub>), cyanide (CN), arsenic (As) and copper (Cu). Over the period 2011-2013, NH<sub>3</sub>-N showed a pronounced decrease in concentration through the wetland, from a mean influent value of ~11 mg/L to a mean of 1.2 mg/L in the wetland discharge (overall reduction of 89% by mass). Over the same period, CN exhibited a mean loading reduction of 78%, with average influent and effluent concentrations of 0.14 mg/L and 0.07 mg/L respectively. Arsenic (primarily as dissolved species) showed a mean loading reduction of 56%, with concentrations being reduced from approximately 0.09 to 0.04 mg/L through the wetland system. Cu loading reductions averaged 33% (2011-2013), with values decreasing from a mean influent concentration of 0.01 mg/L to an outflow mean of 0.007 mg/L. In contrast to Cu and As, Ni shows evidence of remobilization within the wetland complex, with concentrations in the outflow (mean = 0.065 mg/L) exceeding those at the inflow (2011-2013 mean = 0.047 mg/L). Factors likely to contribute to variations in wetland performance include seasonal changes in HRT, redox-controlled processes, metal-CN complexation, and temporal variability in influent chemistry. Overall, the data demonstrate that constructed wetlands can be used effectively for the treatment of gold-mill effluents in cold-interior climates. This is particularly relevant for mine sites that have sufficient water storage capacity upstream of the wetland to allow for seasonal treatment during the warmer months when wetland performance is maximized.

**Keywords:** bioremediation, *Typha*, ammonia, arsenic, copper

## INTRODUCTION

The use of bioremediation in the form of constructed wetlands for the treatment of mine-influenced wastewater has been widely used since the 1980's (Hammer, 1992). The potential utility and performance of constructed wetlands depend on several factors, including climate, the parameters of interest, parameter concentrations, vegetation type, wetland substrate, and the physical characteristics of the wetland itself, such as surface area, water depth, and hydraulic retention time (HRT). The following paper examines a surface flow constructed wetland in northwestern Ontario, Canada, designed for the bioremediation of effluents at an operating gold mine. This study specifically examines the data for the 2011, 2012, and 2013 treatment seasons, which extend from May to October in each year. The primary objectives of this paper are to: (a) quantify the magnitude of contaminant removal, (b) delineate the biogeochemical controls governing contaminant removal within the wetland complex, and (c), provide a general prognosis for the use of wetlands for the treatment of gold mill effluents in cold-interior climates.

## SITE SETTING

The Campbell Complex (51°03'43"N and 93°44'40"W) comprises part of Goldcorp's Red Lake Gold Mines, and is located in Balmertown, Ontario, 7 km northeast of the town of Red Lake. The mine has been in continuous operation since 1949, following discovery of the deposit in the 1940's gold rush in the region. The project is located within the cold-interior region of North America, with mean monthly temperatures ranging from -19.3°C (January) to 18.3°C (July) (annual daily temperature of 0.9°C). The site receives an average of 644 mm of precipitation per year, with rainfall accounting for approximately 73% of the total. Snow cover generally persists from November until late March.

Ores are processed using a combination of grinding, flotation, pressure oxidation and carbon-in-pulp processing. Tailings slurry is sent to an SO<sub>2</sub>/Air treatment circuit for CN destruction prior to discharge to the Main Pond. From the Main Pond, water is recycled back to the process plant and treated with lime to further precipitate dissolved metals. The treated effluent is then conveyed to the Settling/Polishing Ponds which afford 750,000 m<sup>3</sup> of storage for the polishing of treated effluent from the Main Pond. During warmer months (May through October) this polished effluent is conveyed to a 16 ha constructed wetland, which allows for further removal of NH<sub>3</sub>, CN and trace elements. The wetland complex is divided into 34 cells, separated by internal dykes to maximize the length of the flow path and water retention time. All cells host transplanted cattails (*Typha spp.*) in a substrate composed of inert waste rock overlain by *Typha* rootstock, organic matter, and soil/loam. Water depths range from 15 to 70 cm (mean  $\cong$  45 cm). A mean flow of 10,000 m<sup>3</sup>/day, a total surface area of 16 ha, and a mean water depth of 45 cm, translate to a HRT of ~7 days.

Effluents from the Polishing Pond are conveyed to two distinct inputs to the wetland (WET-IN and WET-IN2). These flows are treated in parallel wetland circuits, with the flows merging prior to discharge to Balmer Lake. The discharge from the wetland to Balmer Lake (WET-OUT) serves as the final point of discharge for the mine, and where Ontario compliance standards apply with regards to water quality and toxicity. The system of water management and storage upstream of the wetland facilitates the removal of total suspended sediments (TSS) prior to entry into the wetland (TSS <3 mg/L in wetland inflow). Accordingly, the performance of the wetland relies solely on processes

that remove dissolved species from solution. Inflows to the wetland are circumneutral in pH ( $6 < \text{pH} < 7$ ).

## METHODOLOGY

This study focuses on the treatment seasons of 2011, 2012 and 2013. These years correspond to the period of maximum wetland area (expanded to 16 ha in early 2011) and implementation of a new SO<sub>2</sub>/Air treatment system (commissioned in late 2010). The latter had a significant effect in reducing the concentrations of CN and trace metals in inflows to the wetland complex. The 2011 and 2013 treatment seasons extended from late May to the end of October in each year. A shorter treatment season was implemented in 2012 (May through August).

This paper focuses on parameters (NH<sub>3</sub>, CN, As, Cu, and Ni) although other parameters were examined to elucidate contaminant behaviour (*e.g.*, Fe, Mn, Se and U). Temporal resolution of the data varies by location and parameter. At WET-OUT, samples of all five major parameters as well as sulfate, were collected three times per week through the treatment period. In contrast, influent data (WET-IN) were collected once per week for NH<sub>3</sub>, As, Cu, Fe, and Ni, and once per month for CN and sulfate. Flows are measured continuously at WET-OUT using an OCM III Ultrasonic flow meter. For loading calculations, inflow rates to the wetland were assumed to be equal to those at WET-OUT. Specifically, the influences of precipitation and evapotranspiration were assumed to be negligible given the magnitude of flow through the wetland system. HRT was calculated based on wetland dimensions and flow, and supported with analysis of lag times inferred from the timing of water quality changes between WET-IN and WET-OUT.

To provide additional insight in the role of redox controls within the wetland, high-resolution vertical profiles of dissolved oxygen (DO), temperature, pH, ORP and conductivity were collected from 7 locations within the wetland (July 2014). At these same 7 sites surface (0-3 cm) sediments were collected and analyzed for grain size, organic carbon, major elements and minor elements.

## RESULTS AND DISCUSSION

### Hydrology and Retention Time

Mean daily flow rates through the wetland for 2011, 2012 and 2013 were 11,200 m<sup>3</sup>/day, 10,600 m<sup>3</sup>/day and 7,200 m<sup>3</sup>/day (Figure 1), translating to total volumes of 1.9 million m<sup>3</sup>, 1.1 million m<sup>3</sup>, and 1.1 million m<sup>3</sup>, respectively. Wetland HRT typically varied between 6-7 days depending on the discharge rate from the Polishing Pond, with an overall 2011-2013 mean of 6.4 days. Estimates of HRT are supported by lag times observed between WET-IN and WET-OUT for changes in concentration of conservative parameters such as sulfate.

### Water Quality

#### *Ammonia*

NH<sub>3</sub> originates from the degradation of CN compounds (*e.g.*, cyanate hydrolysis) as well as from the leaching of residual blasting residues used in underground mining. Influent concentrations of NH<sub>3</sub>-N were variable, with values typically ranging between 10 and 15 mg/L (Figure 2). Concentrations at WET-OUT were greatly reduced compared to WET-IN, with the majority of

values in 2012 and 2013 being <0.5 mg/L. Removal rates ranged from 60 to 1,200 mgN/m<sup>2</sup>/day (2011-2013 mean = 482 mgN/m<sup>2</sup>/day), with mass removal showing a general decrease from 2011 to 2013 (overall mass removal efficiency of 89%)

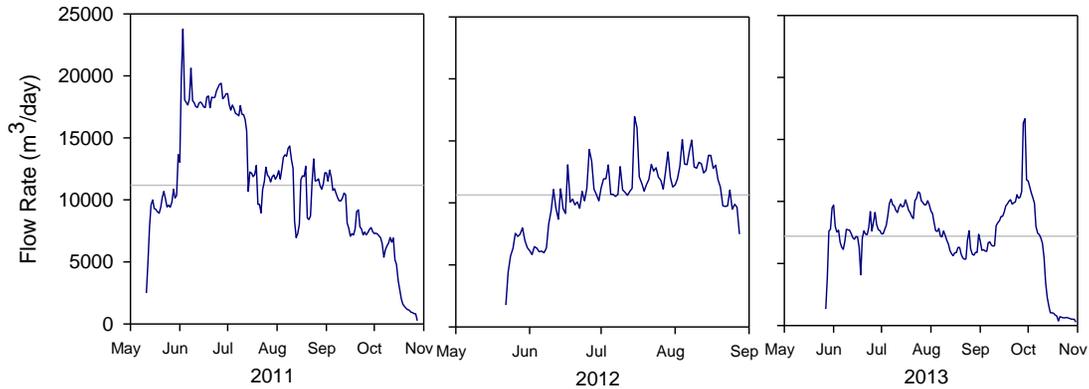


Figure 1 Daily flow rates (measured at WET-OUT) for the 2011, 2012 and 2013 wetland treatment periods.

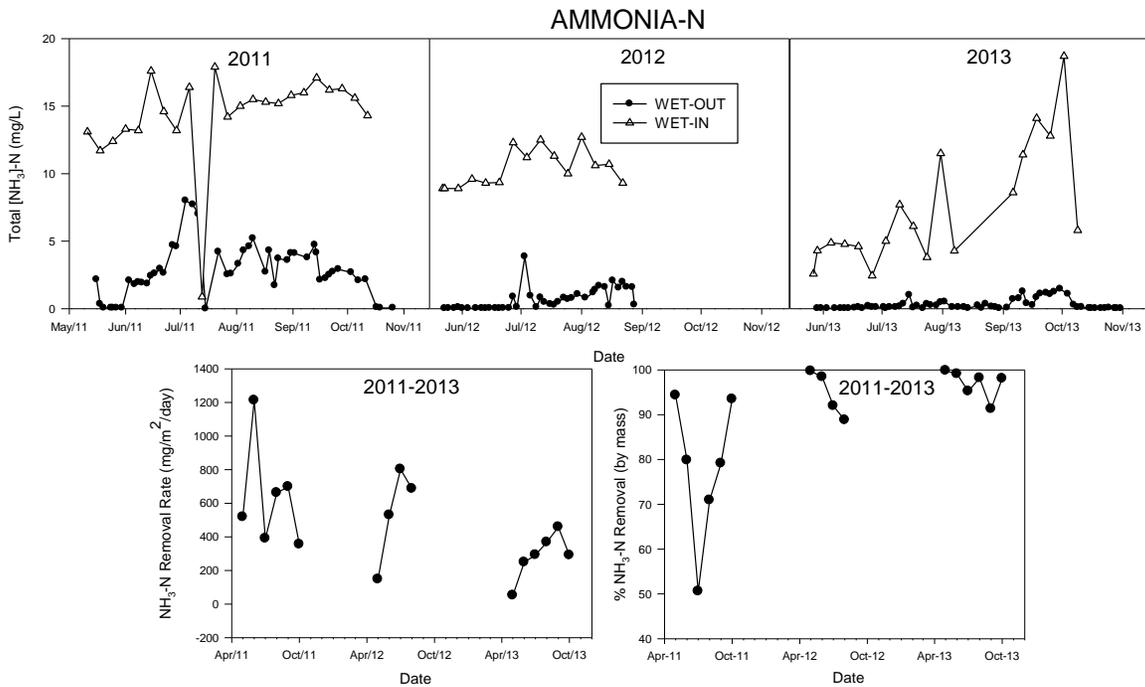
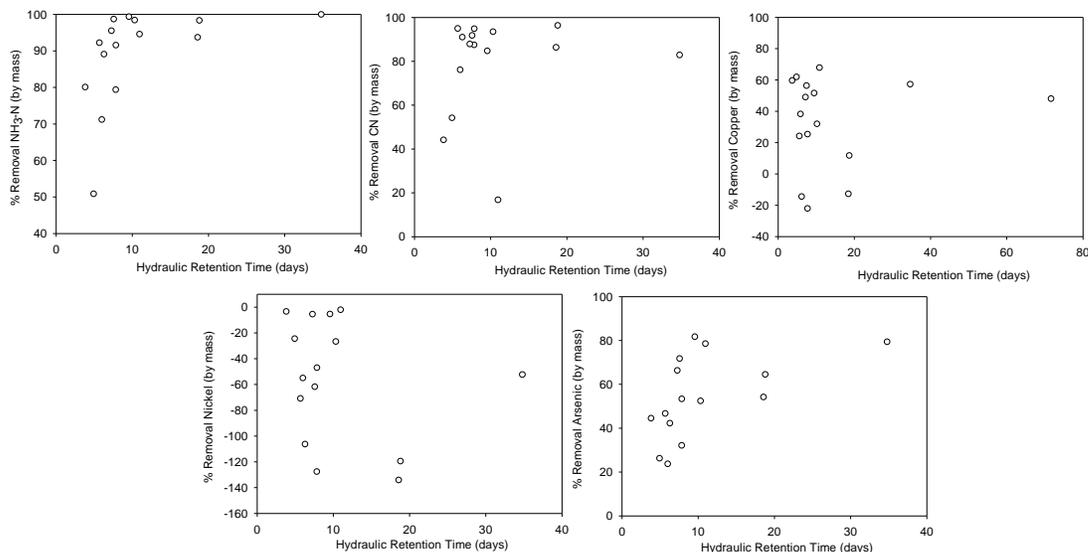


Figure 2 (Top) Time-series of NH<sub>3</sub>-N concentration in the influent (WET-IN) and effluent (WET-OUT) of the wetland complex (May to October of 2011, 2012, and 2013). (Bottom left) Wetland removal rate for NH<sub>3</sub>-N as monthly means for May to October of 2011, 2012, and 2013. (Bottom right) NH<sub>3</sub>-N percent removal efficiency (by mass) as monthly means for May to October of 2011, 2012, and 2013.

(Figure 2). The decrease in removal rate from 2011-2013 is largely due to the general decline in influent NH<sub>3</sub> concentration over the study period. In contrast, removal efficiency generally increased over time, with % removal in 2013 ranging from 91 to 99% (Figure 2). Mechanisms of NH<sub>3</sub> removal include biological uptake (macrophytes, algae), adsorption to substrates and microbially-mediated nitrification (oxidation to nitrate). At the influent pH range (6<pH<7)

ammonia will be predominantly present as ammonium (NH<sub>4</sub><sup>+</sup>); therefore, volatilization of NH<sub>3</sub>(g) is not predicted to be significant (Reddy and Patrick, 1984).

Wetland removal efficiency for NH<sub>3</sub> was seasonal, with maximum efficiency observed at the beginning of the treatment season, and efficiency decreasing to minima in July/August. Comparison of removal efficiency (by mass) as a function of HRT suggests NH<sub>3</sub> percent removal is dependent on flow rate to some degree (Figure 3). Specifically, NH<sub>3</sub> removal efficiency shows an asymptotic relationship with HRT, whereby the two parameters show a positive correlation up to a HRT value of 10 days, beyond which removal efficiency does not improve. The decrease in removal efficiency during the warmer months may also be linked to the remobilization of NH<sub>3</sub> associated with the degradation of organic matter within the wetland, which is expected to be at a maximum during the warmer period. Increased temperatures and rates of organic matter decomposition are also more likely to sustain low oxygen conditions in the water column, which are more conducive to the persistence of NH<sub>3</sub>. Indeed, dissolved oxygen (DO) measurements in the wetland at the end of July 2014 revealed zones of low DO, with values as low as 1 mg/L at some locations. Dunne *et al.* (2013) also showed that releases of NH<sub>3</sub> were greatest during warm periods, and linked this to anoxia in the water column due to increased temperatures and rates of organic matter decomposition.

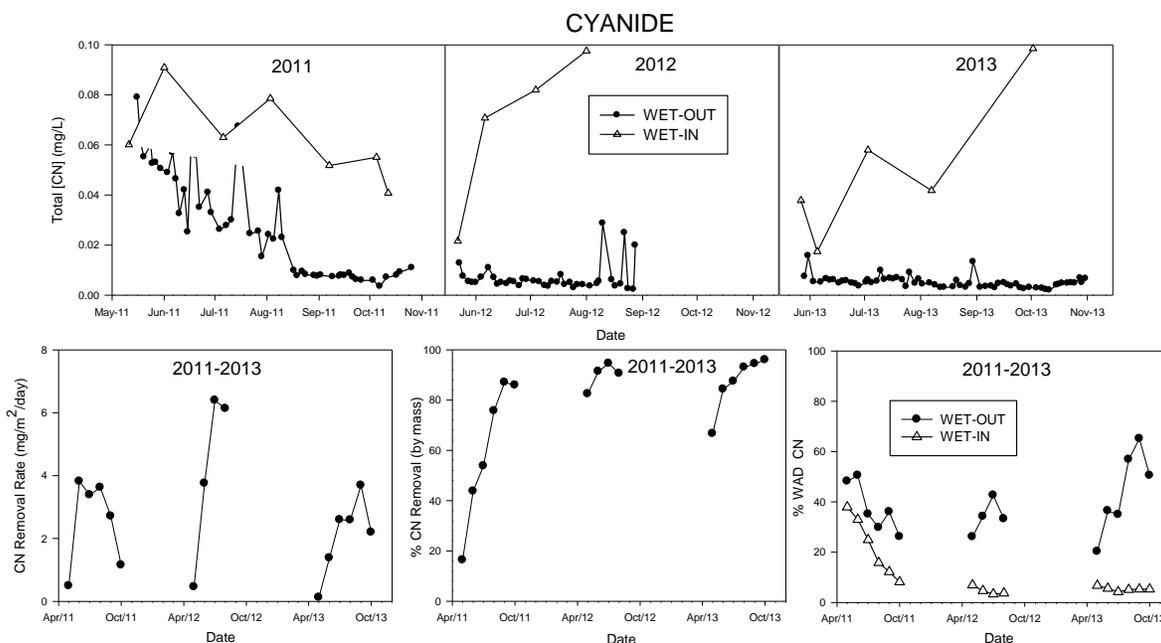


**Figure 3** Wetland removal rates for NH<sub>3</sub>-N, CN, Cu, Ni and As (as monthly means) as a function of hydraulic retention time (monthly means) for the period 2011-2013.

### Cyanide

CN originates from the use NaCN in gold extraction during ore processing. Treatment using SO<sub>2</sub>/Air methods reduced CN to values well below compliance limits prior to entry into the wetland. Values for total CN in the wetland inflow were low and variable (0.02 to 0.1 mg/L) (Figure 4). CN levels were reduced through the wetland system, with most values at WET-OUT in 2012 and 2013 being <0.01 mg/L. Such concentration reductions equate to a mean removal rate of 2.8 mg/m<sup>2</sup>/day (range from 0.2 to 6.4 mg/m<sup>2</sup>/day). Rates of CN removal are variable (17 to 95%; 2011-2013 mean = 78%), and increase over the course of each treatment season in 2011, 2012 and 2013 (Figure 4). For 2012 and 2013, the increases over time in removal efficiency can be explained by the

progressively increasing CN values at WET-IN in combination with the low and invariant values at WET-OUT. Similar to NH<sub>3</sub>, CN removal efficiency shows a weakly-positive relationship to HRT (Figure 3), suggesting flow rate also affects CN removal efficiency.



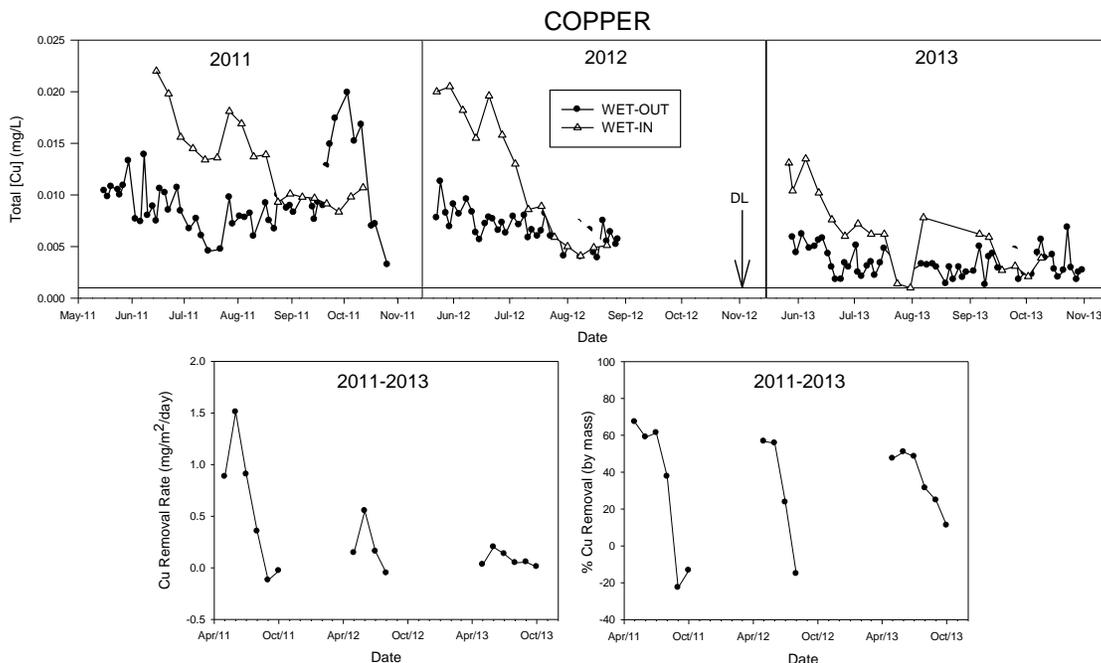
**Figure 4** (Top) Time-series of total CN concentration in the influent (WET-IN) and effluent (WET-OUT) of the wetland complex (May to October of 2011, 2012, and 2013). (Bottom left) Wetland removal rate for total CN as monthly means for May to October of 2011, 2012, and 2013. (Bottom centre) CN percent removal efficiency (by mass) as monthly means for May to October of 2011, 2012, and 2013. (Bottom right) Ratio of WAD CN to Total CN in influent (WET-IN) and effluent (WET-OUT) as monthly means for May to October of 2011, 2012, and 2013.

Mechanisms for CN removal in wetland systems may include volatilization of HCN, UV-catalyzed oxidation, hydrolysis to NH<sub>3</sub> and precipitation of insoluble metal-CN complexes (Gasper and Beck, 1983). Insight into CN behaviour can also be garnered from the proportion of weak-acid-dissociable (WAD) CN in the wetland inflow and outflow (Figure 4). Specifically, in both 2012 and 2013, the %WAD-CN at WET-OUT increases during each treatment season, while the %WAD-CN in the inflow remains constant. These data indicate that either: 1) strong acid dissociable CN compounds (e.g., ferri/ferro CN complexes) are preferentially removed within the wetland complex; and/or 2) WAD-CN complexes are increasingly remobilized into solution over the treatment period. These considerations also have relevance to the interpretation of Ni and Cu behaviour (discussed below).

### Copper

Total Cu concentrations in the wetland inflow were variable, ranging from 0.001 to 0.022 mg/L (mean = 0.011 mg/L) (Figure 5). Over the course of each treatment season, inflow Cu concentration generally declined, which reflects processes occurring upstream in the tailings management area (TMA). Concentrations of Cu were significantly reduced by the wetland, with a mean outflow value of

0.0065 mg/L (range of 0.001 to 0.02 mg/L) (Figure 5). Cu removal rates range from -0.12 to 1.5 mg/m<sup>2</sup>/day.

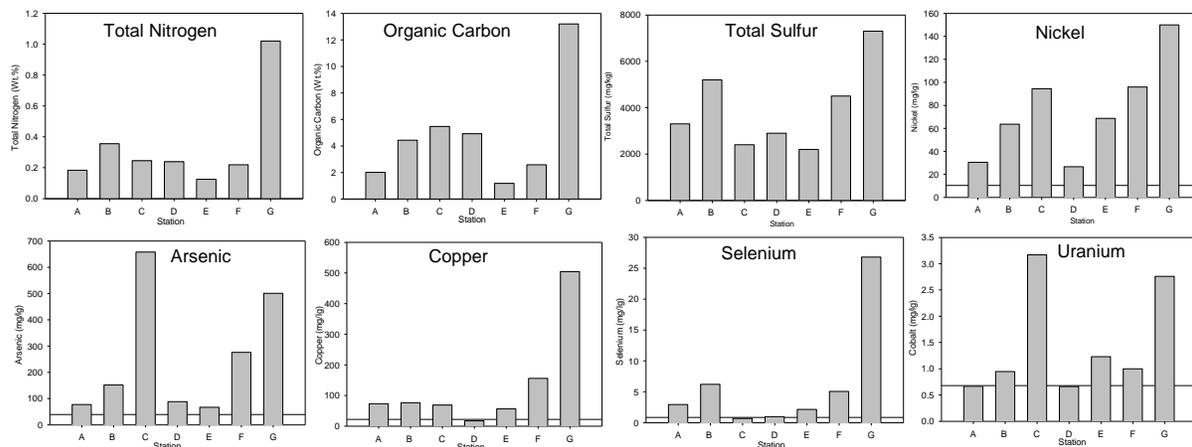


**Figure 5** (Top) Time-series of total Cu concentration in the influent (WET-IN) and effluent (WET-OUT) of the wetland complex (May to October of 2011, 2012, and 2013) DL = detection limit (0.002mg/L). (Bottom left) Wetland removal rate for total Cu as monthly means for May to October of 2011, 2012, and 2013. (Bottom right) Total Cu percent removal efficiency (by mass) as monthly means for May to October of 2011, 2012, and 2013.

Removal rates declined over the course of each treatment season (May – October), possibly related to the decreases that occurred to inflow concentration (*i.e.*, decreased removal efficiency with decreasing inflow concentration). Cu removal efficiency ranged between -23% to 68% (mean = 33%), indicating that in some months, the wetland served as a source of Cu rather than a sink (Figure 5). Cu removal efficiency was weakly proportional to HRT, suggesting flow rate is not a dominant variable with respect to percent removal (Figure 3).

Possible sinks for Cu in wetland systems include adsorption (*e.g.*, Fe oxides, organic matter, clays), biological uptake (bacteria, algae, biofilms, macrophytes), and precipitation as secondary phases (*e.g.*, Cu sulfides) (Sobolewski, 1999). As a means to assess the nature of Cu removal, elemental data for the near-surface substrates were examined. Cu concentrations in surface sediments were variable, and ranged from 17 to 504 mg/kg (mean = 136 mg/kg) (Figure 6). Most values greatly exceeded the local background (22 mg/kg) for sediments collected in a nearby reference wetland, demonstrating that the sediments are serving as a pronounced sink for Cu. Sedimentary Cu showed strong correlations ( $r^2 > 0.7$ ) with parameters indicative of organic matter enrichment, including T-N and TOC (Table 1). Cu also shows strong correlations with parameters that preferentially accumulate in reducing sediments, including S and Se. In particular, the observation of Se enrichment in zones of high organic carbon provide strong evidence to indicate redox controls are important, given the tendency of dissolved Se to accumulate in organic-rich reducing sediments (Martin *et al.*, 2011). This is also supported by observations of low DO values in the wetland water

column (as low as 1 mg/L in some locations). Overall, the data suggest that Cu accumulation in sediments is in part tied to sedimentary organic matter abundance, which may reflect Cu uptake by macrophytes/algae and/or precipitation of Cu as secondary sulfides.



**Figure 6** Solid-phase concentrations of total nitrogen, organic carbon, total sulfur and select trace elements (Ni, As, Cu, Se and U) in the upper 3 cm of wetland substrate at seven locations in the wetland complex. Horizontal lines for trace elements represent background values for a control wetland in the local area.

**Table 1** Coefficients of determination ( $r^2$ ) for correlations between total nitrogen (T-N), total organic carbon (TOC), sulfur (S) and select trace elements (As, Cu, Fe, Mn, Ni, Se and U) in the upper 3 cm of wetland sediments (n=7).  $R^2$  values >0.7 are shaded.

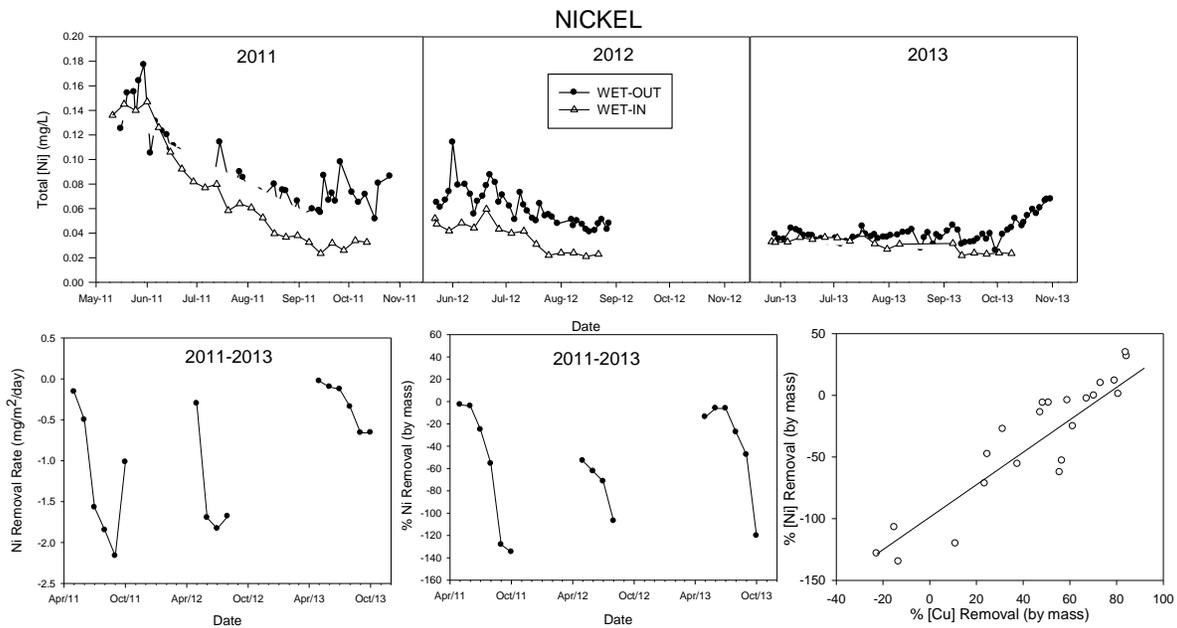
Parameter	T-N	TOC	As	Cu	Fe	Mn	Ni	Se	S	U
T-N	1	0.92	0.24	0.90	-0.02	-0.019	0.57	0.95	0.77	0.29
TOC		1	0.38	0.74	-0.01	0.01	0.52	0.77	0.57	0.42
As			1	0.24	0.31	0.38	0.57	0.15	0.08	0.89
Cu				1	-0.03	-0.05	0.72	0.96	0.74	0.28
Fe					1	0.77	0.11	-0.05	-0.10	0.40
Mn						1	0.03	-0.08	-0.21	0.46
Ni							1	0.61	0.44	0.58
Se								1	0.82	0.20
S									1	0.20
U										1

### Nickel

Ni concentrations in the inflow to the wetland showed a strong decline during the 2011-2012 period, from approximately 0.17 mg/L (June 2011) to 0.02 mg/L (August 2012), after which time values remained relatively invariant (Figure 7). This decrease in Ni may in part reflect implementation of a new SO<sub>2</sub>/Air CN destruction circuit at the end of 2010, which would be expected to lower Ni solubility in response to reducing the potential for CN-Ni complexation. In contrast to Cu, Ni concentrations at the outflow are consistently higher than those at the inflow, demonstrating that the wetland behaved as a net source for Ni during 2011, 2012, and 2013 (Figure

7). Mean loading to the outflow was 0.9 mg/m<sup>2</sup>/day (2011-2013), translating to a mean increase of 54% (by mass) over the same period (Figure 7).

The source of Ni within the wetland complex is not clear. Possible reasons include the leaching of Ni from Ni-bearing waste rock used in wetland construction, or more likely, the remobilization of Ni through desorption processes. With respect to the latter, the remobilization of Ni through CN complexation may play a role. As described in the discussion of CN, the removal of CN appears to be linked to the preferential removal/destruction of strong-acid-dissociable species (*e.g.*, Fe-CN). This process may allow for secondary complexation of liberated CN<sup>-</sup> with other trace elements (*e.g.*, Ni) within the wetland complex. The strong correlation between Ni and Cu removal efficiencies (Figure 7, bottom right) implies that similar factors may apply to Cu.



**Figure 7** (Top) Time-series of total Ni concentration in the influent (WET-IN) and effluent (WET-OUT) of the wetland complex (May to October of 2011, 2012, and 2013). (Bottom left) Wetland removal rate for total Ni as monthly means for May to October of 2011, 2012, and 2013. (Bottom centre) Total Ni percent removal efficiency (by mass) as monthly means for May to October of 2011, 2012, and 2013. (Bottom right) Relationship between Ni and Cu percent removal efficiencies (regression of monthly means for period 2011-2013).

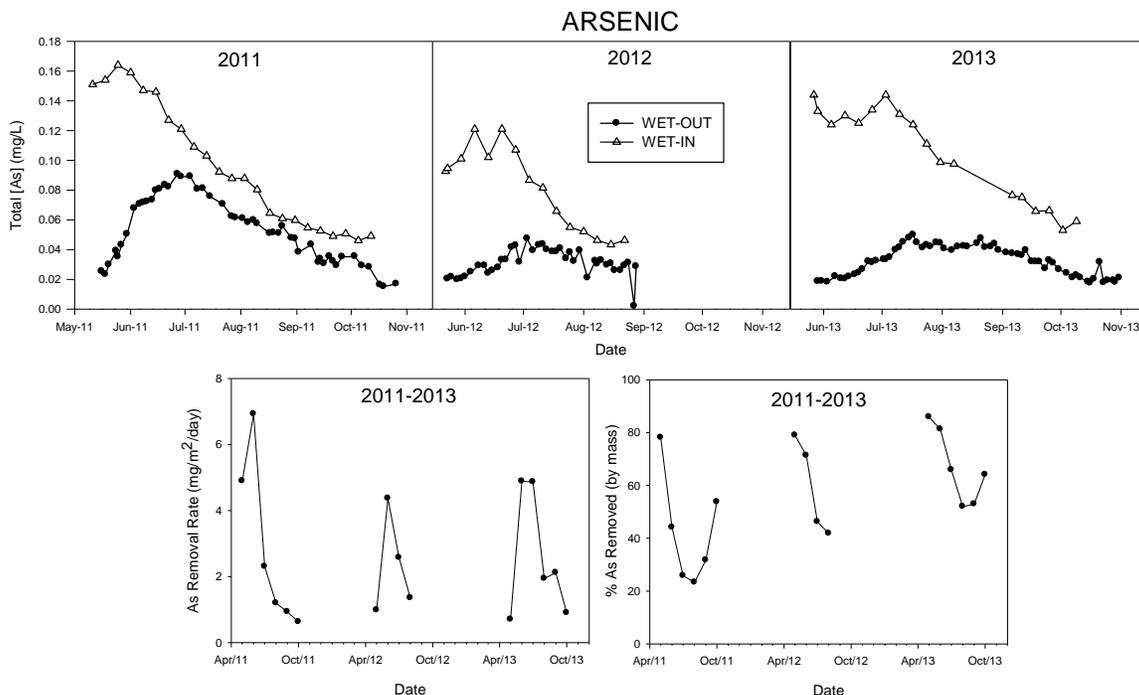
Alternatively, the remobilization of Ni from within the wetland complex may be related to the decline in Ni influent values during 2011-2012, which was likely the result of improved CN treatment upstream. Specifically, the pronounced decline in Ni concentrations in the wetland inflow may have promoted desorption of Ni from binding sites on particle/organic surfaces. Metal concentrations in solution that are governed by sorption reactions can be described in terms of a distribution coefficient ( $K_d$ ) (Tessier, 1989), which is the ratio of sorbed metal concentration (expressed in mg metal per kg sorbent) to the dissolved metal concentration (expressed in mg/L) at equilibrium (Equation 1).

$$K_d = \frac{\text{mg metal per kg sorbent}}{\text{mg/L}} \quad \text{(Equation 1)}$$

According to this equation, if the dissolved metal concentration decreases, the sorbed concentration must also decrease to maintain a constant  $K_d$  value. This mechanism, which relies on the assumption of adsorptive equilibrium, provides a possible explanation of the remobilization of Ni from the wetland complex.

**Arsenic**

In the wetland inflow, As concentration consistently declined between June and October in each of 2011, 2012 and 2013, with values ranging from 0.43 to 0.16 mg/L (mean = 0.095 mg/L) (Figure 8). The decrease in As concentration in the wetland influent within each season likely relates to seasonal redox-related processes in the TMA upstream. Arsenic values were consistently lower at the wetland outflow (mean = 0.039 mg/L), indicating that the wetland served as a sink for dissolved As. Mean monthly As removal rates ranged from 0.6 to 6.9 mg/m<sup>2</sup>/day (mean = 2.3 mg/m<sup>2</sup>/day), translating to a mean removal efficiency of 56%. Arsenic removal efficiency showed a positive correlation with HRT (Figure 3), suggesting flow rate affected wetland performance to some degree. Within each treatment year, removal efficiency showed a seasonal decline to minima typically occurring in July or August. Removal efficiency increased late in the season (September) despite the influent concentration continuing to decrease. Such behaviour may relate to redox-controlled processes in the wetland. Specifically, higher rates of Fe-oxide reductive dissolution may be expected during the peak summer period, when rates of bacterially-mediated organic matter oxidation are at a seasonal maximum. This, combined with lower DO values in the water column, may promote the recycling of As into surface waters (Martin and Pedersen, 2002).



**Figure 8** (Top) Time-series of total As concentration in the influent (WET-IN) and effluent (WET-OUT) of the wetland complex (May to October of 2011, 2012, and 2013). (Bottom left) Wetland removal rates for total As as monthly means for May to October of 2011, 2012, and 2013. (Bottom right) Total As percent removal efficiency (by mass) as monthly means for May to October of 2011, 2012, and 2013.

Arsenic showed significant solid-phase enrichment over background values in reference wetland substrates, indicating that the sediments are serving as a sink for As. Possible accumulation pathways include sorption to Fe/Mn oxides, adsorption of arsenite (As<sup>+3</sup>) to particles, and precipitation of secondary As sulfides. Correlations of As with Fe ( $r^2=0.31$ ) and Mn ( $r^2=0.38$ ) are relevant, and suggest some degree of As adsorption with Fe and/or Mn oxides (Table 1). In contrast, As was poorly correlated with S ( $r^2=0.08$ ), suggesting secondary As-sulfide formation is not a dominant accumulation pathway. Interestingly, As showed the strongest relationship with U ( $r^2=0.89$ ) (Table 1). Like As, U is redox sensitive and tends to accumulate as discrete U phases in anoxic sediments (Lovely *et al.*, 1991). The strong link of As to U, and weak correlations of As with S, Fe and Mn, may suggest that As adsorption as arsenite (*e.g.*, to clays and/or organics) may play a significant role in governing As uptake within the wetland complex.

## CONCLUSIONS

The results of this study demonstrate the successful utility of constructed wetlands for the treatment of neutral-pH gold mill effluents in cold interior climates, with parameters including NH<sub>3</sub>, CN, Cu and As showing robust removal efficiencies. The data suggest that contaminant removal relates to a combination of biological uptake as well as redox-related processes that result in the sequestration of trace elements under conditions of suboxia. The successful application of wetland treatment at the Campbell Complex is strongly tied to the water management system, which facilitates sufficient storage within the TMA to allow all mine waters to be discharged through the wetland during the ice-free period (May through end of October). The data also illustrate the effect of several processes that can influence wetland performance on seasonal and inter-annual scales, including changes to influent composition, variability in HRT, and biogeochemical processes that may promote contaminant remobilization.

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