

The East-Sullivan Mine Site: Merging Prevention and Treatment of Acid Mine Drainage¹

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ABSTRACT

The East Sullivan mine was closed in 1966, leaving 15 Mt of acid-generating tailings free to contaminate the environment. An experimental wood waste cover was implemented in the early 80's in order to prevent sulphide oxidation. Contaminant production and dispersion were investigated in the 90's to validate that concept and guide later actions. Ligneous wastes stop oxygen, and also favour infiltration, rising the watertable and allowing a rapid flush of pre-cover, acid-prone, groundwater. These are replaced by reducing and alkaline waters, a geochemical environment that favours sulphate reduction and base metal precipitation. The initial restoration strategy was modified to take advantage of these properties.

An innovative treatment of acid effluents was put forward, capitalizing on the sulphate reduction expected to occur in the water collected around the impoundment and recirculated through the organic cover. The treatment system was implemented in 1998, and operated until 2005, when not required anymore. Despite acid feed water down to pH 2.5, groundwater's pH next to the dispersal zone was maintained above 6. Alkalinity decreased during the recirculation, but was back to 800 mg/L and more the following spring, thanks to sulphate reduction. Fe²⁺ concentrations were maintained below 2 mg/L. Evolution of dissolved iron mass in the reservoirs surrounding the impoundment suggests that the contaminated groundwater flush was completed in the north sector in 1999, in the west sector in 2001, and in the south sector in 2006. It is near completion in the east sector. Since 1999, the water quality at the final effluent, located south of the pumping station, meets the requirements of the Ministry du Développement durable, de l'Environnement et des Parcs du Québec. The wood-waste cover, besides limiting sulphide oxidation, can fill the role of alkaline reducing bioreactor for the treatment of acidogenic waters, until the mass of alkalinity exceeds that of acidity in the effluents. Such situation occurred in 2006. These unconventional approaches have allowed to cut restoration cost by several M\$ in comparison to those required by standard techniques of the early 90's.

RÉSUMÉ

La fermeture de la mine East Sullivan en 1966 laissait 15 Mt de résidus miniers acides libres de contaminer l'environnement. Les premières mesures de restauration, au début des années 1980, visaient l'interruption de l'oxydation des sulfures avec une couverture ligneuse. Des études de mobilisation et dispersion des contaminants ont été réalisées pour valider ce concept et orienter les interventions. La couverture bloque l'oxygène, mais augmente aussi l'infiltration, rehaussant la nappe et purgeant les eaux acidogènes pré-couverture. L'eau en remplacement, alcaline et réductrice, favorise la réduction des sulfates et la précipitation des métaux. La stratégie initiale de restauration a donc été modifiée pour en profiter.

Un traitement innovateur pour les effluents acides a été mis de l'avant, misant sur la réduction des sulfates affectant les effluents du parc recirculés à travers le couvert organique. De 1998 à 2005, l'eau collectée autour du parc à résidus miniers a été traitée en la pompant sur le couvert ligneux. Malgré des eaux d'alimentation ayant un pH aussi bas que 2.5, le pH des eaux souterraines à proximité du point de déversement se maintenait au dessus de 6. L'alcalinité diminuait au cours de la recirculation, mais remontait à 800 mg/L et plus le printemps suivant, grâce à la réduction des sulfates. Les concentrations en Fe²⁺ se maintenaient à moins de 2 mg/L. L'évolution de la masse de fer dissous dans les réservoirs entourant le parc indique que la purge des eaux de pores contaminées a été complétée dans le secteur nord en 1999, dans le secteur ouest en 2001 et dans le secteur sud en 2006. La purge du secteur est sera incessamment complétée. Depuis 1999, la qualité des eaux à l'effluent final rencontre les exigences du Ministère du Développement durable, de l'Environnement et des Parcs du Québec. Comme attendu, le couvert organique, en plus de limiter l'oxydation des sulfures, joue le rôle d'une barrière réductrice et alcaline pour le traitement des eaux acides et ce jusqu'à ce que la masse d'alcalinité dépasse celle de l'acidité dans les effluents. Une telle situation a été observée en 2006. Ces approches non-conventionnelles ont permis de réduire les coûts de restauration de plusieurs M\$, en comparaison aux coûts des techniques courantes du début des années 1990.

¹ This paper is an update of two papers: Germain et al., (2004) and Tassé et al. (2004).

1. INTRODUCTION

East Sullivan was active from 1946 to 1966 in Abitibi, QC, Canada. The Québec government took over the site responsibility in 1980. Nothing had been done, either during exploitation or after closure, to mitigate the site's impact on the surrounding environment and to insure the long term chemical and physical stability of the acid-producing mining wastes. East Sullivan was soon recognized as one of the 28 sites posing a major threat to the environment and public health, with its acid mine drainage (AMD), amongst the 341 hazardous waste deposits of all types tallied by the Ministère de l'Environnement in the 80's (GERLED, 1990).

The placement of a wood waste cover began in 1984, following early proposals that organic covers can prevent oxidation of sulphidic mine tailings (Reardon and Poscente, 1984, and references therein). In this type of cover, atmospheric oxygen is consumed by the oxidation of carbon, hence its absence in the interstitial gases coming in contact with the sulphides. A restoration scheme putting forward seepage collection around the impoundment followed by passive treatment of the effluents started to be implemented in the early 1990's (SNC-Lavalin, 1992). Effluents with near neutral pH, thanks to the organic contribution of the wood waste cover, were expected, based on the quality of water sampled over the impoundment. However, the effluents collected around it turned out to be strongly acidic, rather than near neutral.

That finding brought forward the necessity to have a good understanding of the hydrogeological and

hydrogeochemical processes going on in that peculiar set up, in order to anticipate the general properties of the seepage waters that would be collected around the impoundment over the mid- and long- terms. Studies carried thereafter not only answered to these concerns, but further allowed the recognition of specific properties of the wood waste cover that could be put at work for the site restoration. The wood waste cover, a prevention only device, was turned into a dual prevention plus treatment system, allowing a cutback of restoration expenses, down to 9.5 M\$ by 2002, from the 50 M\$ required by standard, early 90's, techniques (Cyr, 2002).

2. THE EAST SULLIVAN MINE TAILINGS

The East Sullivan mine tailings impoundment is located 6 km east of Val d'Or (77°42'W; 48°05'N; Figure 1). An extensive survey of the site in the early 90's allowed to build a coherent and integrated data base on all aspects of the tailings pile: topography, thickness and extent of both tailings and forestry wastes; physical and chemical properties of tailings (porosity, permeability, grain-size, mineralogy, composition); composition of pore gases, surface waters, and pore waters (vadose and saturated zones, organic and inorganic parameters); watertable elevations. Methods and details appear in Tassé *et al.* (1992) and Tassé and Germain (1996).

Essential information on the site are summarized in Table 1 and sketched in Figure 2. Thickness, composition and grain-size of 1 m thick intervals, sampled at every 2 m from 0.5 m depth, were determined at each control point pictured, for a total of 199 samples.

Table 1. Main characteristics of the mining and forestry wastes.

<u>Mine tailings:</u>			
metals extracted:	Cu, Zn (Au, Ag)	accumulation:	1949-66
sulphide concentration:	3.6% S	tonnage:	15 Mt
sulphide mineralogy:	FeS ₂ (Fe _{1-x} S, CuFeS ₂ , ZnS)	area:	1.36 km ² (+0.68 km ² of spreading)
acid generating potential:	108.9 kg CaCO ₃ /t	morphology:	4-5 m plateau
acid neutralizing potential:	4.1 kg CaCO ₃ /t	thickness:	2-14 m
<u>Organic wastes:</u>			
	<u>northwest</u>	<u>south</u>	<u>northeast</u>
accumulation:	1984-92, then at low pace	since 1992	since 1999
species:	deciduous dominant over conifers	mainly conifers	conifers and deciduous
types of waste:	barks, logs, presswood waste, mostly overlain by sewage sludge	barks, sawdust, and logs	barks
thickness:	0.2 to 6 m (pre-1990) and 2 m (post-1990)	≈ 2 m	≈ 2 m



Figure 1. Localization of East Sullivan mine tailings impoundment.

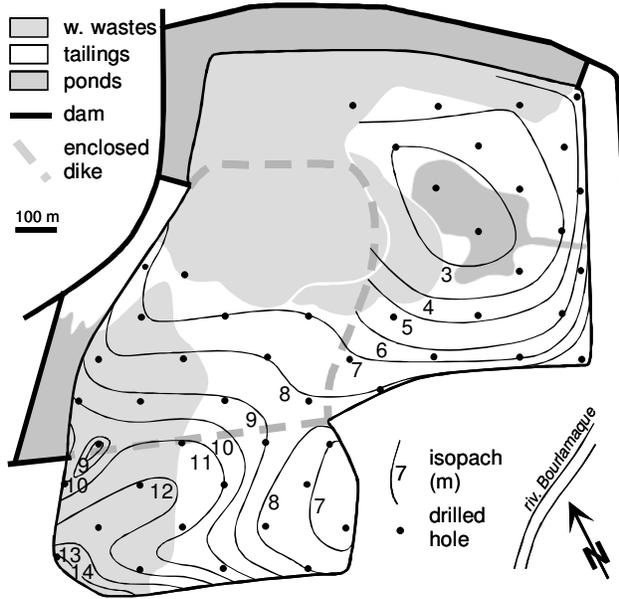


Figure 2. Isopach of the East Sullivan tailings impoundment in 1994.

The initial impoundment, to the south, features coarse grained ($d_{50} = 44 \mu\text{m}$), sulphurous (3.96 % S), and thick tailings (av. 10.1 m). Tailings are getting finer ($d_{50} = 36$ and $27 \mu\text{m}$), less sulphurous (3.15 and 2.10 % S), and thinner (av. 8.1 and 5.1 m) in the later northern (mid-50's) and north-eastern (early 60's) enlargements. In a vertical

profile, coarse ($d_{50} \sim 62\text{-}107 \mu\text{m}$) and fine ($\sim 8\text{-}9 \mu\text{m}$) material alternates in layers 1-17 cm thick (av. 5-7 cm), in coarse/fine proportions that vary from 0.3 in impoundment centres to 3.1 at the margins. The tailings overly an impermeable substratum composed of clayey sediments and bedrock.

3. GEOCHEMICAL PROCESSES IN THE TAILINGS

Continuous core samples recovered from 7 stations (Figure 3) give an insight of the geochemical processes going on in the tailings (Germain *et al.* 1994). Typical results are shown in Figure 4 for st 7, located close to the north-eastern corner of the impoundment. Near-surface pore waters are enriched in H^+ , Fe^{2+} , SO_4^{2-} and metals by sulphide oxidation, either by atmospheric oxygen (Eq. 1) or by ferric iron (Eq. 2). Hydrolysis that follows oxidation of iron (Eq. 3 and 4) produces acid as well. Since the kinetic of oxidation is very slow in acid conditions (Stumm and Morgan, 1996), ferrous iron is allowed to migrate downward in an oxygen-free environment.

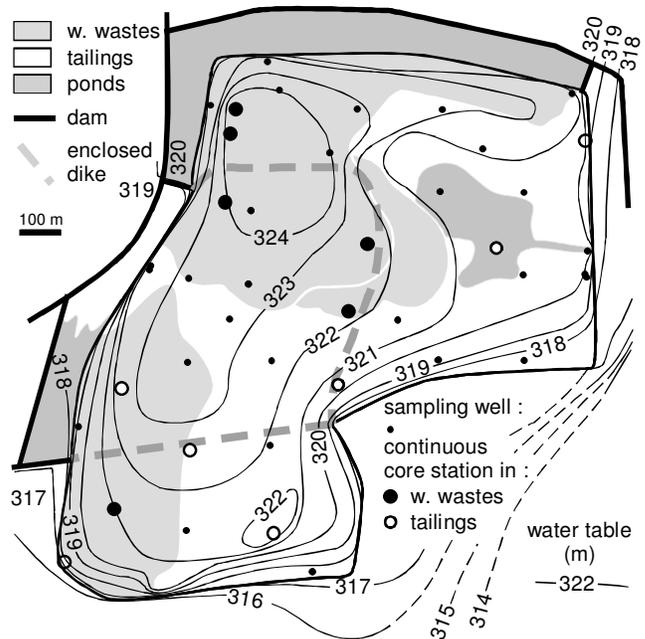
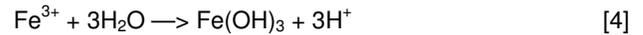
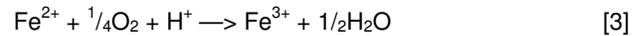
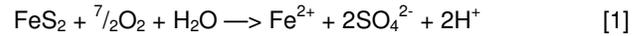


Figure 3. Localization of sampling wells and continuous core stations used in hydrogeochemical groundwater studies; watertable elevation October 24, 1994.

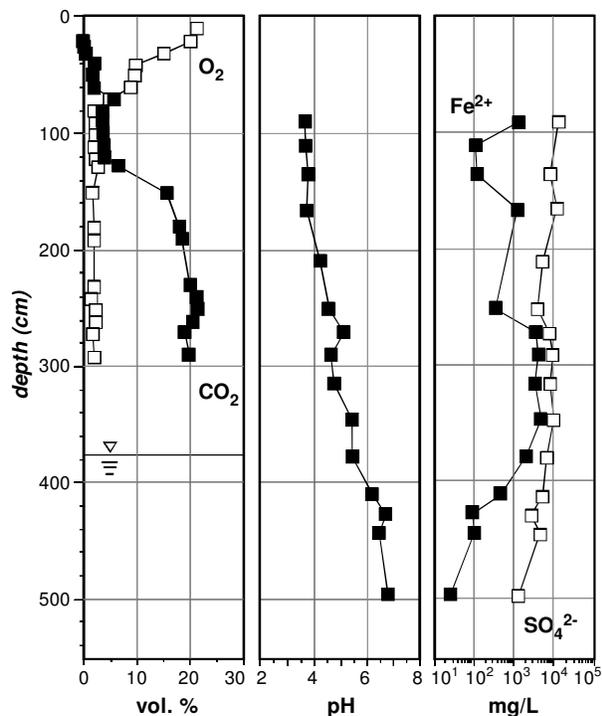


Figure 4. Geochemical profiles at near-margin station st 7

As the pore waters percolate downwards, calcite dissolution releases CO_2 to pore gases and increases pH and Ca^{2+} , easing adsorption and bringing the pore water in the stability field of siderite (FeCO_3) and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). This decreases Fe^{2+} , SO_4^{2-} , and heavy metals concentrations. For instance, concentration of dissolved iron is reduced from 18 000 ppm in the vadose zone to less than 100 ppm in the saturated zone of st 7 (Figure 4). Residual Fe^{2+} is carried to the edge of the tailings pile where it seeps. Oxidation occurs under atmospheric oxygen and hydrolysis can proceed, due to the higher pH acquired upon migration. Alkalinity gained from solid-groundwater interactions cannot always overcome the acid produced this way, especially near margins, where the travel lengths and times are short. Seepage is thus acid, and that this acid is only indirectly related to sulphide weathering at the surface of the tailings. In that scheme, acid production around the impoundment can go on a long time after the placement of an oxygen barrier, since acidity is stored in groundwater as Fe^{2+} , and kept until emergence.

Neutralizing capacities are locally exceeded in the vicinity of enclosed dikes, where coarse grained-tailings favours air infiltration and act as a drain for groundwater (Germain *et al.* 1994). That deterioration represents the long term fate of the impoundment, in the absence of any mitigation, given the acid/neutralizing potentials evaluated from the tailings composition (Table 1).

4. GEOCHEMICAL PROCESSES IN THE WOOD WASTE COVER

The cover functionality in term of oxygen consumption was demonstrated repeatedly (Tassé *et al.* 1994; Tassé, 2000). CO_2 is produced in stoichiometric proportion to the consumption of oxygen during carbon oxidation and varies generally in inverse proportion to that of O_2 , giving a mirror image (Eq. 5; Figure 5).

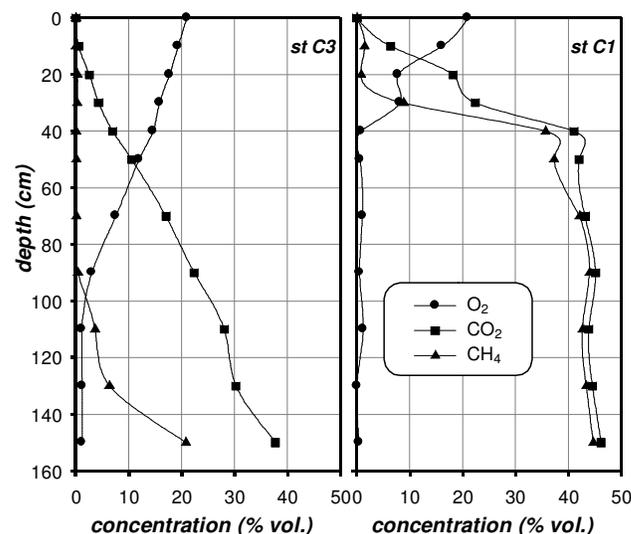


Figure 5. Typical O_2 , CO_2 and CH_4 profiles in East Sullivan forestry wastes (from Tassé, 2000).

Over-representation of CO_2 happens and implies fermentation, releasing both CO_2 and CH_4 from organic substrates (Eq. 6). Methane is the best evidence of strongly anaerobic conditions within the forestry wastes. Tassé (2000) expands on the variables (tree species and part; ages; degree of water saturation, *etc.*) that can affect the degradation processes over the short and long term.



Temperature profiles throughout the cover are influenced by surface temperatures (Figure 6). Freezing did not go below 60 cm in the surveyed year, despite severe winter conditions (average monthly temperature of -17.0 and -15.1°C for January and February). This suggests year-long bacterial activity within the cover. Low temperatures at depth in late April and early May are due to cold water infiltration that follows snow cover melting.

The distribution of alkalinity in the saturated zone below the covered areas shows the beneficial effects associated with the forestry wastes in tailings (Figure 7). Values

exceeding 2000 mg/L-CaCO₃ have been recorded, in contrast to 500 mg/L or less in the sectors dependent on mineral alkalinity alone.

Some of this alkalinity is associated with dissolved organic compounds, and some with dissolved inorganic carbon, due to high partial CO₂ pressures (20-40%) associated to carbon oxidation and near neutral pHs of groundwater. In tailings, similar CO₂ concentrations in the vadose zone result from acid dissolution of calcite. The acid conditions prevent any build-up of comparable alkalinity in the underlying saturated zone.

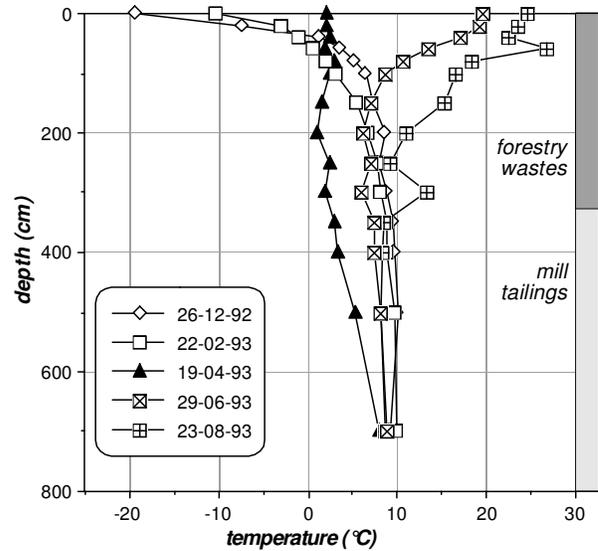


Figure 6. Selected temperature profiles in forestry wastes (from Tassé *et al.* 1997).

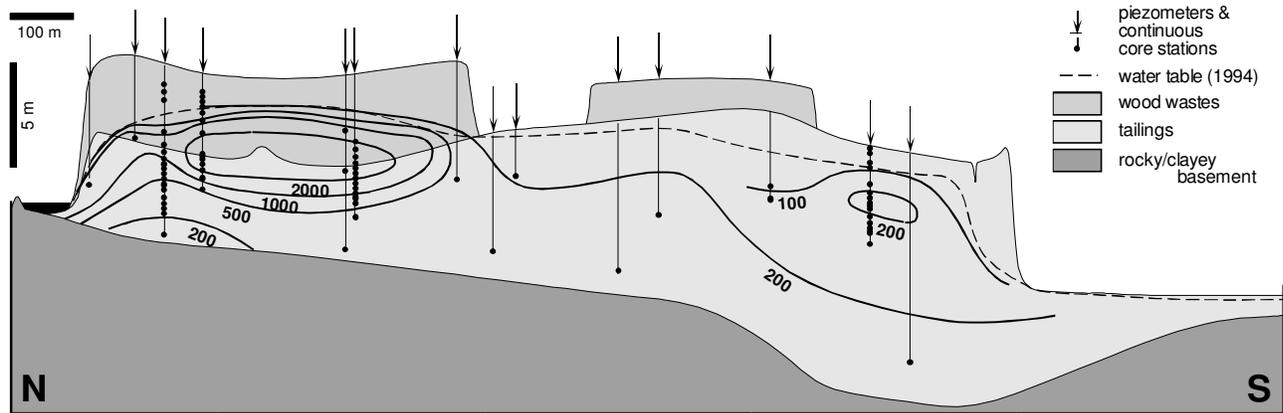


Figure 7. Distribution of alkalinity (in mg/L-CaCO₃) along a N-S section across the tailings pile in 1994 (from Tassé *et al.* 1997).

5. COVER PLACEMENT AND REDUCTION OF AMD

The interruption of sulphide weathering, either by an organic barrier or by some other oxygen-proof cover, does not mean an immediate cessation of AMD. As previously stated, acidity is stored in groundwater as Fe²⁺, and acid-prone waters generated prior cover placement must first be flushed out from the tailings. Acidity/alkalinity balance allows some predictions about the pH of a given pore water that will be eventually discharged around the impoundment (Figure 8). The problematic samples are those for which alkalinity will be totally exhausted, at a ratio of 1.8 mg/L-CaCO₃ for each

1 mg/L Fe²⁺. Samples collected under the oldest parts of the cover (pre-1992, north sector) generally present no threat, with high alkalinity and low Fe²⁺. Elsewhere, most acid prone samples are located next to the dikes, including former dikes, now enclosed in finer tailings. They can also be located in more central parts, in tailings directly poured out from the mouth of carrying pipes. These locations correspond to zones of high hydraulic conductivity, where O₂ diffusion favours or has favoured high rates of alteration. Alteration and acid generation is also enhanced near margins, because the vadose zones are deeper than within the impoundment, allowing deeper weathering.

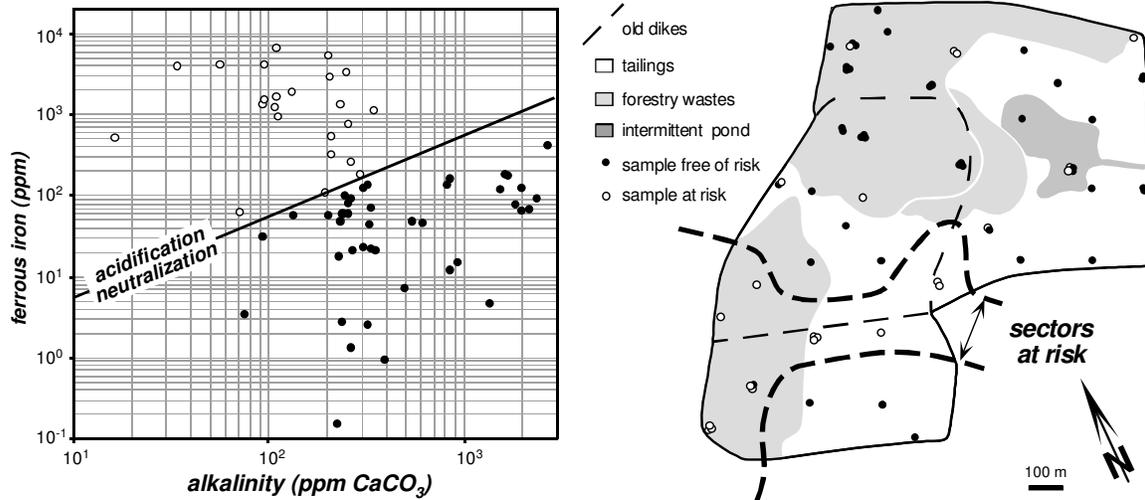


Figure 8. Ferrous iron versus alkalinity diagram for samples from the saturated zone.

Given the importance of transport time and length in neutralization and gain of alkalinity, groundwater flow was modeled in order to estimate the flushing time of acid-prone waters, *i.e.* the time required to evacuate Fe^{2+} derived from sulphide oxidation. The results of the simulations are presented in Tassé *et al.* (2004).

The particle tracking simulations show that flow is relatively rapid within the forestry wastes, above the interface with the mill tailings. It is much slower in the tailings, except in the southern part, *i.e.*, the zone containing, overall, the most contaminated groundwater. Flushing times are expected to be in the order of 5 to 10 years, for the internal parts of that sector, and a matter of a few months, next to the margins. Elsewhere, samples identified as "at risk" are more isolated, and unrelated to zones of high hydraulic conductivity. The transport distances and times for these waters, therefore, could be long, with the possibility that they may acquire additional alkalinity through interactions with the mineral medium, and become potentially inoffensive, *i.e.* able to keep near neutral pH upon seepage oxidation.

6. TREATMENT OF AMD

The geochemical environment below the wood wastes cover is reducing and alkaline (Figure 7). In organic-rich anoxic environments, specialized bacteria can live and fulfil their energy needs by mediating redox reactions between organic and inorganic compounds. Manganese oxide, nitrate, iron oxide and sulphate are reduced, while organic compounds are oxidized. Reduction occurs in that order, according to the Gibbs free energy involved in the reactions, in environment allowing for segregation of bacterial communities (*e.g.* deep sea; Froelich *et al.* 1979). If SO_4 and sulphate reducing bacteria (SRB) are available, and if the environment is reducing enough to allow H_2S production, metals such as Fe, Ni, Cu, Co, Zn,

Cd, *etc.* are precipitated as insoluble sulphide minerals. At East Sullivan, the breakdown of the forestry wastes provides organic compounds easy to metabolise. The "acid", in "AMD", is sulphuric acid, and SO_4^{2-} can be involved in the above redox reaction. Actually, the rotten eggs smell of H_2S can be noted at some sampling sites, pointing that this reaction is going on.

Tassé *et al.* (1997) believed that the alkaline and reducing milieu underlying the wood waste could be put at work for the site rehabilitation, simply by driving the acid residual waters through it. A side effect of recirculation would be the increase water infiltration, raising the watertable, and allowing a faster purge of acid-prone groundwater generated prior to organic cover placement. SRB are used in many varieties of bioreactors, from constructed wetlands (Wildeman *et al.* 1992) to reactive walls (Benner *et al.* 2000). Conceptually, a wood waste cover allowing prevention and treatment of AMD has several advantages over these "classic" alternatives:

- the system is a priori less sensitive to the hydrogeologic and vegetative cycles that affect purifying wetlands, because anaerobic conditions in an oversized organic cover can persist despite strong irregularities in influent discharge and climatic conditions;
- the insulating capacity of the forestry wastes keeps the temperature almost constant and above the freezing point at shallow depths all year round, providing an effectiveness over an extended season, a factor of importance in a nordic region;
- the extent of the cover is directly proportional to the volume of tailings to be covered, so that the volume of "reagents" poses no problem of space management;
- contact time can be long relative to a constructed wetland, and, despite increased infiltration, levels of supersaturation are easily attained.

Nevertheless, some uncertainties had to be faced. A reactive cover is a bioreactor, and as such, will perform well as long as its bacterial flora is fed with the proper

reactive material, in a suitable geochemical environment. The feed is less a concern. The carbon reservoir can be replenished if showing any weakness on the long term. However, the impact of the acid pH on the bacteria and the organic quality of the effluents were more worrying:

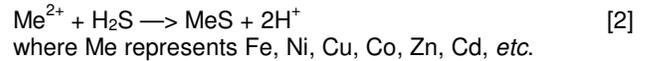
- anaerobic and aerobic oxidation that contribute to establish an adequate environment for SRB are controlled by bacteria; their collective resistance to acid pHs could not be defined; nevertheless, SRB are most functional at pH's between 5 and 9, but can tolerate pH's as low as 2.6 (Bolis *et al.* 1991, in Eger and Wagner, 1995); actually, SRB could overcome pH 1.6 conditions and rise the pH to 6 in closed vessel experiments with tree barks and artificial AMD (Tassé and Germain, 2002);
- organic leachates can be associated with the presence of undesirable substances, such as phenols or chelating complexes; actually, phenols and phytotoxic substances are released by humification and composting of forestry wastes, but significant attenuation occurs within only a few weeks, if the mass of organic waste is not fed with new reactive products (Solbraa *et al.* 1983).

7. RECIRCULATION

The climate normals in the region from 1971 to 2000 are precipitation of 0.91 m, and temperature daily maximum and minimum of 7.0 and -4.7°C.

A forestry-waste cover can be exploited for the treatment of acid prone groundwater flushed after cover placement. Its essential geochemical characteristics are similar as passive treatment systems such as constructed wetlands and reducing barriers, in which redox reactions improve the effluent quality (*e.g.* Wildeman *et al.* 1992; Benner *et*

al. 2000). In those anoxic environments, sulphate-reducing bacteria, in the absence of molecular oxygen, secure their energy needs by oxidizing organic compounds, using the sulphur of sulphate as the terminal electron acceptor (Eq. 1). Metals are precipitated by reduced sulphur (Eq. 2).



As mentioned above, the characteristic H₂S smell of reduced sulphur was noted at some observation wells, which is clear evidence that sulphate reduction is going on. Alkalinity, below the wood-waste cover, reaches considerable values, up to 2000-2500 mg/L-CaCO₃ (Figure 7).

These alkaline and reducing properties observed below the wood waste cover at East Sullivan are essentially those of a constructed wetland. In the same way, reduction of sulphate to sulphide in these geochemical conditions can provide the counter-ions required for the precipitation of iron and other metal as sulphides. Laboratory tests were performed to determine the efficiency of various types of barks fragments (Tassé and Germain, 2002). The results indicate sulphate reduction occurs more readily in a media with deciduous and coniferous barks than with coniferous bark alone. Nonetheless, both types of media do favour sulphate reduction but it is only the residence time that differs. Considering these results, the treatment system was done simply by recirculating the acid effluents collected around the impoundment through the wood waste cover.

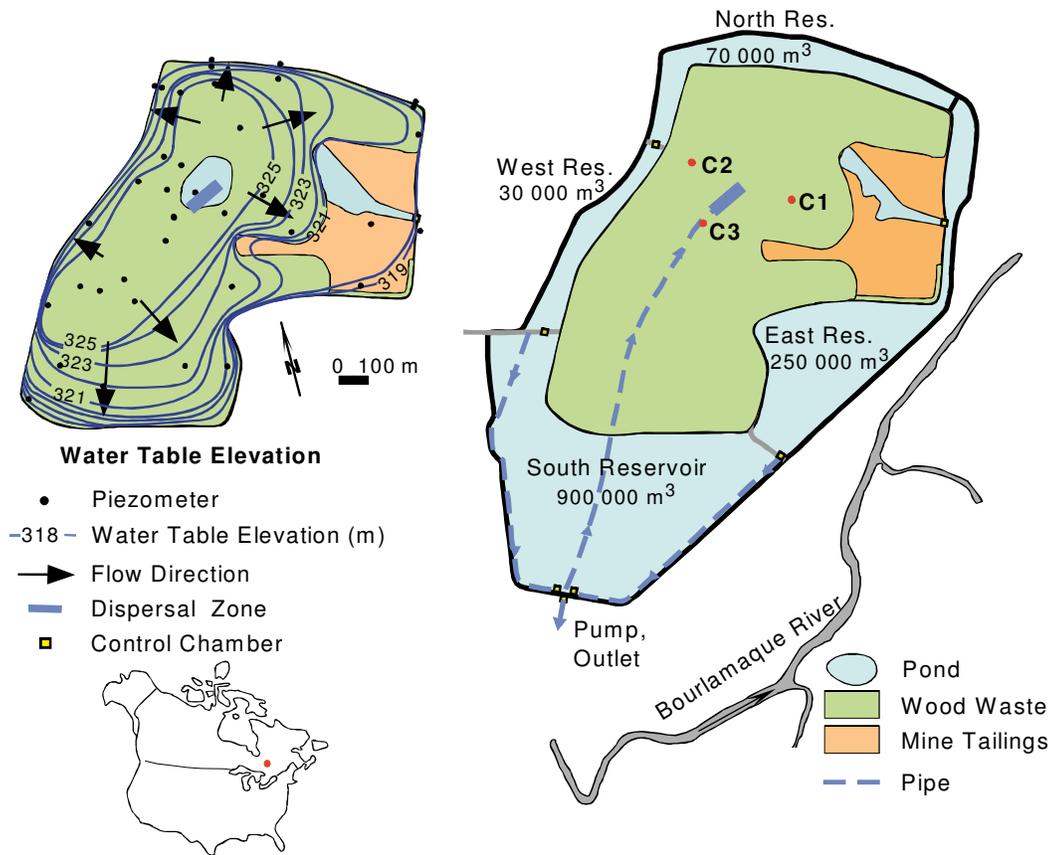


Figure 9. The East Sullivan tailings impoundment with the extent of the organic cover, the watertable elevation when the recirculation is in operation, and locations of observation wells and dispersal zone.

8. COLLECTION AND RECIRCULATION SYSTEMS

The collection system of the effluents, constructed in 1998 and 1999, consists of four reservoirs: North (70 000 m³), West (30 000 m³), East (250 000 m³), and South (900 000 m³) (Figure 9). The recharge rates, in m³/d, are, for North: 1 170; West: 430; East: 6 600. The recharge rate of the South reservoir could not be calculated since it was either pumped or open to the natural environment. The high rate for the East reservoir reflects the fact that it drains most of the impoundment (see flow directions, Figure 9).

The water from the North reservoir flows into the West reservoir. Pipes are connected to the outlets of the West and East reservoirs and water is directed by gravity either to a pumping station or to the outlet at the southern end of the site. There, water from either West, East or South reservoirs can be pumped back over the organic cover, or is allowed to flow by gravity into the natural environment (Figure 9). The dispersal zone, 120 m long perforated pipe, is located over the oldest part of the cover (mid-eighties). The recirculation is in operation from the beginning of May to the end of

October. Table 2 indicates the mean pumping rate and the volume of water treated from 1998 to 2005.

In 2006, the recirculation was no longer required since the alkalinity present in the North, West and South reservoirs was higher than the acidity present in the East reservoir. Therefore, since 2006, the acid water from the East reservoir is discharged by gravity into the South reservoir, where the acid water is neutralised.

Table 2. Average pumping rate and total volume of water treated from 1998 to 2005.

Year	Mean pumping rate (m ³ /d)	Volume (m ³)
1998	2 500	420 000
1999	3 750	630 000
2000	5 390	650 110
2001	5 928	741 530
2002	5 760	798 555
2003	5 060	724 783
2004	6 095	975 363
2005	4 617	572 526

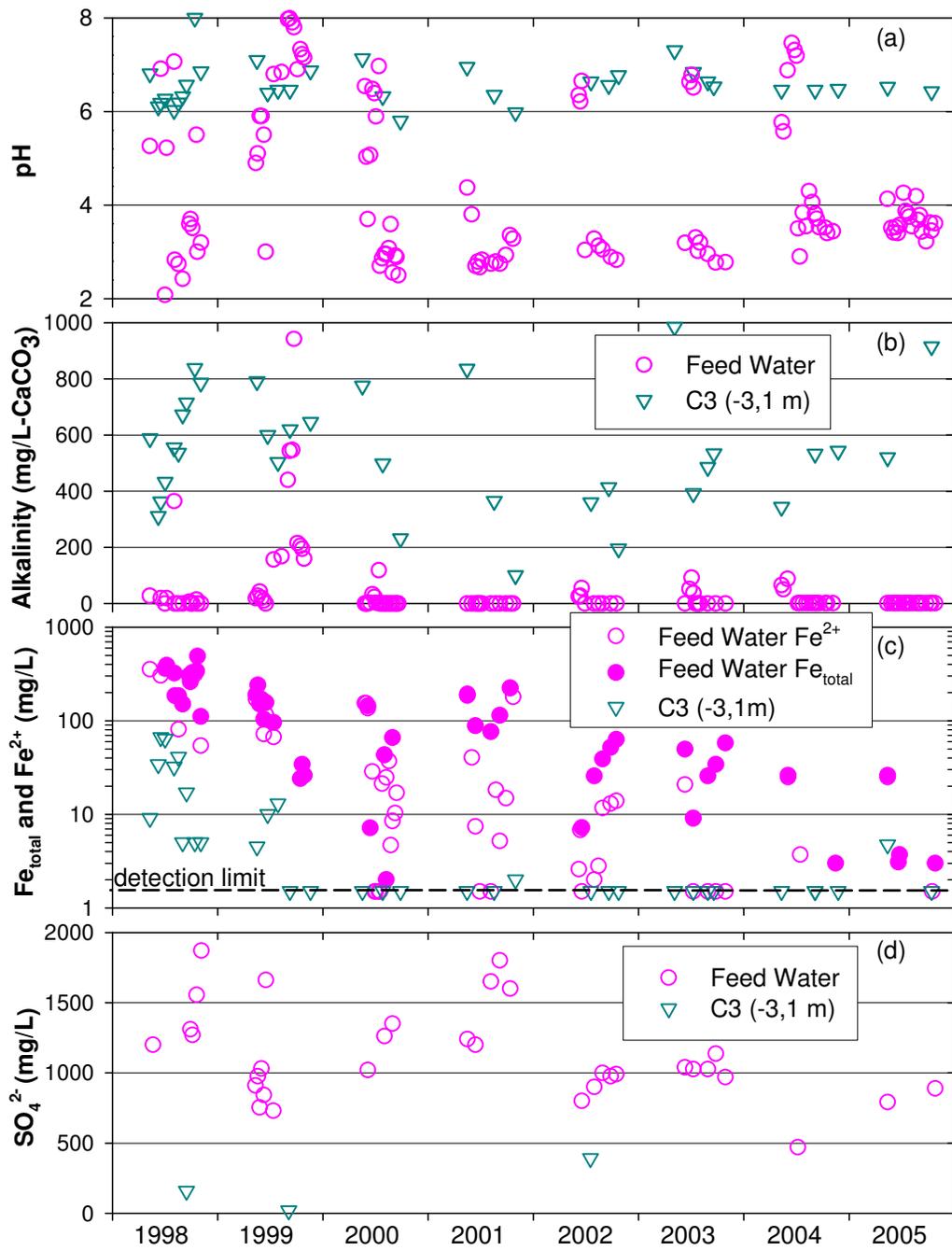


Figure 10. Feed-waters and groundwater quality at station C3: (a) pH , (b) alkalinity, (c) Fe_{total} and Fe²⁺, and (d) sulphates.

8.1 Water Quality at the Dispersal Zone

pH, alkalinity, total iron, ferrous iron (Fe^{2+}), and sulphates concentrations in feed-waters are shown in Figure 10. Since 2000, the first reservoir being pumped at the beginning of the recirculation period, starting in May, is the East reservoir until its quality meets the criteria shown in Table 3, thereafter its water is released to the natural environment, and only then the South reservoir is pumped until the end of October. Since the beginning of the water recirculation, the Fe_{total} varies between 500 and 2 mg/L. During the last five recirculation periods, pH was below 3.5 most of the time.

Table 3. Main water quality criteria.

Inorganic parameters	Mean Conc. (mg/L)	Max. Conc. (mg/L)
Cu	0.3	0.6
Ni	0.5	1.0
Pb	0.2	0.4
Zn	0.5	1.0
Fe_{total}	3.0	6.0
pH between 6.5 and 9.5		
Organic parameters		
Biological oxygen demand		50
Phenols		0.05

8.2 Efficiency of the Water-Treatment System

The efficiency of the biofilter can be assessed by comparing feed-water descriptors to those of groundwater collected in a nearby observation well, C3-p4. It is located next to the dispersal zone at 3.1 m depth (Figure 9).

From 1998 to 2005, the pore waters quality at observation well C3-p4 also improved over time with respect to ferrous iron (Figure 10). More specifically, the average Fe^{2+} concentration decreased from 28.2 mg/L in 1998 to less than 2 mg/L from 2000 to 2005, except in May 12, 2005 (5 mg/L). The fraction of ferrous iron removed increased progressively from 86% to 97% from 1998 to 2001 (Table 4). The increased efficiency of the biofilter over time is likely due to the establishment of bacteria colonies responsible for sulphate reduction.

pH was always near neutral in the six years surveyed, despite the more than often acid feed-waters (Figure 10). This is because pH and alkalinity are two interrelated parameters: if an acid is added to an alkaline solution, pH is preserved while alkalinity is consumed. In 2001 and 2002, we calculated an average consumption of -4.25 mg/L- CaCO_3 per day at C3-p4 over the recirculation period (Germain *et al.*, 2003). Despite these drops, alkalinity went back to 800 mg/L the following spring. Unfortunately, no data were gathered at this station in May 2002. In 2003, the

organic cover was completed in the vicinity of the observation well C3-p4, which decreased significantly the vertical hydraulic gradient. Thus, the flow rate migrating through this well is smaller now, which explains the lower decrease in alkalinity (down to 400 mg/L- CaCO_3) despite acid feed-waters. In 2004, the low alkalinity value was significantly less (343 mg/L) than that observed in 2005 (520 mg/L), since the volume of acid water being treated in 2004 represents 70% more than the ones of 2005 (see Table 2). The re-establishment of alkalinity during the non-recirculation period is mainly due to in situ alkalinity generated by on-going sulphate reduction (Eq. 1) and somewhat to alkalinity associated with the alteration of organic components.

Table 4. Average Fe^{2+} in feed-waters and at observation well C3-p4, and its corresponding percentage removed from 1998 to 2005.

Year	Average Fe^{2+} (mg/L)		% removed
	Feed-waters	C3-p4	
1998	201.8	28.2	86
1999	93.3	5.9	94
2000	39.1	<1.5	>96
2001	47.3	1.6	>97
2002	3.6	<1.5	>58
2003	3.3	<1.5	>55
2004	n.a.	<1.5	
2005	25	3.25	87

9. IMPACT ON GROUNDWATER QUALITY

pH, alkalinity and Fe^{2+} have been monitored since 1998 in the groundwater. Here, Fe^{2+} is the key parameter for water quality, since its oxidation and hydrolysis are responsible for the acid mine drainage. Despite the recirculation of acid and ferrous waters, none of these fronts was developed under the dispersal zone at any time. In fact, the pH of all groundwater samples underlying the zone varies between 5.0 and 8.0 for the entire 1998-2005 database, which suggests efficient H^+ neutralization by local alkalinity and/or adsorption by colloidal and particulate wood waste. There is more. Fe^{2+} is less than 3 mg/L since 2000 in all eight near-surface observation wells in the vicinity of the dispersal zone, except once at C3-p4 in 2005/05/12 (5 mg/L). Since feed-water with sometimes over 100 mg/L- Fe^{2+} was pumped directly over the area, it can be concluded that Fe^{2+} attenuation did occur and that the recirculation did not compromise the groundwater quality.

On the contrary, the recirculation has allowed a more rapid flushing of Fe^{2+} rich pore waters, as shown at observation wells C1 and C2 (Figure 11). These stations are in the vicinity of the dispersal zone, over the old cover (see Figure 9). Two flushing effects are observed: the natural one by the increase recharge due to the coarse fragments of the cover and the imposed one by the recirculation. The natural flushing is

observed at these wells, with a decrease of yearly average Fe^{2+} concentrations (Figure 11). That decline starts over the first years of the available database, in 1992. The effects of water treatment and flushing are noted at C1 in 2001 and at C2 in 2005. The flushing delay observed at C2 is due to a deeper location (8.45 vs 4.87 m) and a lower hydraulic conductivity ($2.0\text{E-}7$ vs $7.5\text{E-}7$ m/s).

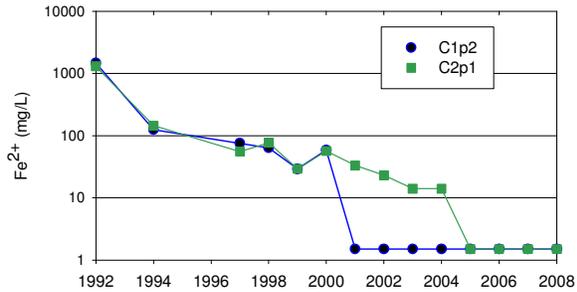


Figure 11. Evolution of Fe^{2+} at observation wells located in tailings at C1 and C2 from 1992 to 2008 (see Figure 9 for location).

10. IMPACT ON SURFACE WATER QUALITY

Data on water composition of each reservoir were gathered from May to November, each year since their completion (North: 1994; East: 1997; West: 1994; South: 1998).

10.1 Fe Mass in Each Reservoir

Figure 12 shows the evolution of the Fe mass present in each reservoir in the spring from 1998 to 2008. When the water treatment was on, the sampling was taken prior to the beginning of the recirculation, *i.e.* from 1998 to 2005. The Fe mass in the North reservoir is the lowest since it is adjacent to the oldest section of the cover. Therefore, the natural flushing of the pore water had occurred. Since 1998, its quality complies with the target values (Table 3) without any treatment.

The West reservoir drains a small section of the impoundment but made of highly permeable porous media, mainly dikes (Figure 9). These pore waters were characterised with high Fe_{total} concentrations: 540 mg/L in 1994. With the recirculation, the watertable rises and increases the hydraulic gradient, which accelerates the flushing of these altered pore waters. Thus, despite a more recent cover in comparison to the north section, its waters also do not require any treatment since 2001.

The impact of flushing altered pore waters due to the recirculation is also observed in the East Reservoir, in particular during the first year of the recirculation. Also, additional forest wastes were deposited along the dikes next to this reservoir allowing a greater natural infiltration. These combined flushing mechanisms have allowed the steepest Fe mass drop observed within a

year (79%). In 2007 and 2008, the mass of iron has increased due to additional forest wastes were deposited along the dikes.

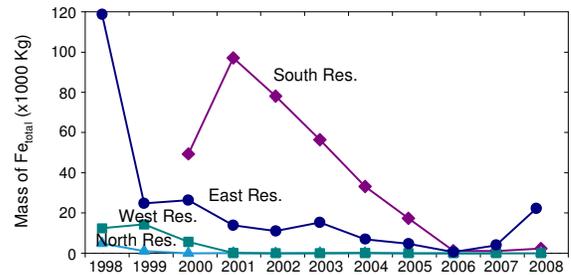


Figure 12. Evolution of the Fe mass in the reservoirs in the spring from 1998 to 2008.

The impacts of the recirculation and the increase natural infiltration are particularly well illustrated by the evolution of the Fe mass in the South reservoir, where it has decreased by more than 65%, at a constant rate of $-19\ 193$ Kg/a from 2001 to 2006. As for the North and West Reservoirs, its water quality also complies with the target values (Table 3) without any treatment. In fact, since 2006, the mass balance between alkalinity and acidity in the reservoirs (Figure 13) indicates that the alkalinity present can counterpart the acidity. Therefore, since 2006, the acid water of the East reservoir is neutralised directly with that of the South reservoir, which also collect by gravity the water from the West and North reservoirs.

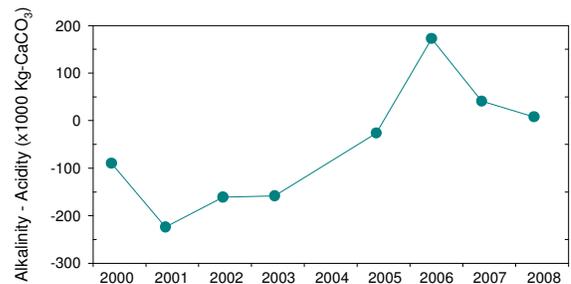


Figure 13. Mass balance between alkalinity and acidity (Kg-CaCO₃) in the four reservoirs from 2000 to 2008.

The tailings adjacent to the South reservoir are fine-grained on the east side but coarse-grained on the west side. Iron concentrations in pore waters are low on the east side, except along dikes. However, coarse material on the west side did favour sulphide oxidation and high Fe^{2+} in the pore waters, by maintaining an important vadose zone, not only in the dikes' vicinity but also deep into the impoundment. Moreover, the Fe^{2+} rich plume is migrating southward and is discharging in the South reservoir (see Figure 2, Germain *et al.*, 2003). Nonetheless, the plume extent has considerably decreased; the concentration at the

heart of the dissolved iron plume has constantly decreased passing from a peak at 18 767 mg/L in 1998 to 2 857 mg/L in 2008. The decrease concentration is also observed at observation wells 15E-18N, 13E-12N, and C19. We observed a small downward movement of the plume since the concentration at observation well C20 p3 is slightly increasing since 2006 (Figure 14).

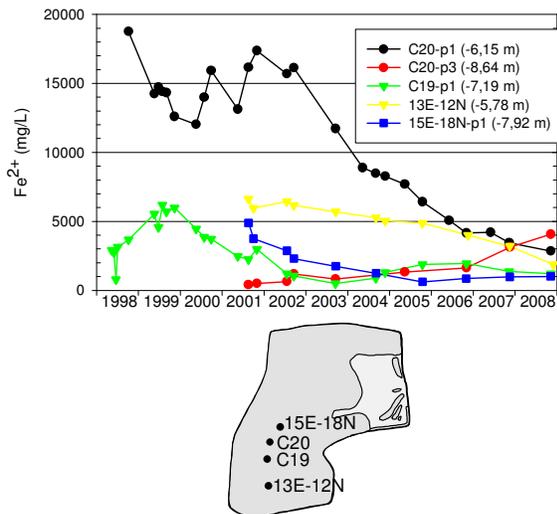


Figure 14. Evolution of dissolved Fe²⁺ in some observation wells located within the plume.

10.2 Detailed Surface Water Quality - East Reservoir
Only the surface water quality of the East Reservoir is presented in detail. This particular reservoir drains most of the groundwater of the impoundment; consequently during the recirculation period, after pumping its water for 3 to 4 weeks in the spring, its water quality was significantly improved and meets the target values. Now, this reservoir contains the worst water quality at this mine site, due to an uncovered section of the impoundment adjacent to it.

10.2.1 pH, Fe_{total}, Copper and Zinc

The presence of a pond in the north-east section of the impoundment since its closure testifies of the fine-grained nature of the tailings (Figure 9). Similar muddy sediments were accumulated next to the eastern dike, to the south. These relatively high levels of water saturation contributed to limit sulphide oxidation in

those areas, prior to cover placement. Significant alteration was limited to coarser-grained dikes and former dikes. It follows that, notwithstanding these dikes, pore waters showed initially relatively low dissolved iron. Cover placement stopped oxidation and also increased infiltration, flushing these relatively poor Fe_{total} pore waters into the reservoir.

Figure 15 shows the evolution of pH, Fe, Copper and Zinc of the East reservoir. pH varied between 3.3 and 8.5 between 1999 and 2008; in 2009 its lowest value was 5.2 indicating an improvement in its water quality in comparison to 2008. From 1998 to 2005, the pH increases over each recirculation period since a significant volume of treated water was discharged into it. When the recirculation was not further needed, the pH trend is reversed, it decreases. This pattern last from 2006 to 2008, partly because the mass of alkalinity discharging into the reservoir was less important then when the recirculation was active. Furthermore, additional organic waste was added over the dike adjacent to the East Reservoir, favouring infiltration and pore water flushing rich in dissolved iron.

The maximum Fe_{total} concentration, drops gradually from 108 mg/L in 2000 to 26 mg/L in 2005, representing a drop rate of -16 mg/L per year (Fig. 5, slope of pink line). This trend stops with the end of the recirculation in 2006. Furthermore, the pore water flushing related to additional organic waste in the vicinity of the dike is also contributing to alter the decrease trend of total iron concentration. During each recirculation period, the total iron decreases as significant treated water is discharged in the reservoir. In the fall, when the recirculation stopped, iron concentration goes back up.

With regard to metals (cadmium, chrome, copper, and lead), their concentrations have always been below 0.2 mg/L since 1999. While for zinc, its concentration is less than 2.2 mg/L in the spring and is generally less or near 0.5 mg/L in the summer and fall, from 1999 to 2008.

10.2.2 Organic parameters: DCO, DBO₅, and phenols

Figure 16 shows the evolution of the chemical and biological oxygen demands, and phenols, from 1999 to 2008. Both demands significantly decrease within the first two years; then the BOD₅ meets the target value of 50 mg/L for the remaining period. A similar behaviour is also observed for phenols

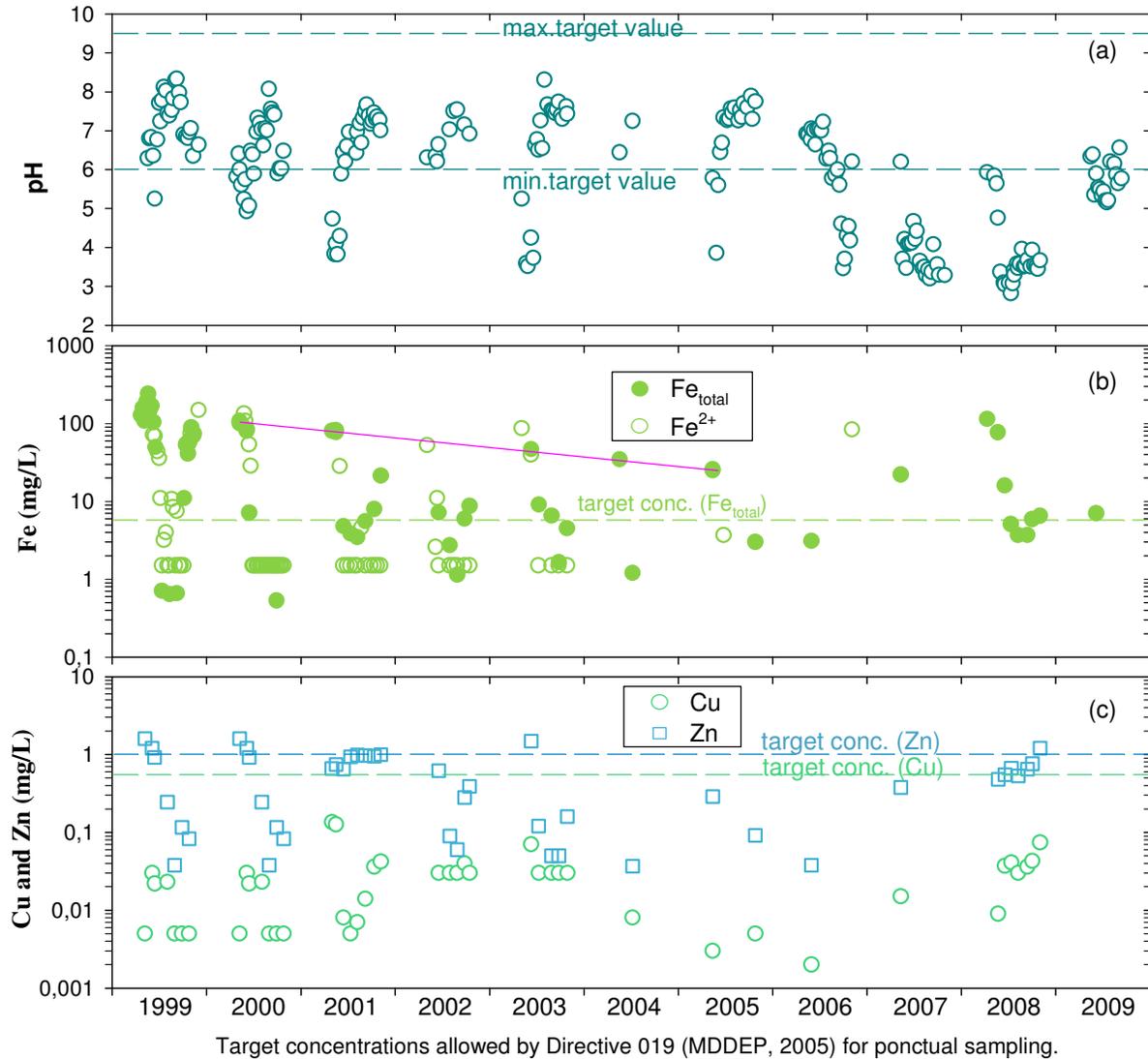


Figure 15. Evolution of (a) pH, (b) Fe_{total} and Fe^{2+} , and (c) copper and zinc in the East reservoir; the pink line represents the Fe-drop trend in the spring.

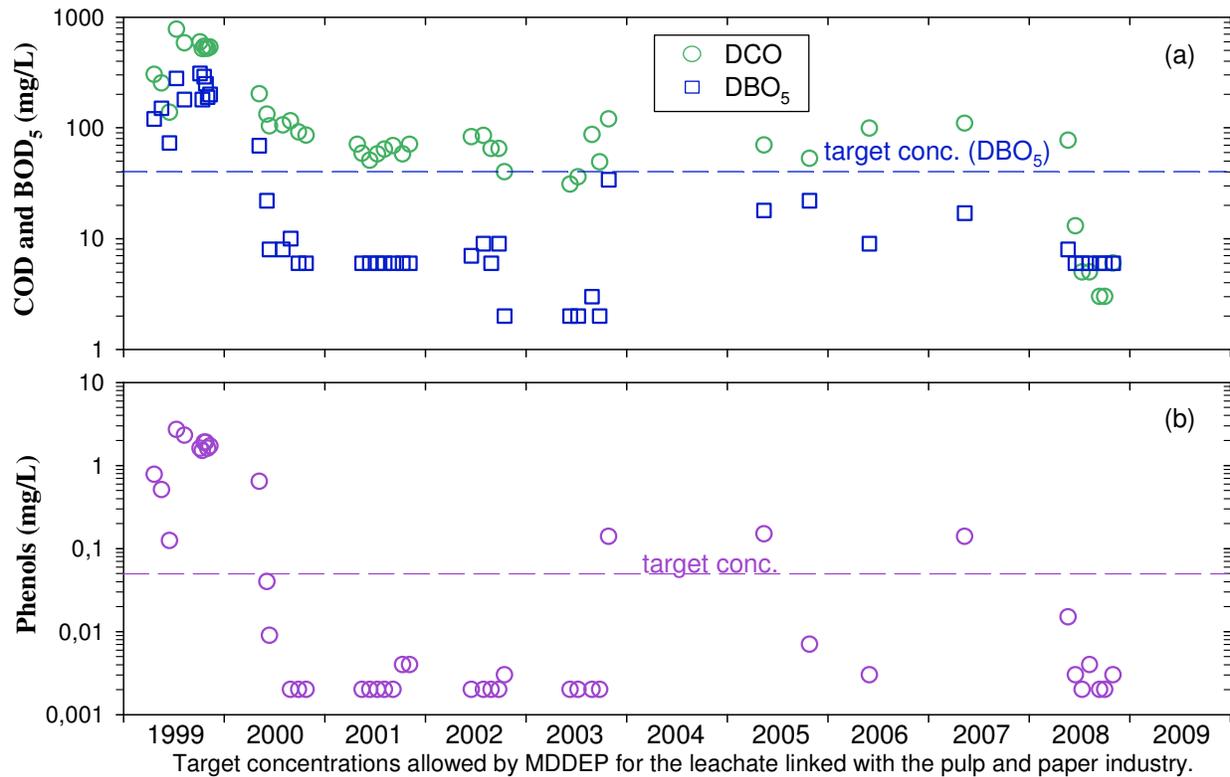


Figure 16. Evolution of (a) chemical and biological oxygen demands, (b) Phenols in the East reservoir.

11. CONCLUSIONS

A cover of forestry wastes features several qualities with respect with other types of cover: no disturbance of the natural environment (industrial waste rather than natural material); sturdy and simple to erect. Since the placement of such a cover may require several years, disposal should first target the margin of the impoundment, where contaminant production is at its maximum.

Besides stopping the atmospheric oxygen migration, such cover favours higher infiltration and a watertable rise, which plays the role of a "wet cover". This also allows a more rapid flushing of acidogenic groundwater. Furthermore, a wood-waste cover can fill the role of an alkaline reducing barrier for the treatment of these acidogenic waters, until a balance between acidity and alkalinity in the discharged water is reached. In 2006, the mass of alkalinity in the reservoirs was superior to that of acidity; consequently, the recirculation system was stopped. Since then, acid

waters from the East Reservoir are neutralised by the alkalinity present in the three remaining reservoirs.

The recirculation system holds several advantages over conventional constructed wetlands, which are driven by the same geochemical processes: it is a priori less sensitive to hydrogeologic and vegetative cycles that affect wetlands, because anaerobic conditions can persist despite large fluctuations in influent and climatic conditions; contact time can be long relative to a constructed wetland, and levels of over-saturation solution are likely to be attained and maintained.

Evolution of surface water quality suggests that the contaminated groundwater flush was completed in the north sector in 1999, west sector in 2001, south sector in 2006, and the east sector is near completion.

Since 1999, the water quality at the final effluent, located south of the pumping station, meets the requirements of the Ministry du Développement durable, de l'Environnement et des Parcs du Québec.

It is important to notice that such organic cover should not be applied over mine tailings impoundment containing mercury because of the potential of the formation of toxic compound such as methyl mercury.

It is clear that an organic cover not only can act as an oxygen barrier, but can also be integrated in a strategy for a highly efficient treatment system of transient acidogenic waters rich in metals.

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