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A UNIQUE RESEARCH PROGRAM in Québec

Passive treatment of highly contaminated iron-rich acid mine drainage

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Task Force-ASMR-ARRI Joint Symposium 2017, April 13, WV, USA

Outline

- Context: Fe-rich AMD
  - Occurrence
  - Passive treatment
- Case studies
  - I) Lorraine mine site: lab *vs* field testing
  - II) East Sullivan mine site: 14 y water quality evolution
- Concluding remarks

Mine sites rehabilitation

- Step 1: Control AMD generation
  - Limit the availability of one (or more) of the three main contributing factors (sulfides, oxygen & water), or control tailings temperature
  - Example of developed methods
    - **Oxygen barriers (case study I and II)**
    - Water infiltration barriers
    - Desulphurization
    - Thermal barriers

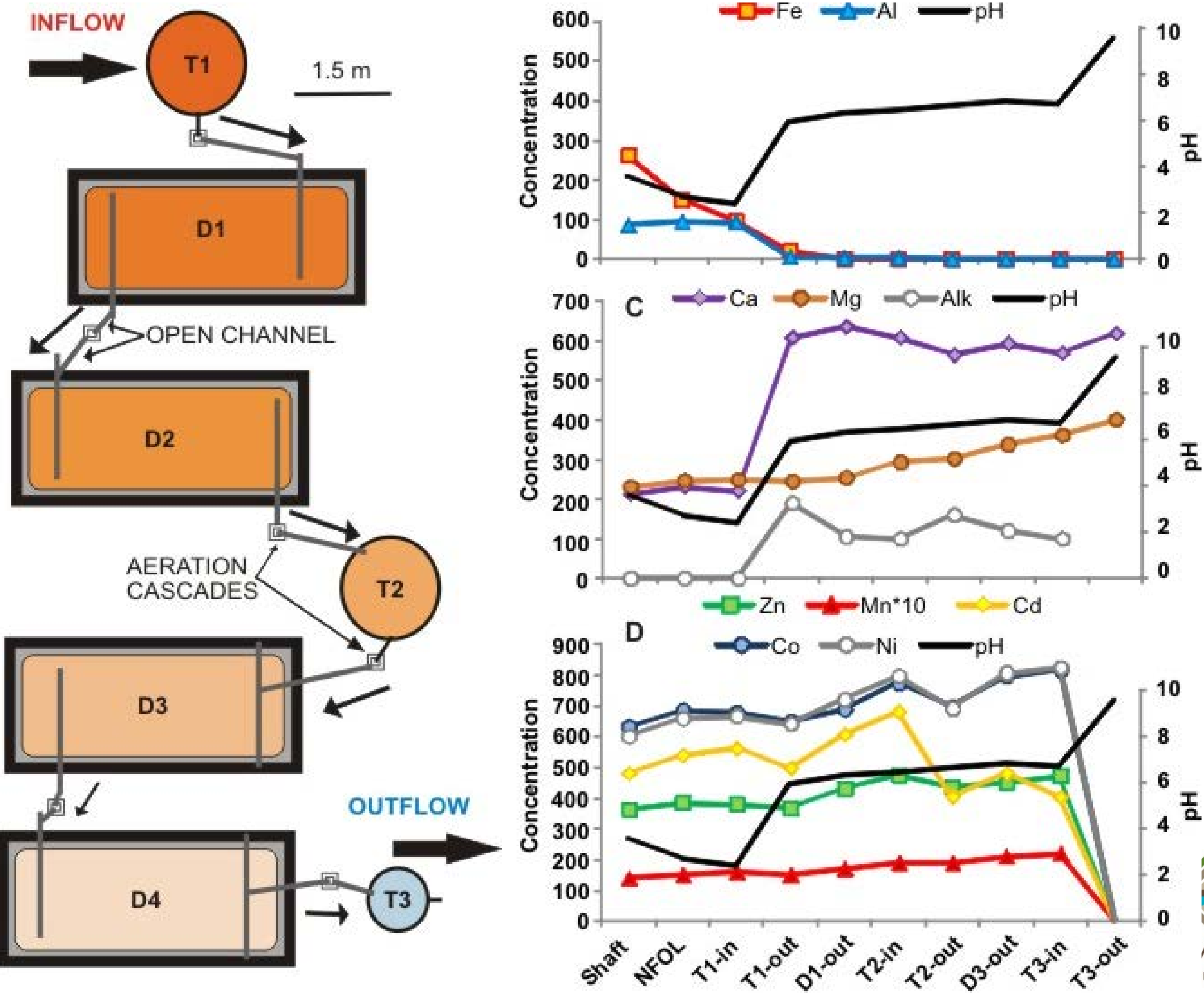
(Bussière and Aubertin, 2016)

Mine sites rehabilitation

- Step 2: Passive treatment of generated AMD
  - **Limestone/dolomite drains (DOL)**
    - pH and alkalinity increase, metals (and sulfate) precipitation
  - **Passive biochemical reactors (PBRs)**
    - Metals and sulfate removal
  - **Wetlands [(an)aerobic]**
    - Polishing of residual contaminants
  - **+ NEWER → Dispersed alkaline substrate (DAS) reactors:** mixtures of highly porous (wood chips) and alkaline (calcite, MgO) materials
    - Pre-treatment of high contamination loads

(Ayora et al., 2013; Genty, 2012)

Pilot-scale DAS reactors (T1-T3)



- **T1 & T2: calcite-DAS**
- **T3: MgO-DAS**

(Ayora et al., 2013)

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Examples of Fe-rich AMD

Comparison of **some of the most acidic waters and highest concentrations of metals** derived from tailings pore water, surface water, and underground mine workings (Moncur et al., 2005)

Parameter (g/L) (except pH)	**pH**	**Cu**	**Zn**	**Cd**	**As**	**Fe_t**	**SO_4^{2-}**
Sheridan tailings (pore water), MB, Canada	**0.67**	**1.6**	**55**	**0.1**	**0.05**	**129**	**280**
Heath Steele (tailings pore water), NB, Canada	**0.80**	**0.6**	**6**	**n/a**	**n/a**	**48**	**85**
Genna Luas (surface water), Sardinia, Italy	0.60	0.22	10.8	0.06	0.07	77	203
Iron Mountain (mine shafts/drifts), CA, USA	-3.6	4.76	23.5	0.21	0.34	141	760
Other sites (mine shafts/drifts/pore water)	0.67	468	50	0.04	22	57	209

Parameter (g/L) (except pH)	pH	Cu	Zn	Cd	As	Fe_t	SO_4^{2-}
Lorraine mine site, QC, Canada (Potvin, 2009)	**3.6**	**n/a**	**0.8**	**0.4**	**n/a**	**6.9**	**15**
East Sullivan mine site, QC, Canada (Germain et al., 1994)	**2**	**n/a**	**n/a**	**n/a**	**n/a**	**7**	**17**
*Carnoulès, France (Giloteaux et al., 2013)	1.2	n/a	n/a	n/a	12	20	29.6
Iberian Belt Pyrite, Spain (Macias et al., 2012)	3	0.005	0.44	n/a	n/a	0.3	3.6

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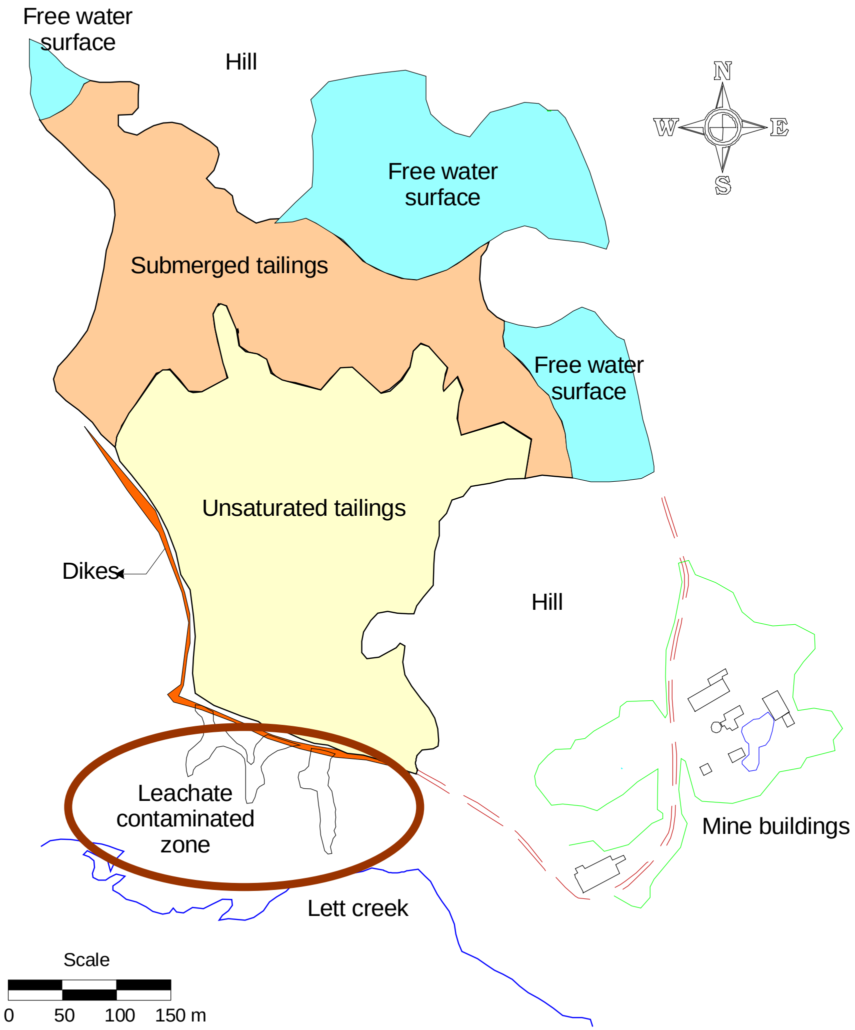
Case study I: Lorraine mine site

- Historic, Progressive Rehabilitation

1 Lorraine mine site: historic



1964-1968 : Cu, Au, Ag, Ni

acid-generating tailings: 15.5 ha (up to 6 m)

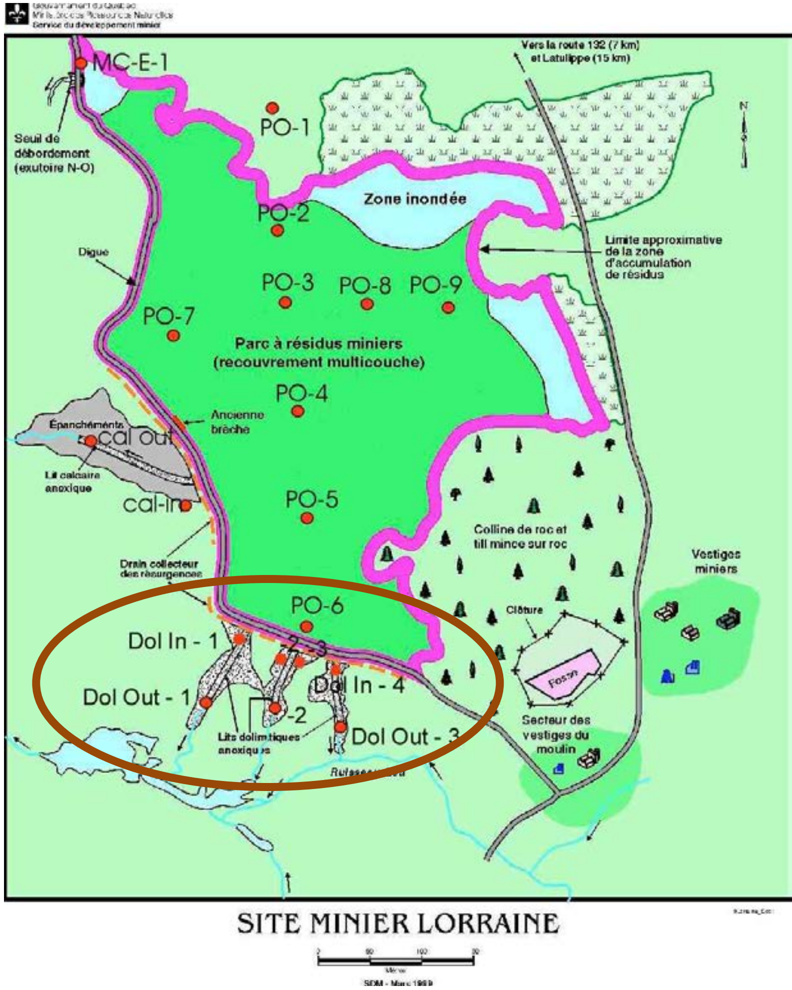
(Nastev & Aubertin, 2000)

Lorraine mine site: rehabilitation

- **Control AMD generation**
  - Multilayer cover
- **Passive treatment of Fe-rich AMD**
  - Phase I: dolomite and calcite drains (1999) - chemical
  - Phase II: 3-unit system (2011) - biochemical
  - Phase III: DAS reactors (?) - biochemical
- **Passive treatment of Fe-rich AMD: challenges**
  - Limited space, topography, high water table
  - Abundant precipitation, harsh winter (7-8 months)
  - <u>Lab testing required</u> prior to construction of a field system

Lorraine mine site: rehabilitation

- **1999**: CCBE (cover with capillary barrier effect = O_2 barrier): control AMD generation
- **1999**: 3 Dolomite drains (Dol-1 to Dol-3) and 1 calcite drain (Cal-1): passive treatment of Fe-rich AMD (**Phase I**)
  - **pH 3.6, 7 g/L Fe, 15 g/L sulfate**

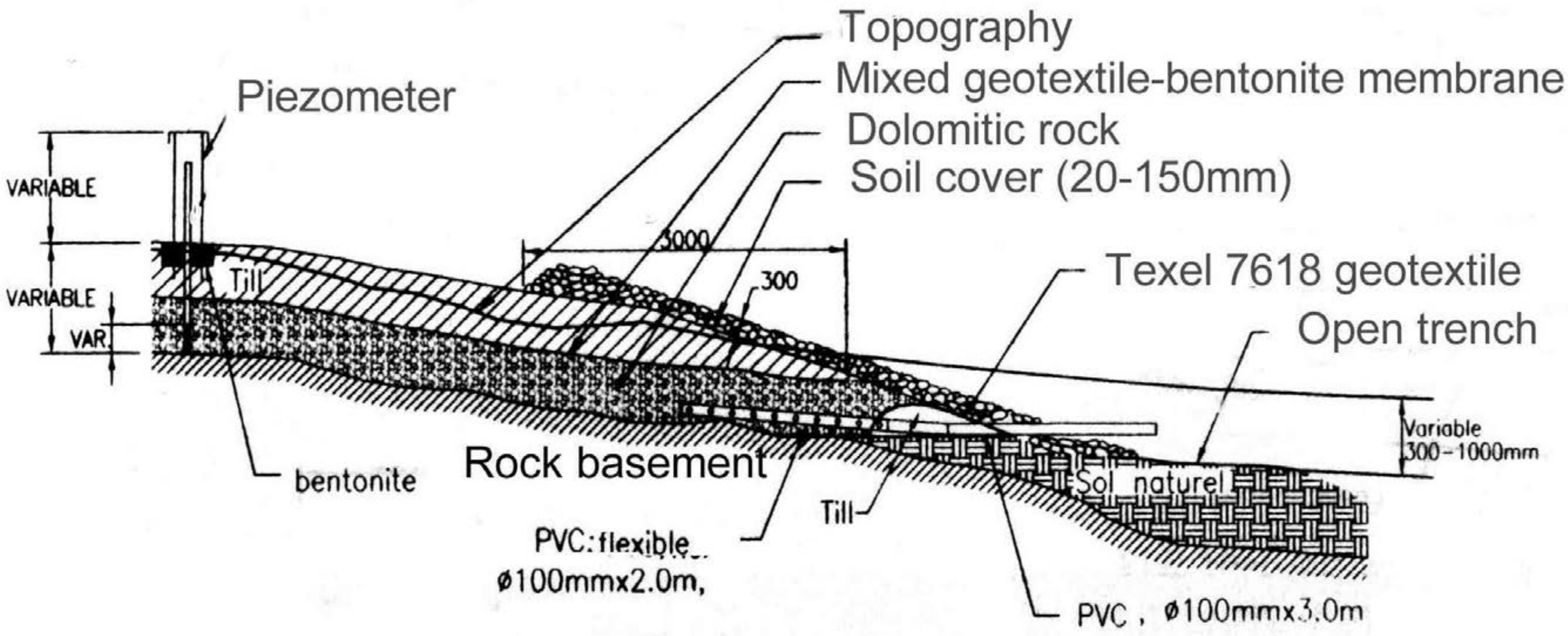


(Potvin, 2009)

1 Dolomite drains: design

Trenches filled with dolomite (70 %) (20-60mm)

- HRT (Dol-1 & Dol-2): 10 to 20 h



Scale 1 : 100 Adapted from Dessau-Soprin 1999

(Fontaine, 1999; Maqsoud et al., 2007)

Cal-1, Dol-1, and Dol-3

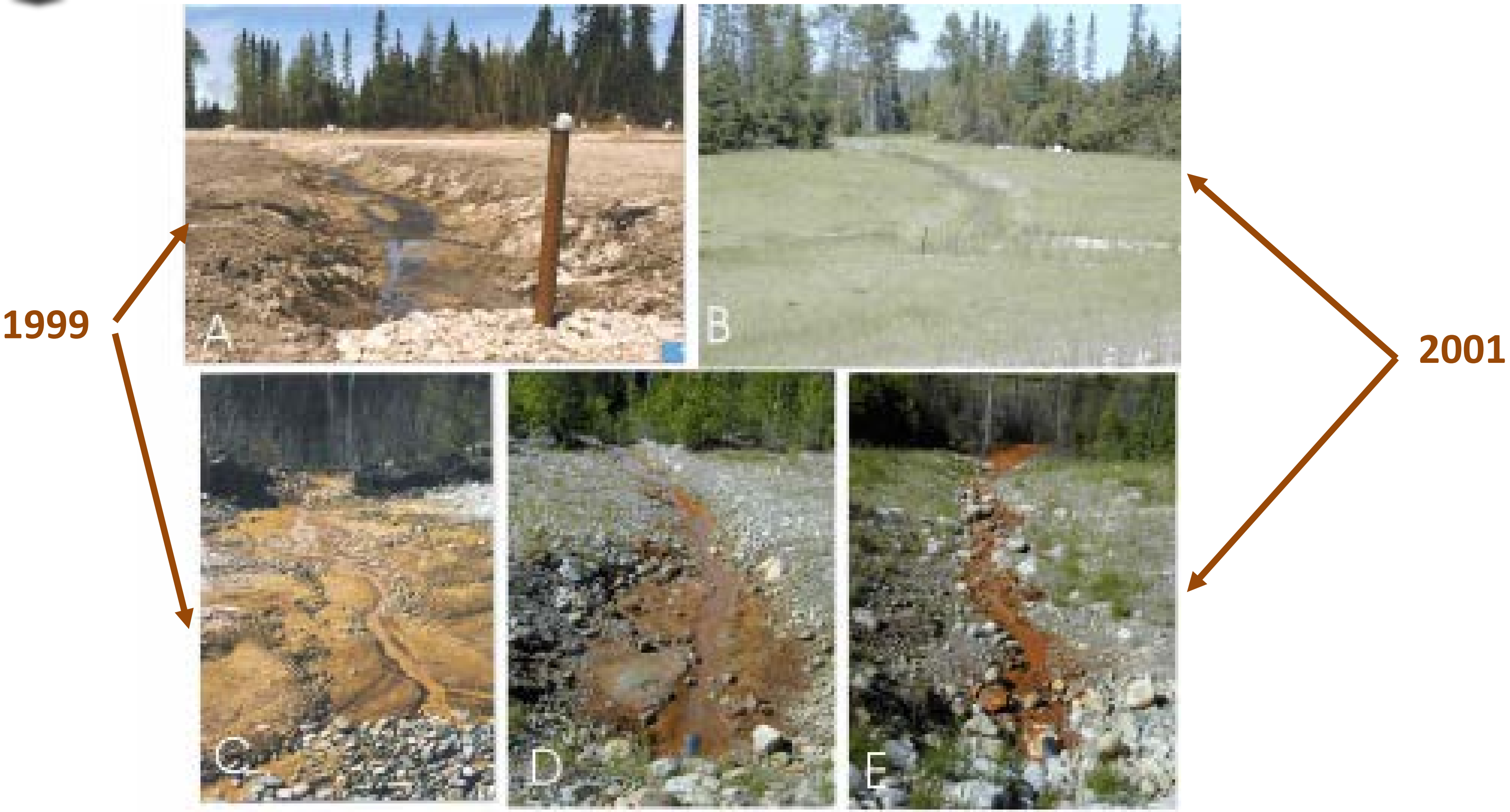
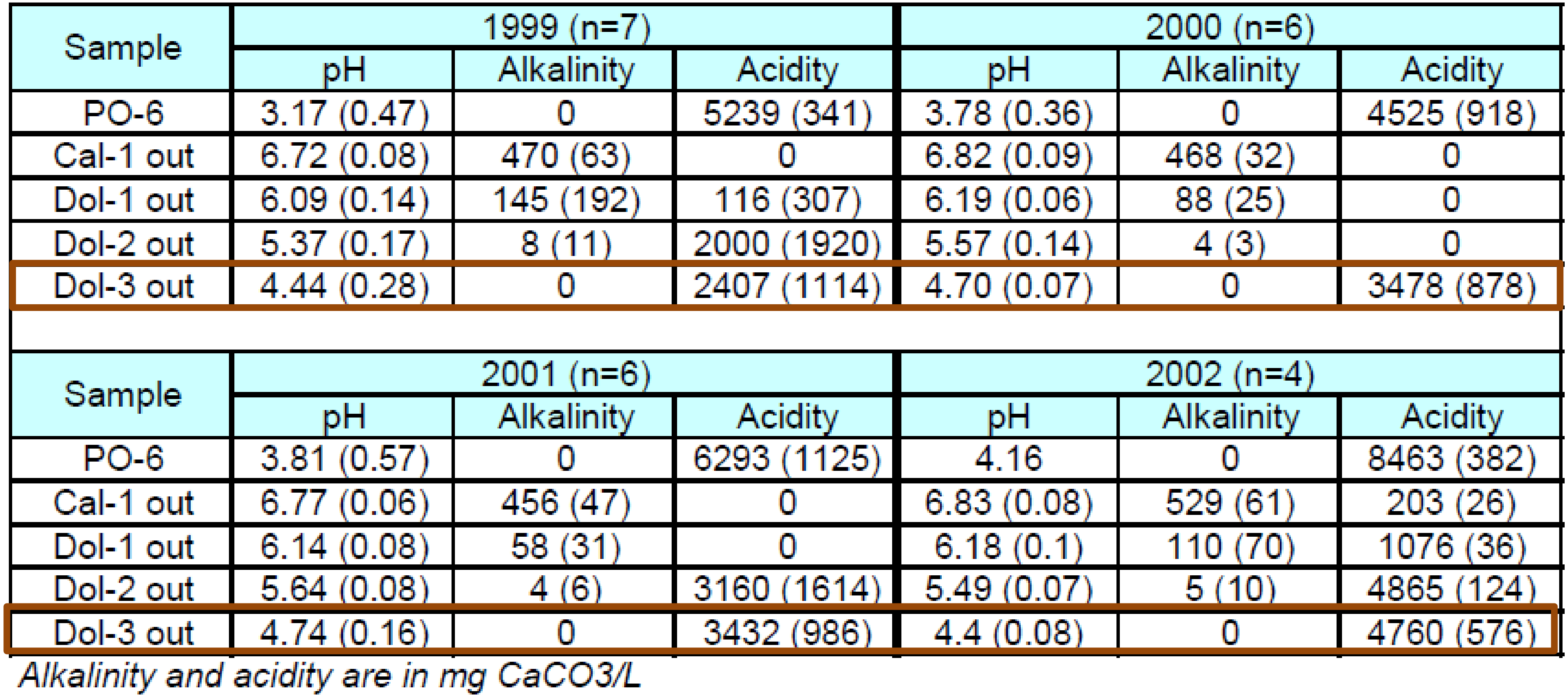


Figure 9 A) View of drain Cal-1 output, May 1999. B) Idem, but in June 2001. C) View of drain Dol-3 output , May 1999. D) Idem , but in June 2001. E View of drain Dol-1 output, June 2001. Notice the iron hydroxides precipitates in the trenches.

(Bernier et al., 2002)

Dolomite/calcite drains: 1999-2001

Tableau 3 Influent and effluent average pH, alkalinity and acidity.



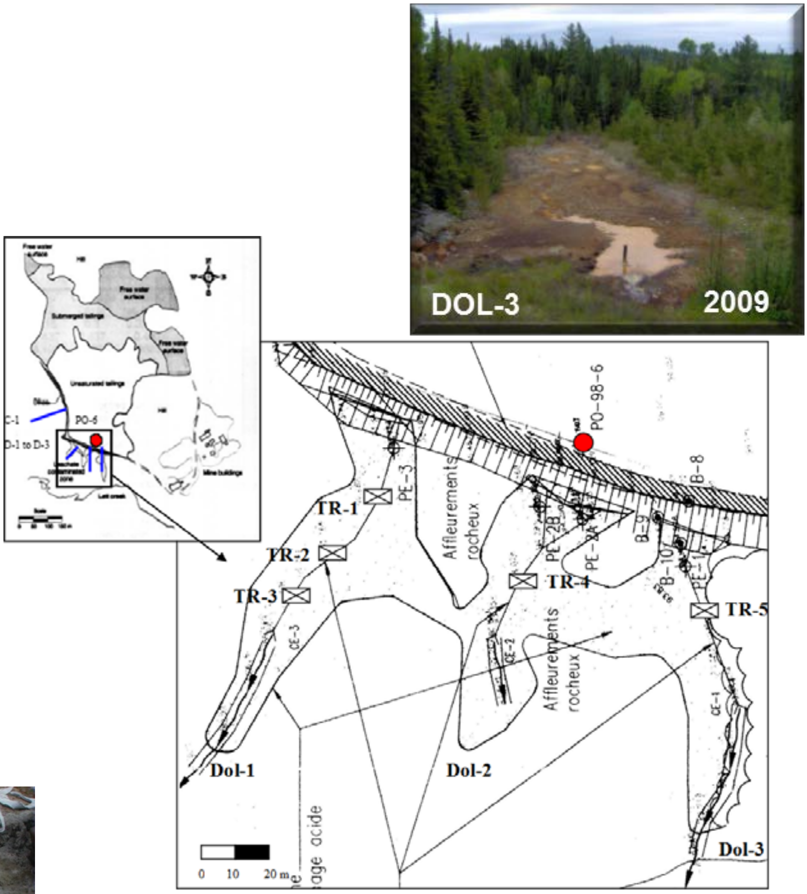
Sample	1999 (n=7)			2000 (n=6)		
	pH	Alkalinity	Acidity	pH	Alkalinity	Acidity
PO-6	3.17 (0.47)	0	5239 (341)	3.78 (0.36)	0	4525 (918)
Cal-1 out	6.72 (0.08)	470 (63)	0	6.82 (0.09)	468 (32)	0
Dol-1 out	6.09 (0.14)	145 (192)	116 (307)	6.19 (0.06)	88 (25)	0
Dol-2 out	5.37 (0.17)	8 (11)	2000 (1920)	5.57 (0.14)	4 (3)	0
Dol-3 out	4.44 (0.28)	0	2407 (1114)	4.70 (0.07)	0	3478 (878)

Sample	2001 (n=6)			2002 (n=4)		
	pH	Alkalinity	Acidity	pH	Alkalinity	Acidity
PO-6	3.81 (0.57)	0	6293 (1125)	4.16	0	8463 (382)
Cal-1 out	6.77 (0.06)	456 (47)	0	6.83 (0.08)	529 (61)	203 (26)
Dol-1 out	6.14 (0.08)	58 (31)	0	6.18 (0.1)	110 (70)	1076 (36)
Dol-2 out	5.64 (0.08)	4 (6)	3160 (1614)	5.49 (0.07)	5 (10)	4865 (124)
Dol-3 out	4.74 (0.16)	0	3432 (986)	4.4 (0.08)	0	4760 (576)

Alkalinity and acidity are in mg CaCO3/L

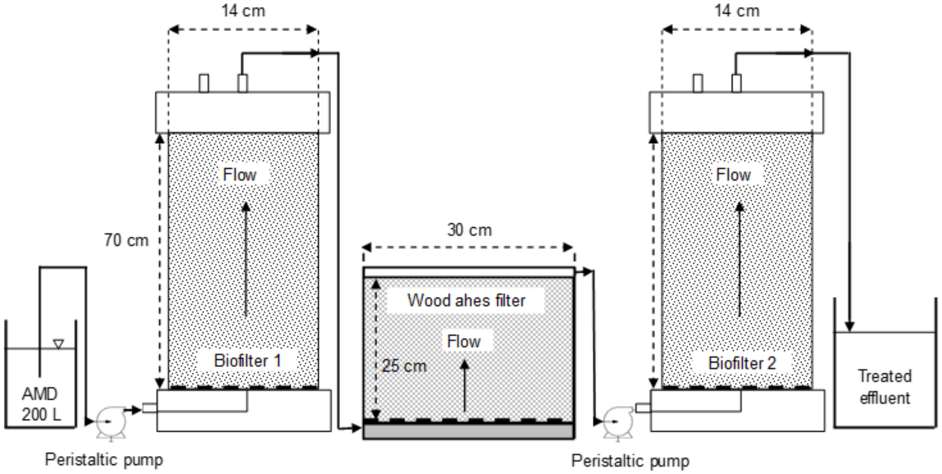
(Bernier et al., 2002)

1 Dol-3 (2009): clogged



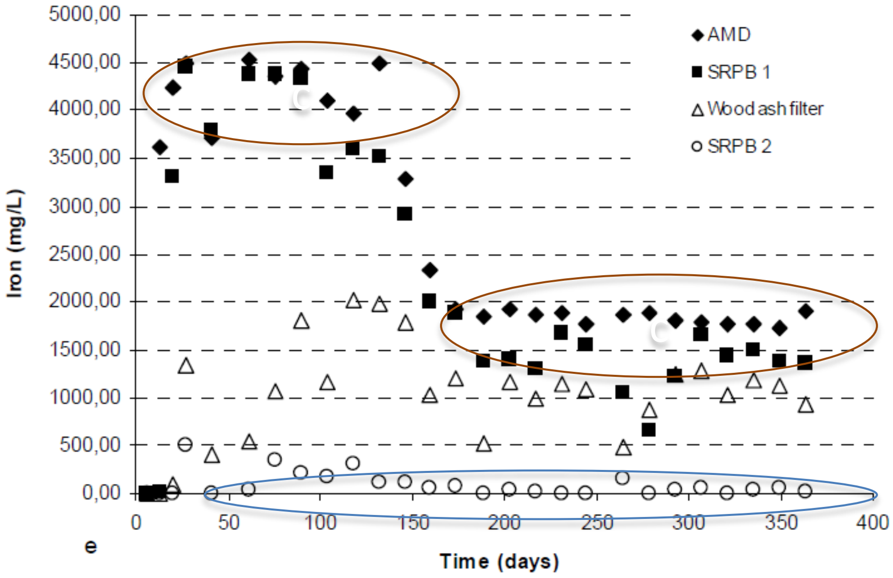
(Potvin, 2009)

1 Phase II: lab testing (6.7L to 2m³)



3-unit train lab system

- **Input Fe: 2-4 g/L**
- **Output Fe: < 1 mg/L**



(Genty, 2012)

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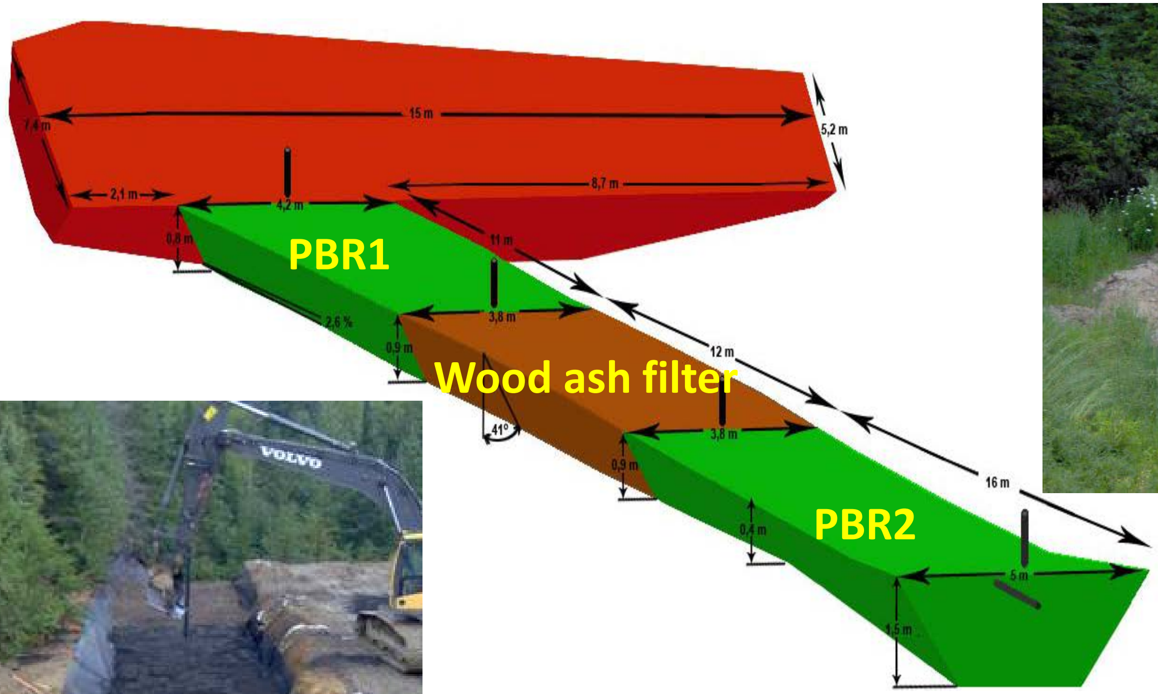
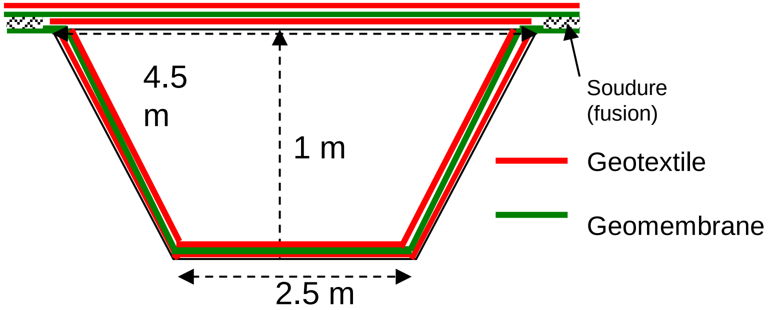
Field pilot construction: design





Components (% dw)	PBR1	PBR2
Wood chips	36	18
Manure	17	10
Compost	24	12
Sand	21	10
Calcite	2	50

(Genty, 2012)

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1 Field pilot construction: within 5 days


Before Dol-3 excavation


Dol-3 excavation


Material mixing


AMD drain collection

1 Field pilot construction: within 5 days

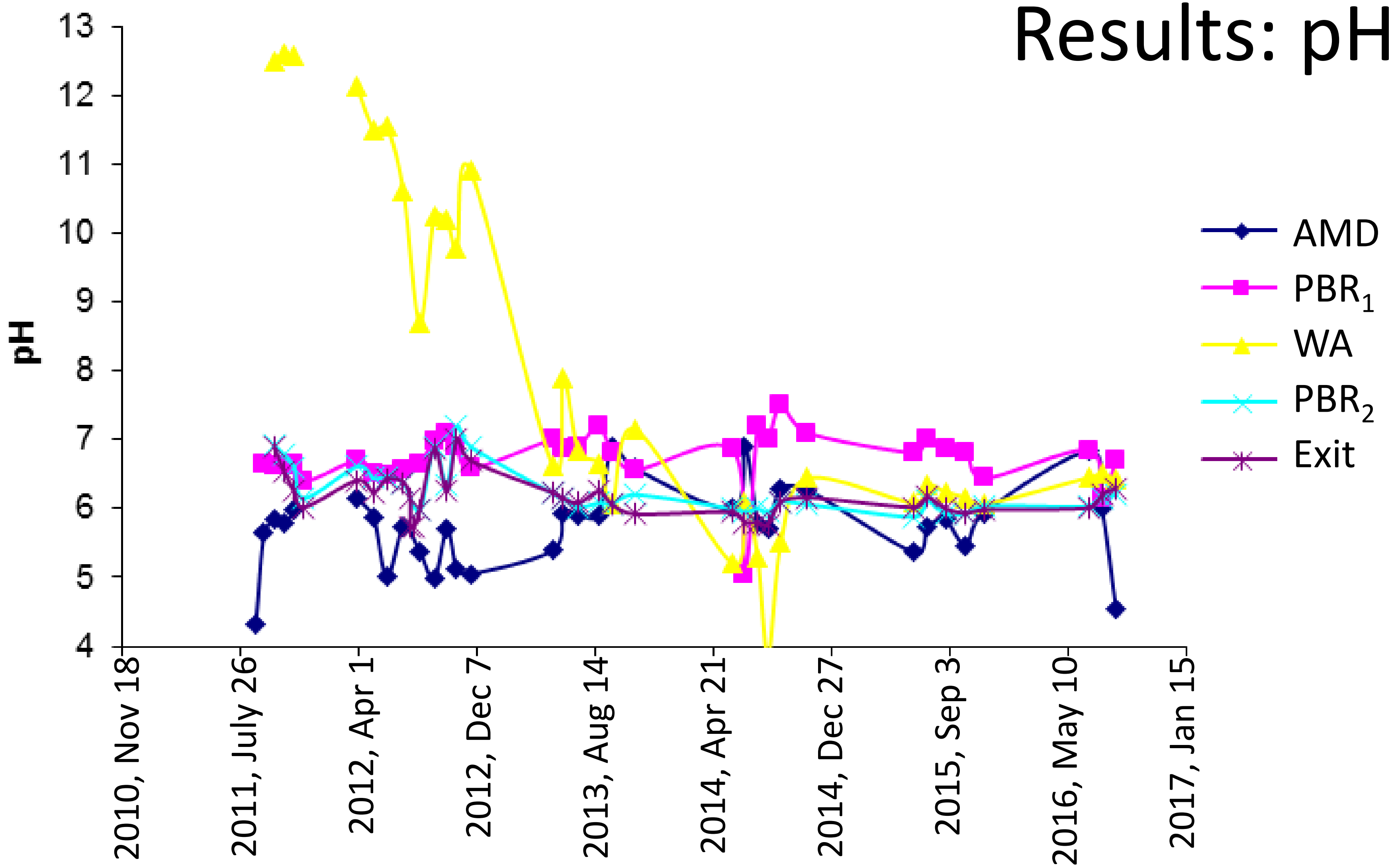

Inferior HDPE membrane placement


Material placement


Superior HDPE membrane


Covering system with soil

Results: pH



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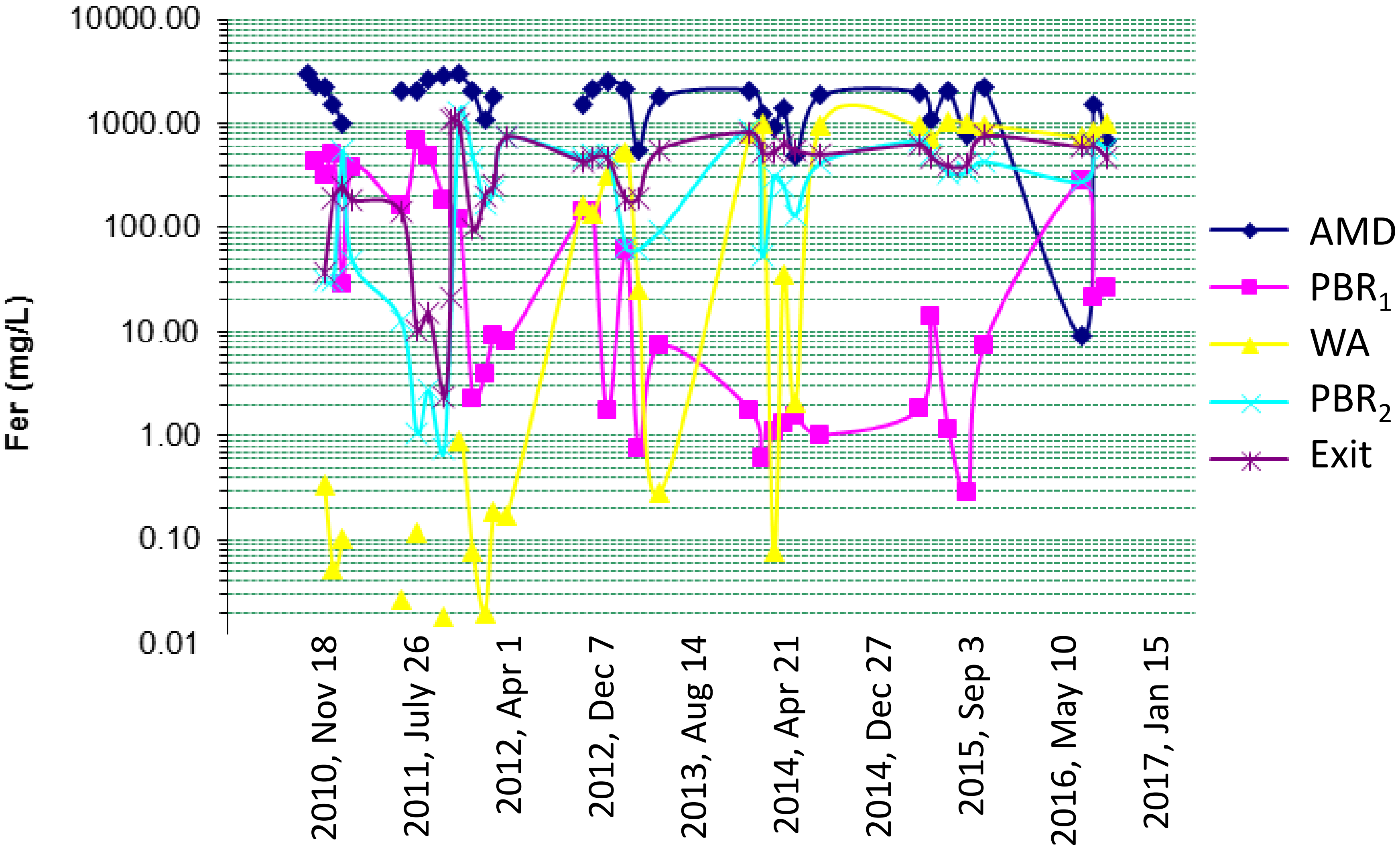
Results: Fe



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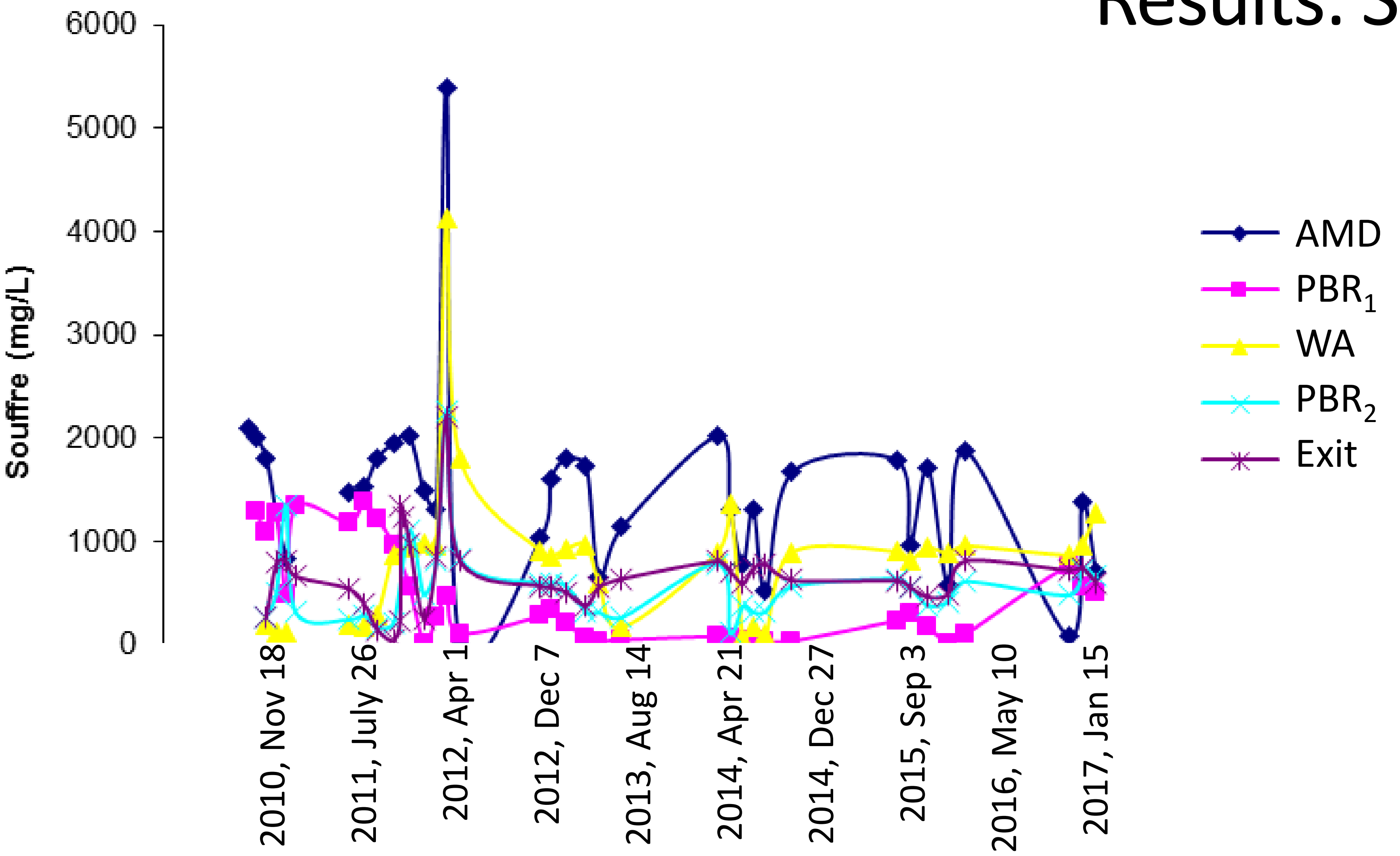
Results: S



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1 Monitoring data (2011-2016)

- Metals / metalloids removal
  - Compliance with regulation, except for Fe (and Mn)

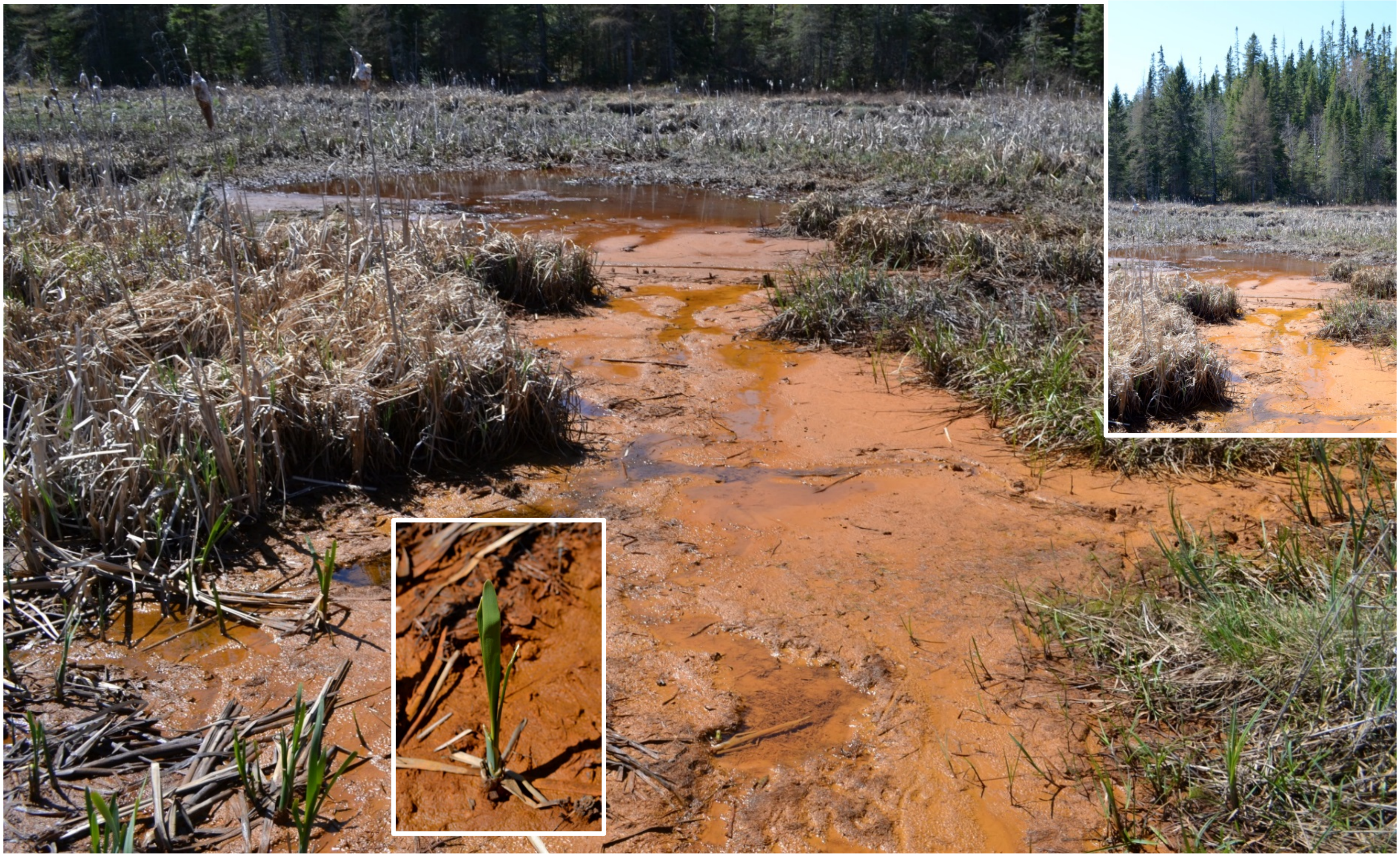
Characteristics	pH	As	Cu	Fe	Ni	Pb	Zn
		(mg/L)					
AMD	4.3 – 6.9	<0.06	<0.003	1 800	0.62	0.19	0.26
Treated effluent	5.8 – 7	<0.01	<0.003	411	0.06	0.03	0.07
Best quality (August 2015)	6	<0.01	<0.01	389	<0.004	<0.07	0.06
Quebec discharge regulation	**6-9**	**0.2**	**0.3**	**3**	**0.5**	**0.2**	**0.5**
Compliance with regulation	**YES**	**YES**	**YES**	**NO**	**YES**	**YES**	**YES**

(Genty et al., 2016)

1 Cascade aeration downstream (2016)

(Rakotonimaro, 2017)

1 Natural wetland downstream (2016)



(Rakotonimaro, 2017)

1 Dolomite drains: 2016

Dol-1

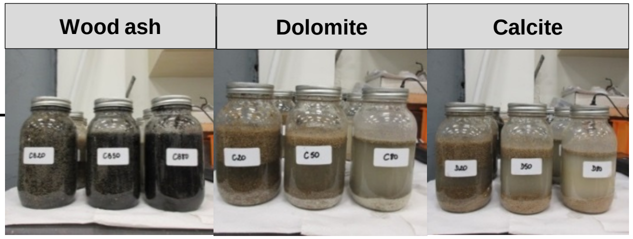
Dol-2

(Rakotonimaro, 2017)

Phase III: lab testing (2 years)

Step 1 – Batch testing (1 L)

Selection the most efficient DAS

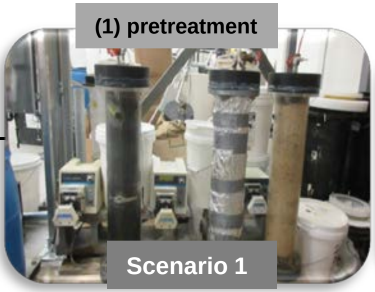


Step 2 – Column testing (1,7 L)

Select optimal HRT (1–5 d);
Evaluate k_{sat} and n

Step 3 – Multi-step (10,7 L)

Performance evolution



Synthetic AMD: pH 4, 2.5 g/L Fe, 5.4 g/L SO_4^{2-}

Monitored parameters: physicochemical, hydraulic, microbiological, mineralogical

HRT: Hydraulic Retention Time; k_{sat}: permeability; n: porosity

1 Results: batch testing

DAS reactors and PBRs

- Most efficient mixture: DAS-wood ash
  - High pH (6.25 - 7.14) and alkalinity
  - 4 h of contact time enough, if Fe < 1.5 g/L
  - 6–11h required, if Fe initial > 1.5 mg/L
  - WA50 (50% wood ash, 50 % wood chips): optimal
- DAS- calcite and DAS-dolomite: comparable efficiency
  - DAS- calcite : more efficient than DAS-dolomite, only temporarily
  - C20 (20% calcite, 80% wood chips): used as post-treatment
- Low SO_4^{2-} removal in all reactors

(Rakotonimaro et al., 2016)

1 Results: column testing

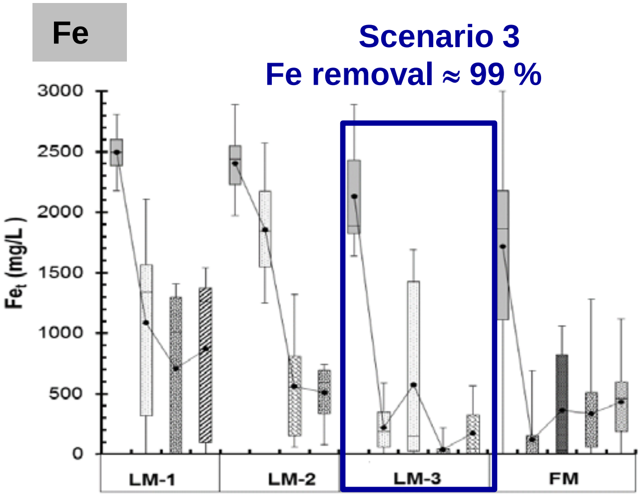
Parameters	**DAS reactors**		**PBRs**	
	WA50	**C20**	**2.5d HRT (R2.5)**	**5d HRT (R5)**
pH	5.3–6.3	6–7	6.2 ± 0.5	6.6 ± 0.5
Alkalinity (mg $CaCO_3$/L)	130–350	16–50	90–2300	430–2800
Acid neutralisation (%)	62	18–47	66	76
Fe removal (%)	up to >96	47–73	77	91
SO_4^{2-} removal (%)	<35	<5	<5	13

- WA50, R5: maximal efficiency at 5d of HRT
- C20: maximal efficiency at 2d of HRT, temporarily
- Low SO_4^{2-} removal in PBRs

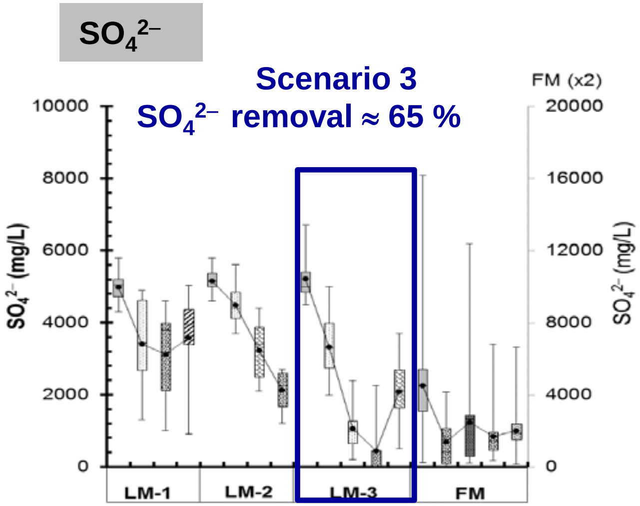
(Rakotonimaro, 2017)

1 Comparative performance: lab vs. field

- **Multi-step** – Laboratory *vs* field (Fe and SO_4^{2-} removal)



Scenario 3
Fe removal ≈ 99 %



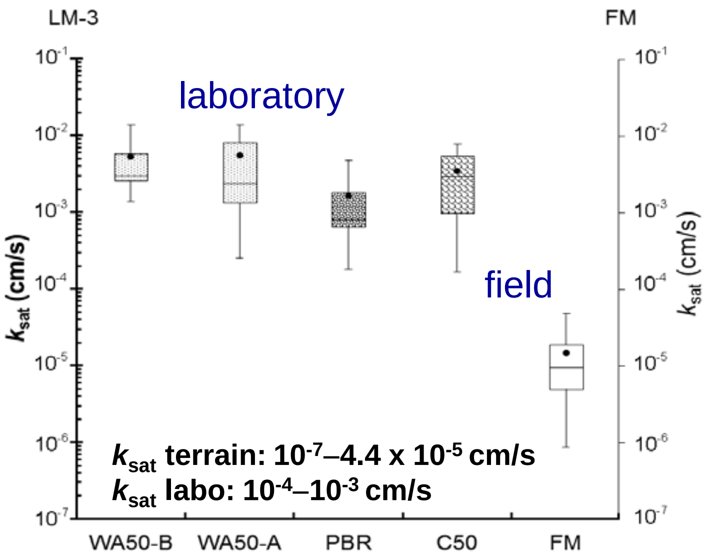
Scenario 3
SO_4^{2-} removal ≈ 65 %

- Lab: best efficiency with scenario 3
- Field: 91 % Fe (first 2 years), then 53 %
  68 % SO_4^{2-} (first 2 years), then 43 %

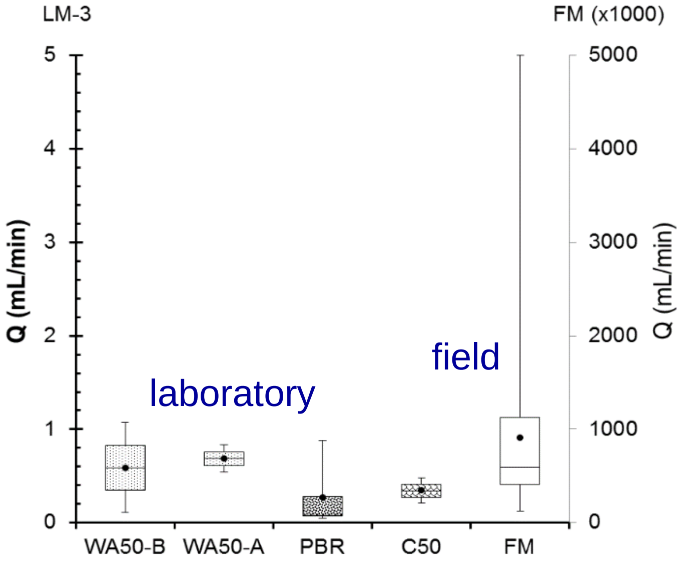
(Rakotonimaro, 2017)

Comparative results: lab vs. field

- **Multi-step** – Laboratory *vs* field (hydraulic evolution)



k_{sat} terrain: 10^{-7}–4.4 x 10^{-5} cm/s
k_{sat} labo: 10^{-4}–10^{-3} cm/s



- ➢ k_{sat} labo = 1–2 order of magnitude higher than k_{sat} terrain
- ➢ Q variable in field (HRT = variable) ≠ Q lab controlled (HRT = ct)

(Rakotonimaro, 2017)

1 Comparative results: literature

System type	Design factors	References
Biochemical		
Anaerobic wetland (AnW)	3,5 g acidity/m^2/d ; 10 g Fe/m^2 /d	Hedin et al (1994); Skousen and Ziemkiewicz (2005)
Vertical flow wetland (VFW)	35 g acidity/m^2/d	Kepler and McCleary (1997)
PBR (mussel shell) (initial Fe = 65,8 mg/L SO_4^{2-} = 608 mg/L)	29 g SO_4^{2-}/m^3 substrate/d (94%)	McCauley et al (2009)
PBR (calcite) (Following two DAS; initial Fe ≈ 35 mg/L; SO_4^{2-} ≈ 1000 mg/L)	**4–73 g Fe/m^3/d, 2–117 g SO_4^{2-}/m^3/d (≈ 99 %)**	**Rakotonimaro (2017)**
Geochemical		
Anoxic limestone drain (ALD)	15 h residence time; 50 g acidity/t/d	Watzlaf (2004); Skousen and Ziemkiewicz (2005)
Limestone leach bed (LLB)	2 h residence time ; 10 g acidity/t/d	Skousen and Ziemkiewicz (2005)
DAS (C20) (initial Fe = 250 mg/L)	HRT (1 d), 42 % Fe	Rötting et al (2008a)
DAS (C20) (initial Fe ≈ 2000 mg/L)	**Fe (73%, HRT = 2 d)**	**Rakotonimaro (2017)**
DAS (C50)- pretreatment (initial Fe = 1800 mg/L)	**Fe (67%, HRT = 3 d)**	
DAS (WA50) (initial Fe ≈ 2000 mg/L)	**Fe (> 89% , HRT = 3 d)**	

(Skousen et al., 2017; Rakotonimaro, 2017)

Summary

- DAS-wood ash: most efficient for Fe pre-treatment
- 2 units of pre-treatment : more efficient than one
- DAS-calcite and DAS-dolomite: comparable efficiency
- No clogging issues in lab testing
- Treatment performance (lab / field) depends on Q, Fe, SO_4^{2-}

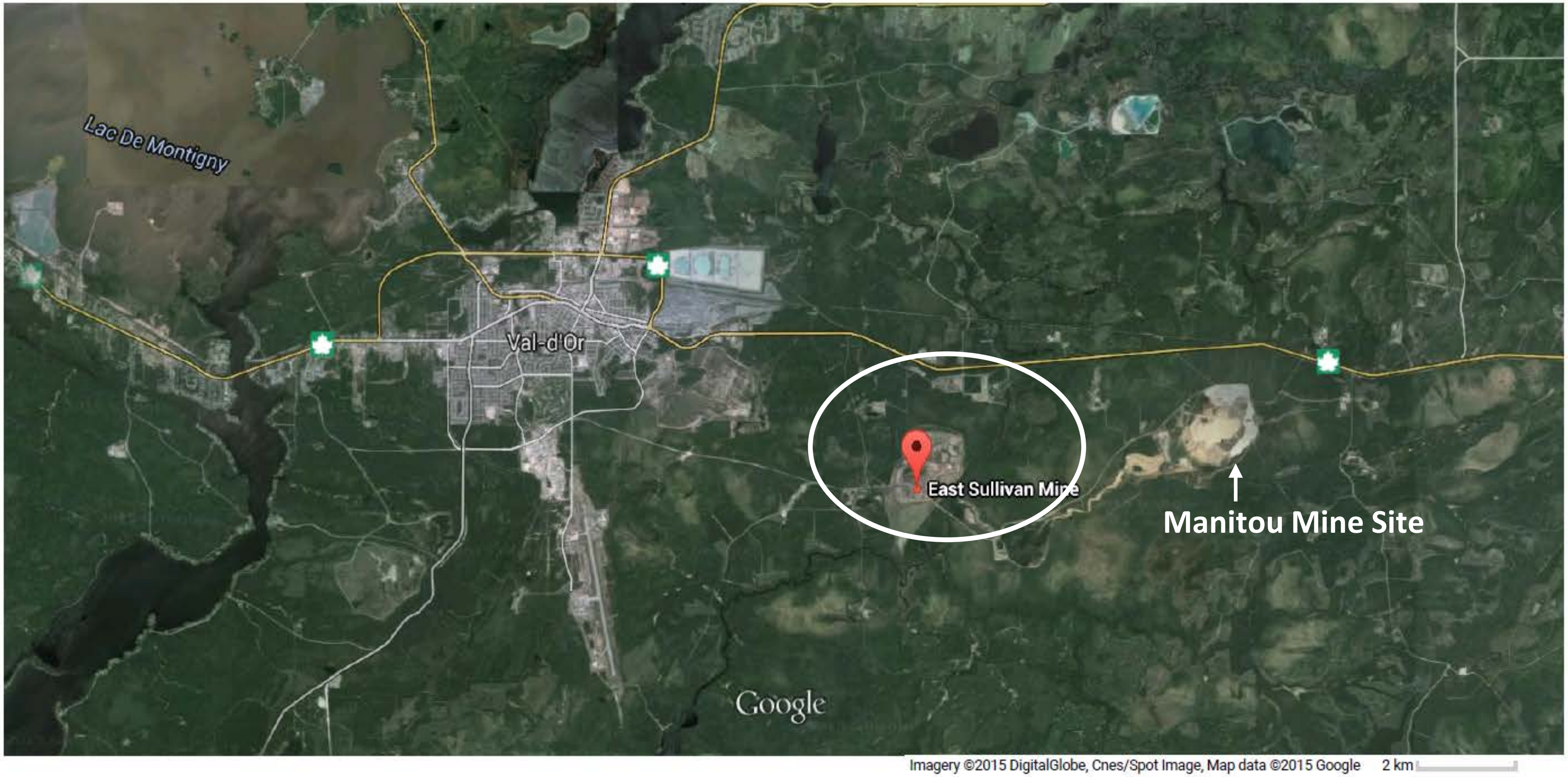
Future work

- Excavation of the 3 units and replacement by 2-3 DAS systems
- Mineralogical and microbiological characterization of solids

Case study II: East Sullivan mine site

- Historic, Rehabilitation

2 Location



Imagery ©2015 DigitalGlobe, Cnes/Spot Image, Map data ©2015 Google

6 km E of the Val-d'Or town, SW QC, Canada

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2 East Sullivan mine site: historic

1945

1990

1942-1979 : Cu, Zn, Au, Ag, Cd

- **15 Mt (200 ha) of tailings, 200kt of acid generating material; 228 ha impacted**
- **3.6% S, thickness of 7.3 m in average**

http://sebastienlavoie.com/maitrise/photos.html

http://www.mrn.gouv.qc.ca/mines/restauration/restauration-sites-east-sullivan.jsp

2 East Sullivan mine site

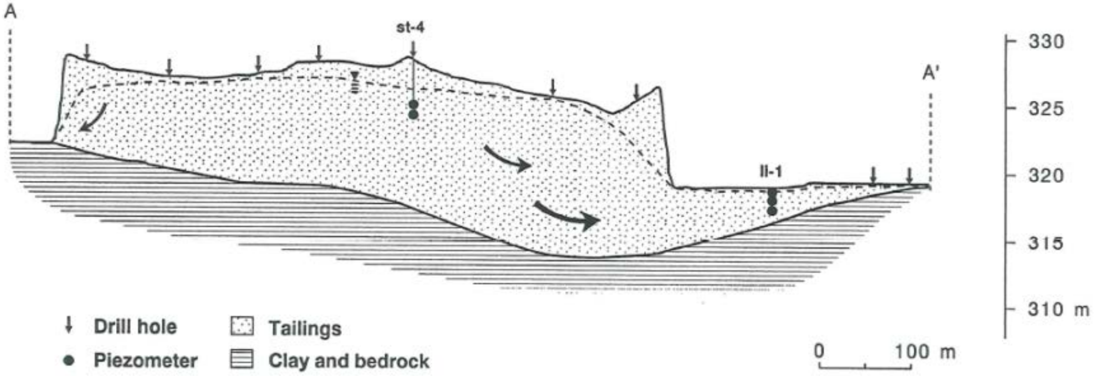

Figure 2. Cross-section A-A' through the East-Sullivan tailings impoundment. Arrows indicate locations of drill holes to the bedrock or clay basement. Dots represent piezometer locations for water sampling.

- **Pore-water quality in 1990**
  - pH ≈ 2
  - Fe (Fe^{2+}): up to 17 g/L
  - SO_4^{2-}: up to 37 g/L
  - Cu, Pb, Zn : 0.1-1 g/L

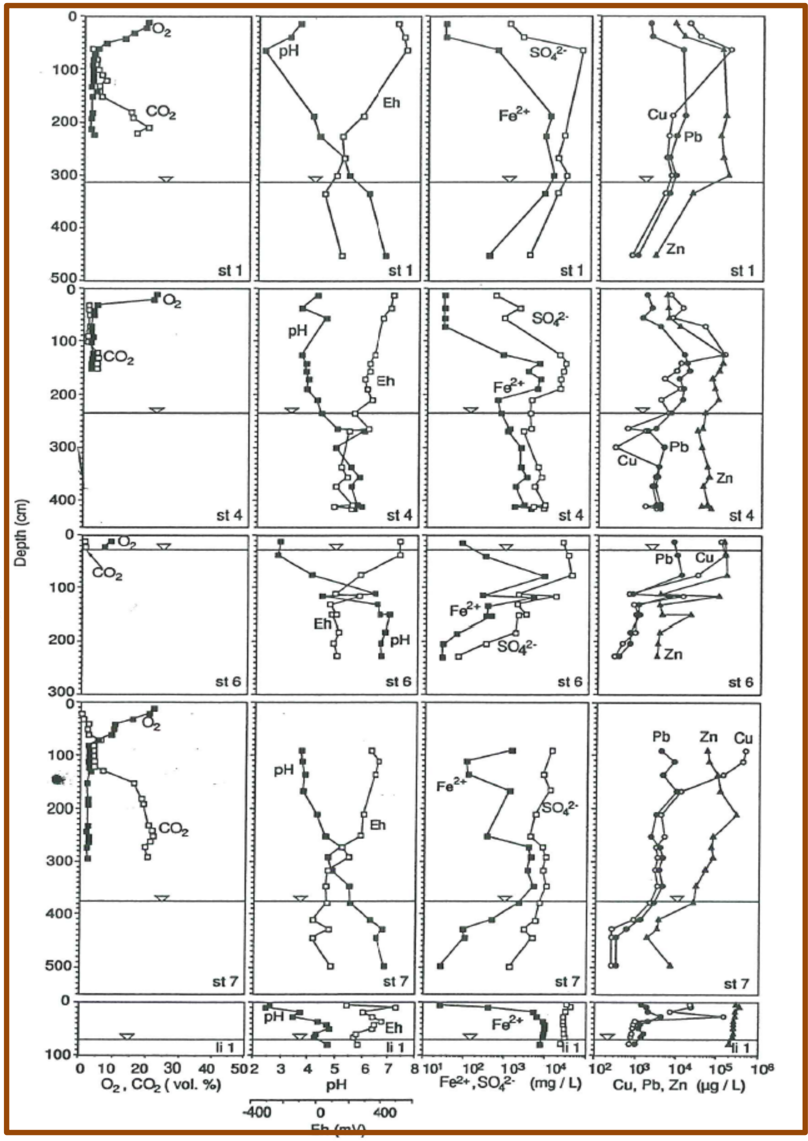

Figure 3. Profiles of O_2 and CO_2 in pore gases and of pH, Eh, Fe^{2+}, SO_4^{2-}, Cu, Pb, and Zn in pore-waters at the five selected stations located in Figure 1. The line with the triangle indicates the average water-table depth during the sampling period.

(Germain et al., 1994)

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2 East Sullivan: rehabilitation

- **1984:** **Wood waste cover (prevention and treatment)**
- **1990:** Seepage collection system
- **1992-1996:** Confining dike (6 km)
- **1998-2005:** “Active” treatment of collected AMD in wetlands
- **[2014:** Wood cover of the eastern sector, not completed**]**

⇒ **Some effluents are still acidic**

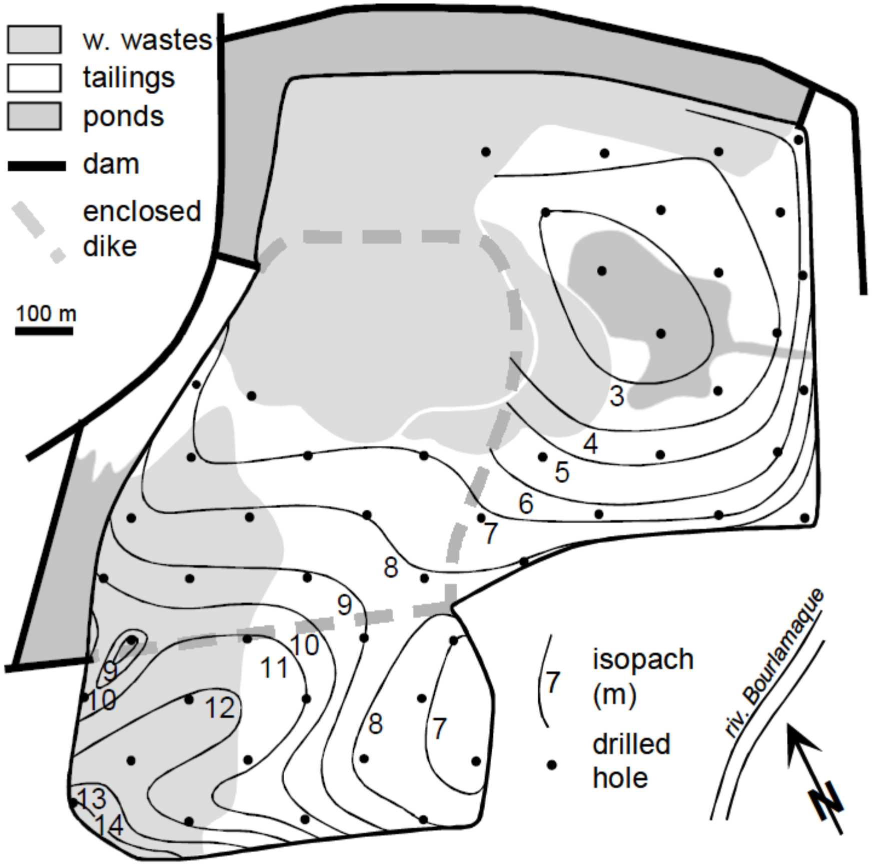
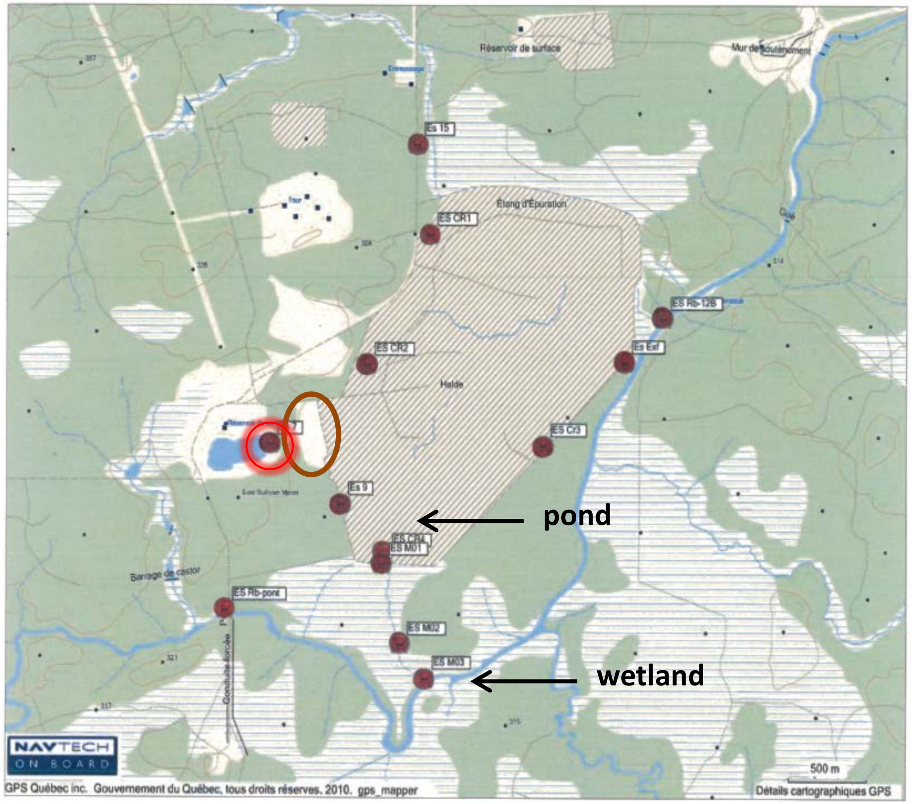


Figure 1. Map of the East Sullivan tailings impoundment in 1994

(Tassé and Germain, 2004)

East Sullivan: monitoring (2000-2014)

- 12 sampling points
  - 7 points: dam and settling ponds
  - 5 points: tailings edges
- Parameters
  - pH, alkalinity, TDS, Fe, Al, Mn, Cu, Zn, Pb, SO_4^{2-}
- Compliance, except for the uncovered tailings area



GPS Québec inc. Gouvernement du Québec, tous droits réservés, 2010. gps_mapper

(Rakotonimaro et al., 2015)

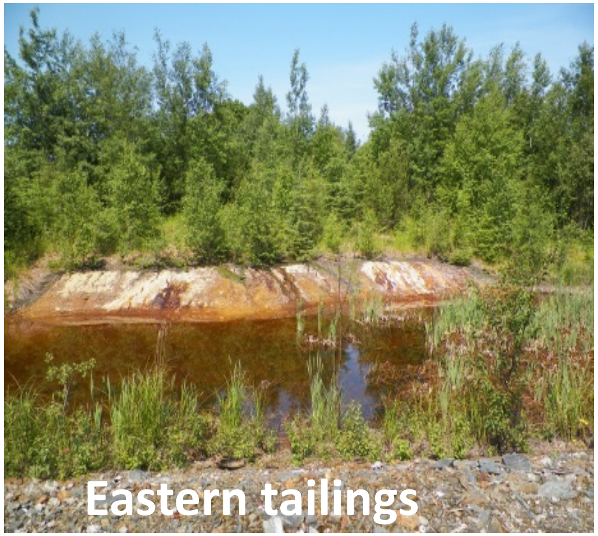
2 Summary

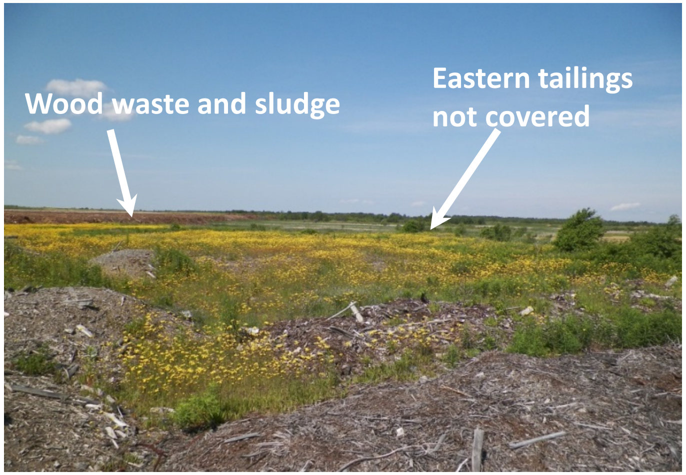
- Efficiency of wood-waste cover for over 14 years
- Significant improvement of water quality
- Site presently turning into <u>birds' refugee</u> (southern and eastern ponds, more than 190 species listed)

Future work

- Completion of the eastern part of tailings by wood-waste and sludge (< 10% of total)
- Mineralogical / microbiological characterization of solids
- Further risk assessment

2 East Sullivan: 2015





(Rakotonimaro et al., 2015)

Concluding remarks

- Use of residual materials (dolomite, wood ash, compost, manure): low cost
- Relatively easy to install and operate
- Maintenance (more or less) required

BUT

- Limited performance at high contamination level: **multi-step systems (?)**
- Unpredictable long-term efficiency
- Solutions not available for sludge management

However, sometimes is the only available option

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