# PROJECT REPORT 2000-6

sustainable forest management network

réseau sur la gestion durable des forêts

# High Temperature Biological Treatment of Foul Evaporator Condensate for Reuse

A Network of Centres of Excellence
Un réseau de centres d'excellence

**Pierre R. Bérubé and Eric R. Hall**

For copies of this or other SFM publications contact:

Sustainable Forest Management Network
G208 Biological Sciences Building
University of Alberta
Edmonton, Alberta, T6G 2E9
Ph: (780) 492 6659
Fax: (780) 492 8160
http://www.ualberta.ca/sfm/

ISBN 1-55261-064-0

# High Temperature Biological Treatment of Foul Evaporator Condensate for Reuse

SFM Network Project: **Membrane Bioreactors for Contaminant Control in Closed Pulp and Paper Mills**

by

**Pierre R. Bérubé and Eric R. Hall**

Department of Civil Engineering and the Pulp and Paper Centre
The University of British Columbia, Vancouver, B.C.

**April 2000**

## ABSTRACT

There is increasing interest in the treatment and reuse of the sewered portion of the evaporator condensate from kraft pulp mills. Reusing the treated condensate would reduce the raw water requirements, reduce the contaminant load to the existing combined mill effluent treatment system and potentially result in significant energy savings if the heat content of the condensate can be recovered. In addition, some legislation proposes incentives for treating and reusing the condensate.

This study indicated that it was possible to biologically remove methanol (primary contaminant of concern) from synthetic condensate using a high temperature membrane bioreactor (MBR), over the entire expected range of temperatures for evaporator condensate (55 to 70 $^{o}$C). However, the operating temperature exerted a significant impact on methanol removal kinetics. Methanol removal was optimal at a temperature of 60 $^{o}$C. The removal of methanol was significantly inhibited above 60 $^{o}$C, indicating that the inactivating effect of temperature on the microorganisms in the MBR must be considered at elevated temperatures. A model was proposed and used to accurately estimate the effect of high temperatures on methanol removal.

Further investigations revealed that the removal of methanol was significantly impacted by the presence of the other (non-methanol) contaminants contained in real condensate. These contaminants resulted in a shift in the composition of the microbial community present in the MBR, which in turn reduced the rate of methanol removal. This indicates that to maximize the removal of methanol, the condensate should be treated separately from other waste streams that may contain high concentrations of other contaminants. Nonetheless, the removal of methanol from real condensate at the optimal temperature of 60 $^{o}$C using an MBR was substantially more efficient than that using conventional technologies.

High temperature treatment using an MBR also successfully removed the other contaminants of concern (non-methanolic organic compounds and reduced sulphur compounds) contained in real condensate. Furthermore, real condensate treatment for reuse using a high temperature MBR was shown to be 40 to 50 % less expensive than treatment using conventional technologies. Therefore, treatment of real condensate for reuse using a high temperature MBR is not only technically feasible, but is more economically attractive than treatment using conventional technologies.



## ACKNOWLEDGEMENTS

This research was funded in part by the Sustainable Forest Management Network, Natural Science and Engineering Research Council of Canada and the Pulp and Paper Research Institute of Canada. Special thanks go to the Western Pulp Limited Partnership bleached kraft pulp mill, Squamish, Canada, for assisting with the characterization of the evaporator condensate and for providing process information.

# INTRODUCTION

## Problem Definition

Tighter regulatory requirements and public interest in “environmentally friendly” pulp and paper products have encouraged the pulp and paper Industry to reconsider its wastewater treatment practices (Mannisto et al. 1996; NCASI 1998). As an alternative to conventional end-of-pipe wastewater treatment, some mills are considering closing up selected process water systems to reuse the wastewater as process water (Bérubé and Hall 1996). Reusing the wastewater can reduce the contaminant load to the existing combined mill effluent treatment system, reduce the raw water requirements, and potentially reduce the impact of discharging treated wastewater to the environment (Vora and Venkataraman 1995; NCASI 1998; Blackwell et al. 1979). The ultimate goal would be to eventually reuse all the wastewater as process water.

Under current operating conditions, kraft pulp mills typically reuse a portion of the cleaner fraction of the evaporator condensate along with clean water as process water in pulp washing and recausticizing as illustrated in Figure 1 (NCASI 1998). This fraction of the evaporator condensate is clean enough to be reused directly. However, the foul fraction of the evaporator condensate is too contaminated to be reused directly. Reusing the foul evaporator condensate without treating it could result in ambient air quality problems because of the subsequent release of hazardous air pollutants (HAP) and foul odorous compounds it contains (Venkatesh et al. 1997; Jain 1996; Jett 1995; NCASI 1994b-f). Such emissions can cause unpleasant or even hazardous working conditions for mill staff (ACGIH 1999). The foul evaporator condensate is typically sewered and then treated in a combined mill effluent treatment system before being discharged to the environment. Some mills steam strip the foul evaporator condensate before sewering it to minimize potential ambient air quality problems that could occur during subsequent treatment in the combined mill effluent treatment system (NCASI 1994a).

There is increasing interest in the treatment and reuse of the foul evaporator condensate, hereon referred to as evaporator condensate (Barton et al. 1996; Vora and Venkataraman 1995). The treated evaporator condensate could be reused in pulp washing, recausticizing, bleaching and papermaking instead of clean water as illustrated in Figure 2 (Sebbas 1987; Pekkanen and Kiiskilä 1996). In addition to reducing the contaminant load to the existing combined mill effluent treatment system, reducing the clean water requirements and potentially reducing the impact of discharging the treated condensate to the environment, reusing the condensate could also result in significant energy savings if the heat content of the evaporator condensate can be recovered (Sebbas 1987; Durham 1991). Also, some legislation proposes a number of incentives for treating and reusing the condensate as process water (Vice and Carroll 1998). The ability to treat and reuse the evaporator condensate would be a significant step towards the ultimate goal of producing a zero effluent mill.

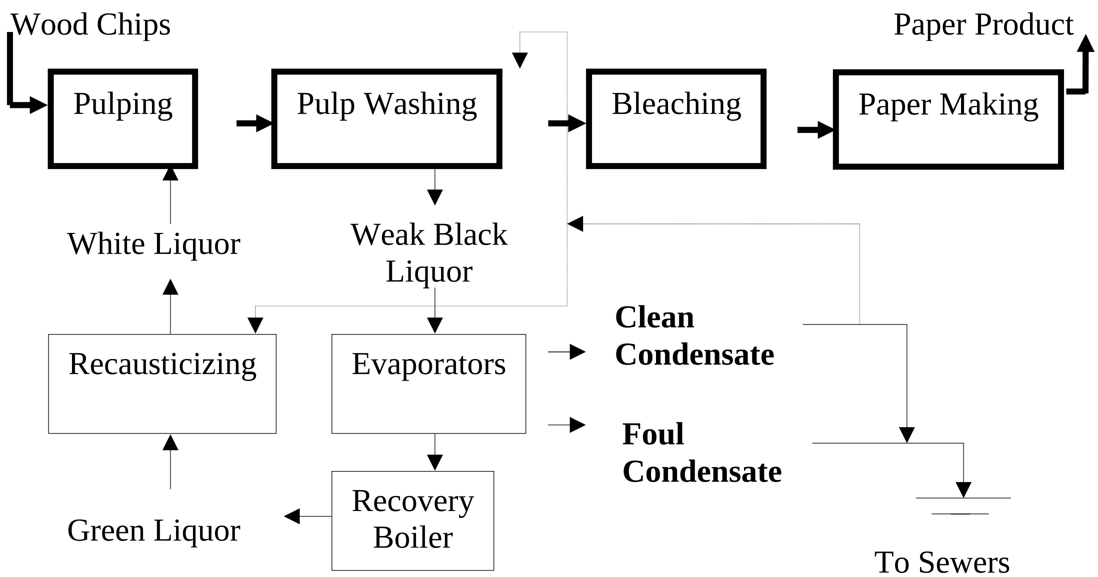



Figure 1 - Schematic of a bleached kraft pulping process and chemical recovery cycle. (bold lines: pulping process; medium lines: chemical recovery cycle; dashed lines: reuse of evaporator condensate)

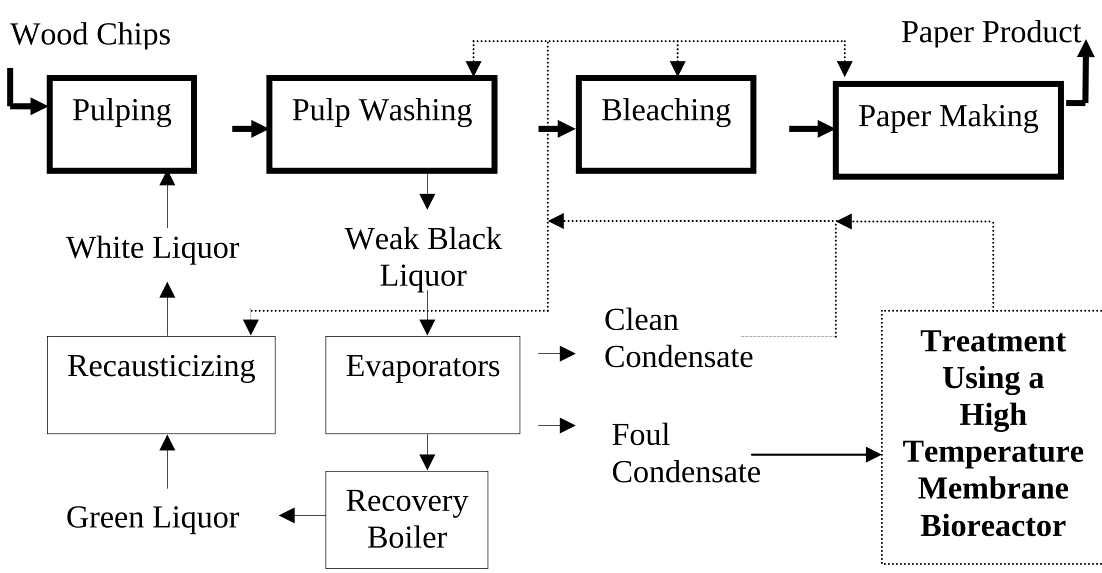



Figure 2 - Schematic of a bleached kraft pulping process with treated evaporator condensate reuse.

A number of conventional technologies exist that could be used to treat the evaporator condensate for reuse. However, the relatively poor treatment efficiencies and/or the high costs associated with these conventional systems provided incentives to investigate and develop a better treatment technology for evaporator condensate treatment for reuse (Bérubé 2000).

## Treatment Requirements for Reuse

To be reused as process water, the evaporator condensate must be treated. Of particular concern are the hazardous air pollutants (HAP) and foul odorous compounds contained in the evaporator condensate. These contaminants could volatilize to the atmosphere, potentially resulting in unpleasant or even hazardous working conditions for mill staff (Jain 1996; Venkatesh et al. 1997; Jett 1995; NCASI 1994b-f). In addition, the presence of other trace organic compounds and particulate matter could disrupt pulping processes and impact pulp quality (Sebbas 1987; Annola et al. 1995; Niemela et al. 1999)

The treatment requirements for the reuse of evaporator condensate are summarized as follows.

1. ***Hazardous air pollutants***
   NCASI (1994a) suggests that for reuse, the concentration of methanol in the treated condensate should be less than 20 mg/L. Methanol accounts for up to 90 % of the organic contaminants present in evaporator condensate and is classified as a hazardous air pollutant.

2. ***Foul odorous compounds***
   The foul odors associated with kraft pulp mills are mainly due to the reduced sulphur compounds (RSC) contained in the evaporator condensate. Hydrogen sulphide and methyl mercaptan, the main contributors to the foul odor, should be completely removed from the evaporator condensate before reuse as process water to prevent any further increase in the ambient air concentrations of these RSC at kraft pulp mills. Under current operating conditions, the ambient air concentrations for these RSC at many mills periodically, or even consistently, exceed the maximum allowable levels (Jappinen et al. 1993; Kangas et al. 1984; Leech and Chung 1982; ACGIH 1999). These RSC are not only of concern because of their foul odor but also because of their toxicological effects on humans (ACGIH 1999)

3. ***Trace organic compounds***
   In addition to methanol and RSC, the evaporator condensate contains other alcohols, terpenes, ketones, phenolics and numerous other organic contaminants at trace levels. These trace organic contaminants can increase the consumption of process chemicals, form potentially hazardous by-products and negatively impact the pulping process and pulp products. Consequently, these trace organic contaminants must be removed before the evaporator condensate can be reused.

4. ***Solids***
   The treated evaporator condensate should contain no suspended solids that could potentially clog process showers and felts when reused.

5. ***High temperature***

The expected temperature range for evaporator condensate is approximately 55 to 70 $^{o}$C and the temperature of the various processes where the treated evaporator condensate could be reused ranges from 65 to over 100 $^{o}$C. Consequently, significant energy savings could be achieved if the evaporator condensate were treated without pre-cooling, as would be required for conventional aerobic biological treatment, and reused with no or minimal external heating.

## Evaluation of Treatment Technologies

Based on the treatment requirements and a literature review, a number of conventional technologies were evaluated for the treatment of evaporator condensate for reuse (Bérubé 2000). As previously mentioned, the relatively poor treatment efficiencies and/or the high costs associated with conventional treatment systems provided incentives to investigate and develop a better technology for the treatment of evaporator condensate for reuse. To address the disadvantages associated with conventional technologies, a novel technology consisting of a high temperature membrane bioreactor (MBR), was considered for the treatment of evaporator condensate for reuse. An MBR is an aerobic biological treatment system that consists of a reactor tank, where a mixed culture of microorganisms consume the contaminants of concern, and a recycling loop that separates the treated water from the mixed microbial culture using an ultrafiltration membrane (Figure 3).

The efficiencies of the different treatment systems considered at meeting the treatment requirements for reuse are presented in Table 1. Based on a literature review, a high temperature MBR appeared to be the most promising technology for the treatment of evaporator condensate for reuse. It can potentially achieve higher methanol, RSC and trace organic compound removal efficiencies than anaerobic biological treatment or steam stripping. An MBR can also achieve a much higher suspended solids removal efficiency than conventional biological treatment systems, especially at elevated temperatures, since the membrane component of the MBR retains all solids. In addition, because of its closed configuration, an MBR could be operated at a high temperature without exposing the microorganisms in the treatment system to significant temperature fluctuations, which would reduce the efficiency of the biological treatment system, and the emission of HAP and foul odorous compounds could be minimized. Finally, because of the high temperature operation, the heat content of the evaporator condensate can be recovered. A potential recycling scheme for evaporator condensate treated for reuse using a high temperature MBR is presented in Figure 2.

Table 1 - Summary of the evaluation of potential technologies for the treatment of evaporator condensate for reuse

| Treatment Requirements | *Steam Stripping | *Anaerobic Biological Treatment | *Aerobic Biological Treatment | High Temperature MBR |
|---|---|---|---|---|
| Ability to remove methanol | Moderate | Moderate | Good | Potentially good |
| Ability to remove RSC | Good | Moderate | Good | Potentially good |
| Ability to remove trace organic contaminants | Moderate To poor | Moderate | Good | Potentially good |
| Suspended solids in effluent | Relatively low | Relatively high | Relatively high | None |
| Cooling required | None | Possibly | Yes | None |

(* conventional treatment technologies)

**Research Requirements**

There is limited information available regarding the biological treatment of evaporator condensate, especially at elevated temperatures. Consequently research was required to improve our understanding of the physical, chemical and biological processes that occur during high temperature biological treatment of evaporator condensate using an MBR. A better understanding of these processes is required to properly evaluate, design and operate a high temperature MBR to treat evaporator condensate for reuse. To improve our understanding of these processes, a number of fundamental questions needed to be addressed. These questions are summarized as follows.

1. What are the effects of high temperature operation on the activity of the microorganisms in the biological treatment system? Conventional biological treatment systems can typically operate at temperatures up to 35 $^{o}$C. The proposed high temperature MBR will operate at temperatures ranging from 55 to 70 $^{o}$C (expected temperature range for evaporator condensate stream).

2. Do the contaminants present in the evaporator condensate matrix inhibit microbial activity in the biological treatment system? The evaporator condensate contains numerous contaminants that are known to inhibit microbial activity. Therefore, the microbial activity could be inhibited to a point at which the biological treatment of evaporator condensate would not be technically feasible.

3. Can all of the contaminants of concern be removed during high temperature biological treatment using an MBR?

4. Is the high temperature biological treatment of evaporator condensate for reuse using an MBR economically attractive?

**Experimental Set-Up (Bench Scale MBR)**

To address the fundamental questions listed above, a series of experiments were designed and performed (Bérubé 2000). A bench scale MBR, presented in Figure 3, was used for the different experiments. The MBR consisted of a biological reactor tank, a ceramic tubular ultrafiltrafiltration membrane and a recycling pump.

The MBR was fed semi-continuously by adding a mixture of evaporator condensate and nutrients, once every 3 hours. Semi-continuous feeding was chosen because it can yield more information about removal kinetics than experiments performed under strict continuous flow conditions. A detailed description of the MBR and the operating conditions is presented in Bérubé (2000).

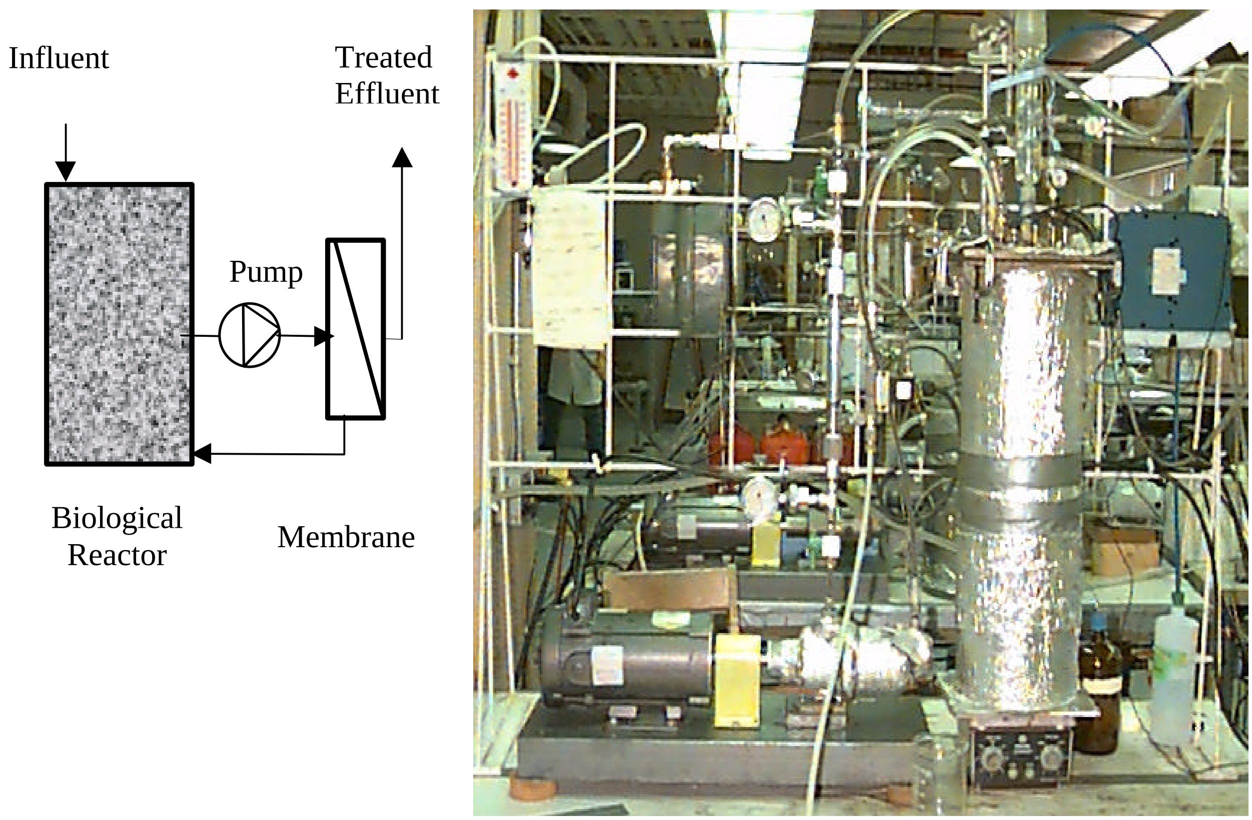



Figure 3: Bench-scale MBR.

# SUMMARY OF RESULTS

**Effects Of High Temperature Operation On The Activity Of The Microorganisms In The Biological Treatment System** (Bérubé and Hall 2000a)

The effect of elevated operating temperature was investigated by progressively increasing the operating temperature of the MBR from 55 to 70 $^{o}$C (expected range of temperatures for evaporator condensate stream).

It was possible to biologically remove methanol from synthetic evaporator condensate using a high temperature MBR, over the entire expected range of temperatures for evaporator condensate. Synthetic evaporator condensate was used when investigating the effect of high temperature operation on the activity of the microorganisms in the biological treatment system because of the difficulty associated with using real evaporator condensates due to their extremely foul odorous nature. The synthetic evaporator condensate contained only methanol as substrate for the microorganisms in the MBR.

The operating temperature exerted a significant impact on the activity of the microorganisms in the MBR. A maximum specific methanol utilization coefficient of approximately 0.84 ± 0.08 /day was observed at an operating temperature of 60 $^{o}$C as illustrated in Figure 4. The specific methanol utilization coefficient was used as a surrogate measurement to estimate the activity of the microorganisms in the MBR. Above 60 $^{o}$C, the specific methanol utilization coefficient declined sharply, suggesting that at high operating temperatures, the inactivating effect of temperature on microorganisms must be considered.

The relationships commonly used to model the effect of temperature on biological treatment systems do not take into account the inactivating effect of temperature on the microorganisms and therefore, can significantly overestimate contaminant removal rates at elevated temperatures. A model, presented in Equation 1, was proposed and used to accurately estimate the effect of high temperatures on methanol removal kinetics in an MBR over the temperature range investigated as illustrated in Figure 4. Based on the model, the optimal operating temperature for the biological removal of methanol by a mixed microbial culture was determined to be approximately 60 $^{o}$C. These results indicated that it is not only possible to operate an MBR at high temperatures, but also that a higher specific methanol utilization coefficient can be achieved at a higher operating temperature. As illustrated in Figure 4, a much higher specific methanol utilization coefficient can be achieved at 60 $^{o}$C than those reported by others for conventional biological treatment systems treating evaporator condensate at much lower temperatures (Barton et al. 1996). However, care may need to be taken not to exceed the critical operating temperature of 60 $^{o}$C.

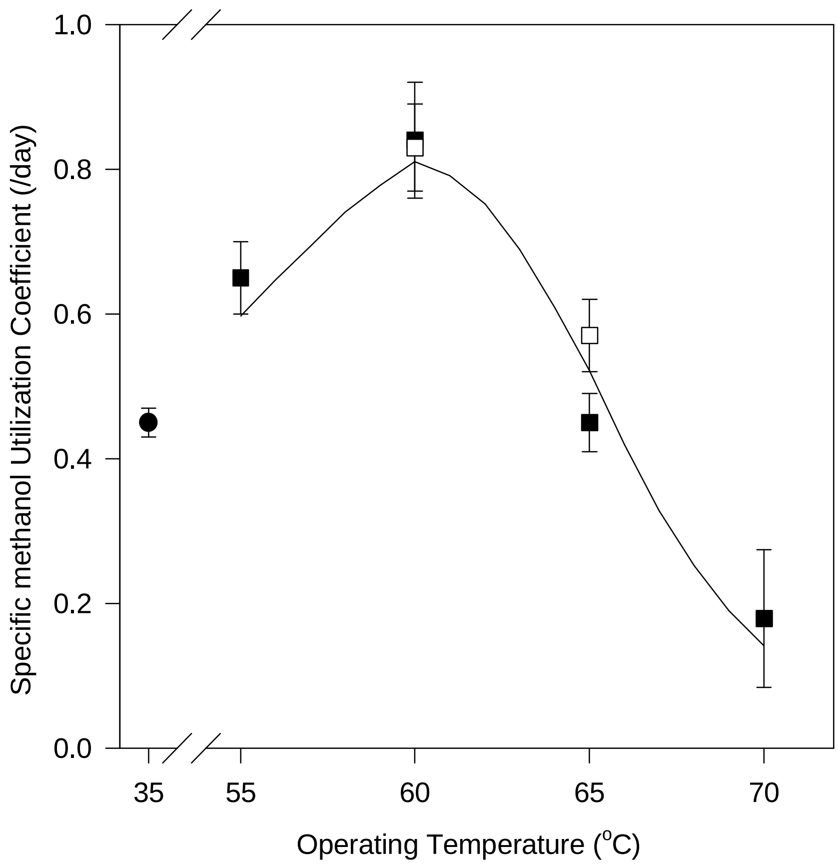



Figure 4: Effect of operating temperature on the specific methanol utilization coefficient. (circle: specific methanol utilization coefficient reported by others at a lower operating temperature (Barton et al. 1996); error bars represent the 90 % confidence intervals for measurements)

$$m_{T'} = m_D q^{(T'-T'_D)} \left( \frac{1}{\left(1 + B' q'^{(T'-T'_D)}\right)} \right) \qquad [1]$$

where:

$m_{T'}$: specific growth coefficient at operating temperature T' (/day)
$m_D$: specific growth coefficient at operating temperature $T'_D$ (/day)
U: specific methanol utilization coefficient (/day) ($m$ = U/Y)
Y: observed growth yield (microorganisms produced/methanol consumed)
$q$ : temperature activation coefficient
T': operating temperature (°C)
$q$': the temperature inactivation coefficient (-)
B': inactivation constant (-)
D: at datum operating temperature

The operating temperature also had a significant effect on the observed microbial growth yield in the MBR. At increasing operating temperatures, a larger fraction of the methanol consumed was converted to energy, reducing the observed growth yield.

**Effect Of Contaminants Present In The Evaporator Condensate Matrix On The Microbial Activity In The Biological Treatment System** (Bérubé and Hall 1999)

Methanol can account for up to 90 % of the mass of organic contaminants contained in evaporator condensate (Blackwell et al. 1979). The remaining fraction consists of numerous non-methanolic organic contaminants present at trace levels. Over 60 trace organic contaminants have been identified in evaporator condensate (Blackwell et al. 1979). Many of these trace organic contaminants are known to inhibit microbial activity in a biological treatment system. Therefore, the microbial activity could be inhibited to a point where the biological treatment of evaporator condensate would not be technically feasible.

The effect of the trace organic contaminants present in the evaporator condensate matrix on the biological activity in the MBR was investigated by progressively replacing the feed to the MBR from 0% real evaporator condensate (100% synthetic evaporator condensate) to 100% real evaporator condensate. The MBR was operated at the optimal temperature of 60 $^oC$. As the fraction of real condensate in the feed increased, the concentration of trace organic contaminants in the feed also increased.

The specific methanol utilization coefficient measured during the treatment of real evaporator condensate was significantly lower than that observed when treating synthetic evaporator condensate as illustrated in Figure 5. However, the reduction in the specific methanol utilization coefficient was not due to the inhibition from the trace organic compounds present in the real evaporator condensate matrix.

The reduction in the specific methanol utilization coefficient was attributed to a shift in the composition of the microbial community present in the MBR. As presented in Figure 6, the morphology of the microorganisms present in the MBR when treating synthetic and real evaporator condensate was substantially different. When treating synthetic evaporator condensate, the microbial community appeared to consist exclusively of small rod-shaped microorganisms (Figure 6a). These microorganisms, hereafter referred to as methylotrophic microorganisms, were capable of growth with methanol as a sole substrate (i.e. capable of growth on synthetic evaporator condensate that contained methanol as only sublstrate). As expected, a more diversified microbial community was observed when treating real evaporator condensate that contained trace organic compounds as well as methanol. In addition to the previously observed small rod-shaped methylotrophic microorganisms, larger rod-shaped and filamentous microorganisms were noted with real evaporator condensate as feed (Figure 6b). These “additional” microorganisms were apparently only capable of growth when trace organic compounds, such as those contained in the real evaporator condensate matrix, were present.

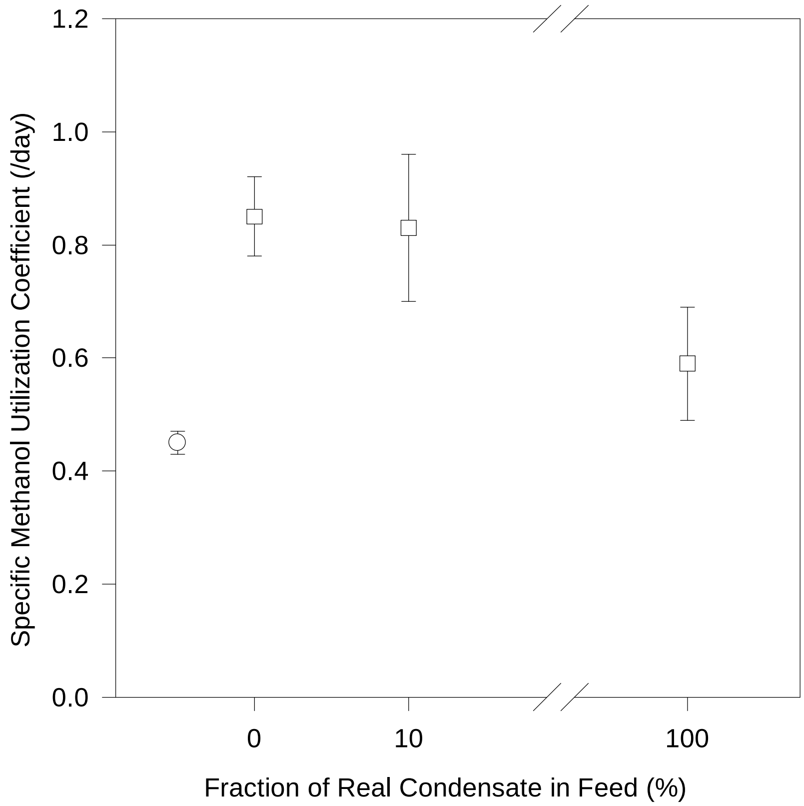
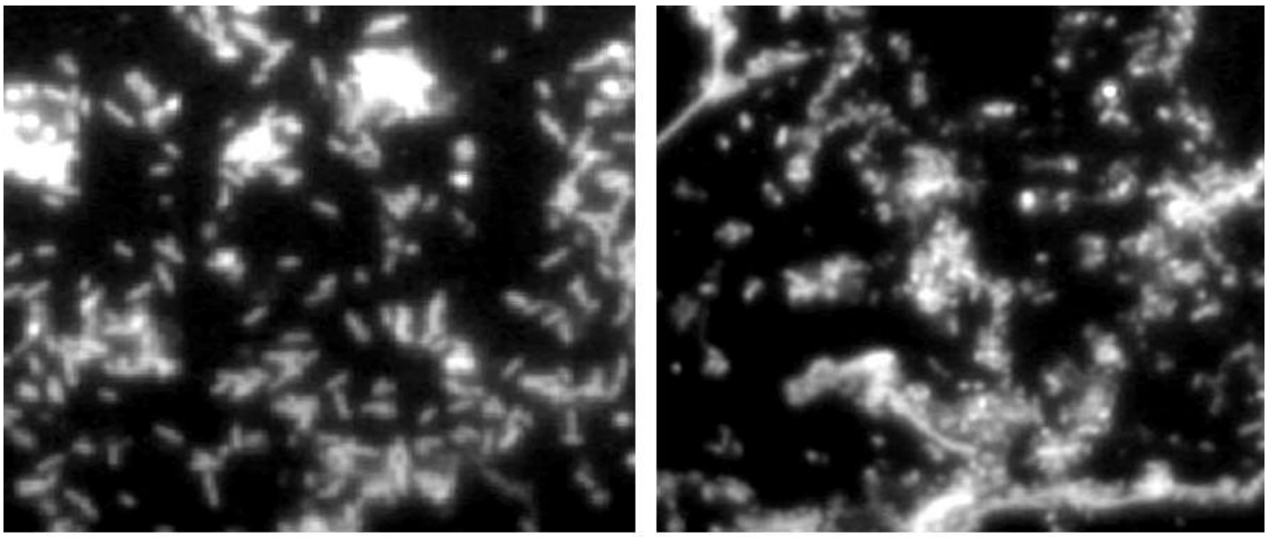



Figure 5: Effect of Feed Composition on the Specific Methanol Utilization Coefficient (circle: specific methanol utilization coefficient reported by others at a lower operating temperature (Barton et al. 1996); error bars represent the 90 % confidence intervals for measurements)

Tests using radio-labeled methanol indicated that although the "additional" microorganisms, from hereon referred to as partial-methylotrophic microorganisms, were not capable of growth with methanol as a sole substrate, they were capable of metabolizing methanol as an energy source. This competition for the available methanol reduced the concentration of methylotrophic microorganisms and increased the concentration of partial-methylotrophic microorganisms in the MBR as illustrated in Figure 7.

A relationship describing the overall removal of methanol in the MBR, presented in Equation 2, was proposed and used to investigate the effects of the two groups of methanol-utilizing microorganisms in the MBR on the overall specific methanol utilization coefficient. From Equation 2 and the estimated concentrations of each group of methanol-utilizing microorganisms (Figure 7), the specific methanol utilization coefficient for the partial-methylotrophic microorganisms ($U_{P\text{-}M}$) was estimated to be approximately 0.29/day. This value is substantially lower than the specific methanol utilization coefficient measured for methylotrophic microorganisms only ($U_M$ = 0.84/day). These results indicated that when trace organic contaminants were present, more of the methanol was consumed by the less active partial-methylotrophic microorganisms, leaving less methanol available for the more active methylotrophic microorganisms. This in turn produced a lower overall specific methanol utilization coefficient of 0.59/day and a corresponding lower rate of methanol removal as illustrated in Figure 8. Again, the specific methanol utilization coefficient measured during the present study when treating real evaporator condensate at 60 $^{o}$C was substantially higher than those reported by others for conventional, lower temperature, biological treatment systems as illustrated in Figure 5 (Barton et al. 1996).



(a) (b)

Figure 6: Qualitative examination of microbial communities in MBR.
((a): 100% synthetic evaporator condensate in feed; (b): 100% real evaporator condensate in feed. Note: The shutter speed for (b) was less than for (a) because the larger microorganisms in (b) were larger and therefore brighter. Consequently, the smaller rod shaped microorganisms seen in (a) are not as clearly identifiable in (b).)

$$R_{B\text{-}MeOH_f} = U_M X_{M_f} + U_{P\text{-}M} X_{N_f} \qquad [2]$$

where: $R_{B\text{-}MeOH}$: overall rate of biological methanol removal (mg/L•minute)
X: concentration of microorganisms as mixed liquor volatile suspended solids (mg/L)
M: methylotrophic microorganisms
P-M: partial-methylotrophic microorganisms
f: fraction of real evaporator condensate in the feed.

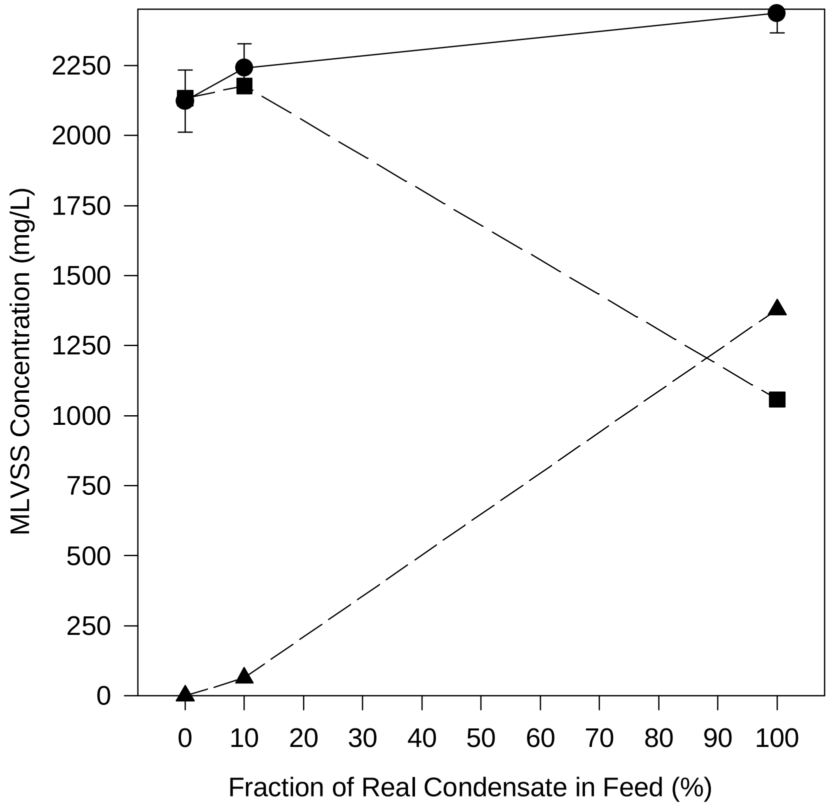



Figure 7: Estimated concentration of methylotrophic and partial-methylotrophic microorganisms in the MBR for different feed compositions.
(● and solid line: measured total MLVSS; ■ and long dashed line: estimated concentration of methylotrophic microorganisms; ▲ and short dashed line: estimated concentration of partial-methylotrophic microorganisms)

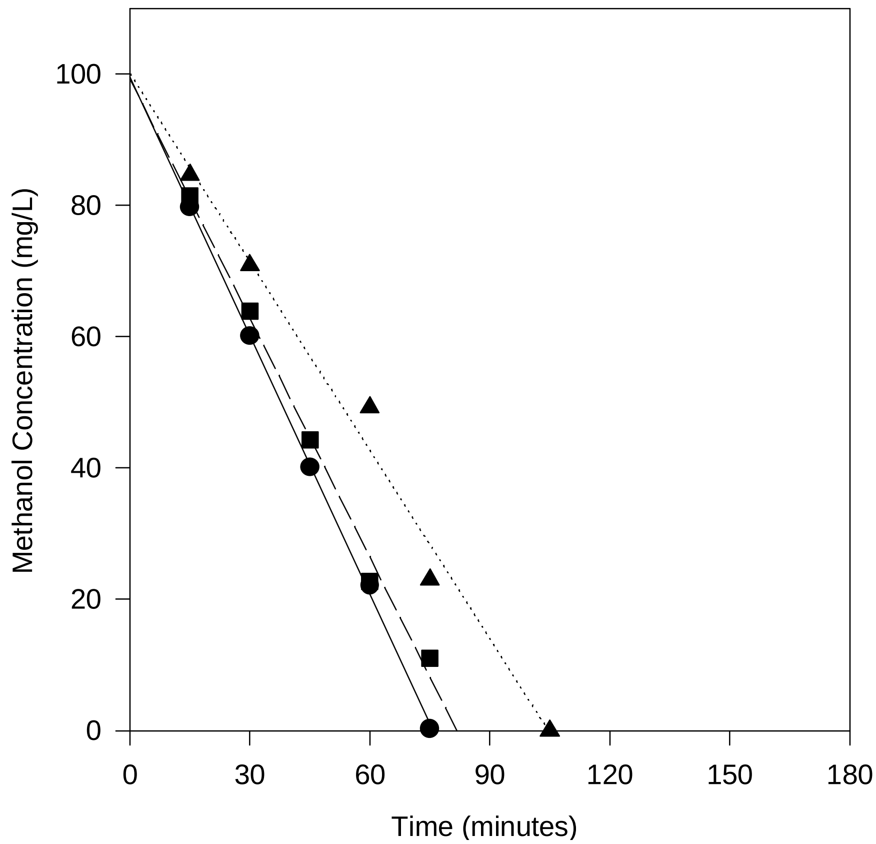



Figure 8: Methanol concentration in MBR during typical batch feed cycles for the different feed compositions investigated.

(●: 0 % real condensate in feed; ■: 10 % real condensate in feed; ▲: 100 % real condensate in feed; lines: Equation 4.4 fitted to the concentration of methanol in the MBR for the different feed compositions examined)

**Removal Efficiency Of Contaminants Of Concern During Treatment For Reuse Using A High Temperature MBR** (Bérubé and Hall 2000b)

High temperature biological treatment using an MBR successfully removed all of the contaminants of concern contained in evaporator condensate. Over 99 % of the methanol and RSC contained in the evaporator condensate was removed during high temperature treatment using an MBR. The concentration of methanol in the evaporator condensate was reduced from approximately 964 mg/L to below detection limits (0.5 mg/L). The concentrations of hydrogen sulphide, methyl mercaptan, dimethyl sulphide and dimethyl sulphide in the evaporator condensate were reduced from approximately 78, 79, 39 and 13mg/L, respectively, to below detection limits (approximately 0.4 mg/L). Approximately 93 % of organic compounds, measured as total organic carbon (TOC), contained in the evaporator condensate were removed.

The concentration of TOC in the evaporator condensate was reduced from 504 ± 137 mg/L to 52 ± 3.6 mg/L. Approximately 78 % of the reduction in TOC was due to the biological removal of methanol. The remaining 22 % of the reduction was due to the biological removal of trace organic contaminants. There were no suspended solids in the treated effluent. The removal efficiencies for the different contaminants are presented in Table 2.

Table 2 - Summary of fate of the contaminants of concern contained in evaporator condensate during high temperature biological treatment using an MBR

| **Influent**<br>504 mg/L | | | **Processes Occuring in Membrane Bioreactor** | **Effluent**<br>52 mg/L |
|---|---|---|---|---|
| Organic Contaminants | Other Organics | Particulate | Potential refractory compounds (removed with waste sludge)<br>Hydrolyzed to soluble products | (n.d.) |
| Organic Contaminants | Other Organics | Soluble | Potential refractory compounds<br>Biodegradable compounds consumed as substrate<br>Some refractory microbial products formed | 52 mg/L |
| Organic Contaminants | Methanol | | 91 % Oxidized to carbon dioxide and water<br>9 % Synthesized to biomass<br>Some refractory microbial products formed | (n.d.) |
| Organic Contaminants | Reduced Sulphur Compounds | DMS | > 99 % Stripped with off-gas | (n.d.) |
| Organic Contaminants | Reduced Sulphur Compounds | DMDS | > 99 % Stripped with off-gas | (n.d.) |
| Organic Contaminants | Reduced Sulphur Compounds | CH3SH | Abiotically oxidized (some to DMDS)<br>33 % Stripped with off-gas | (n.d.) |
| H2S | Reduced Sulphur Compounds | H2S | Abiotically Oxidized<br>3 % Stripped with off-gas | (n.d.) |

(mg/L as TOC; n.d.: non-detectable; H2S: hydrogen sulphide; CH3SH: methyl mercaptan; DMDS: dimethyl disulphide; DMS: dimethyl sulphide)

**Economic Evaluation Of High Temperature Biological Treatment Of Evaporator Condensate For Reuse Using An MBR** (Bérube and Hall 2000c,d)

Based on assumed removal efficiencies of 99, 90 and 99 % for methanol, TOC and RSC (as hydrogen sulphide and methyl mercaptan), respectively, as well as the characteristics of the evaporator condensate from a local kraft pulp mill, a conceptual design for a full-scale, high temperature MBR to treat an evaporator condensate for reuse was developed. Capital and operating costs were estimated and compared to the costs for a steam stripping system capable of achieving comparable contaminant removal efficiencies. Steam stripping is considered by many as the most applicable currently available technology to treat evaporator condensate for reuse. The capital and operating costs are resented in Tables 3 and 4, respectively.

Table 3 - Capital Cost Estimates

| **Cost Component** | **Cost** |
|---|---|
| **Membrane Bioreactor** | |
| Piping | 500 |
| Storage Tanks & Pumps | 180 |
| Chemical Addition | 65 |
| MBR Tank | 175 |
| Aeration System | 1,100 |
| Membranes | 1,300 |
| Civil/Electrical | 660 |
| **TOTAL** | **$3,980** |
| | |
| **Steam Stripping** | |
| Yard Piping | 500 |
| Storage Tanks and Pumps | 180 |
| Steam Stripper | 4,800 |
| Kiln Combustion System | 200 |
| Civil/Electrical | 660 |
| **TOTAL** | **$6,280** |

The capital cost for the MBR system was approximately 40 % less than the capital cost of a steam stripping system. The operating costs for the MBR system were also approximately 50 % less than the operating costs for a steam stripping system. The combined capital and operating cost of treating evaporator condensate for reuse using a high temperature MBR is equivalent to approximately $2.7 per ton of pulp produced (air dried metric ton – ADMT). By comparaison, the combined capital and operating cost for a steam stripping system is approximately $5.2 per ADMT produced.

Table 4 - Operating Cost Estimates

| Cost Component | Cost |
|---|---|
| **Membrane Bioreactor** | |
| Power | |
| Membrane | 0.34 |
| Aeration System | 0.12 |
| Chemicals | 0.25 |
| Labor | 0.60 |
| Equipment | 0.05 |
| **TOTAL** | **$1.36** |
| | |
| **Steam Stripping** | |
| Steam | 2.32 |
| Fuel Economy | -0.10 |
| Labor | 0.60 |
| Equipment | 0.15 |
| **TOTAL** | **$2.97** |

## MANAGEMENT APPLICATIONS

The results from the present study indicated that high temperature biological treatment of evaporator condensate for reuse using and MBR is technically feasible and economically attractive. For a relatively minimal cost (approximately $2.7 per ton of pulp produced), the raw water requirements can be reduced. This in turn can reduce the amount of wastewater to be treated in the existing combined mill effluent treatment system. This reduced load can potentially increase the efficiency and reduce the operating cost of the combined mill effluent treatment system and potentially reduce the impact of discharging treated mill effluent to the environment. This is of particular importance since potentially harmful contaminants have been identified in treated mill effluents (Karels et al. 1999).

Although further research is required, the present study is a significant step towards the ultimate goal of a zero effluent kraft pulp mill. A zero effluent mill would assist in ensuring the responsible and sustainable use of our natural resources.

# CONCLUSIONS

The present study was designed to improve our understanding of the physical, chemical and biological processes that occur during the high temperature biological treatment of evaporator condensate for reuse using an MBR, so that the fundamental questions presented earlier could be addressed. The significance of the results obtained during the present study, to environmental process engineering, is summarized as follows.

1. High temperature biological treatment of evaporator condensate is not only feasible but can be more efficient than conventional treatment technologies.
   - The contaminant removal rates measured during high temperature biological treatment were significantly higher than those reported for conventional, lower temperature, biological treatment systems (Barton et al. 1996).
   - The contaminant removal efficiencies measured during high temperature biological treatment were much higher than those reported for steam stripping systems (NCASI 1998).
   - All of the contaminants of concern were removed during treatment using a high temperature MBR.
   - High temperature treatment for reuse allows the heat content of the evaporator condensate to be recovered.

2. Based on the kinetic information collected, a model was developed to simulate the effect of elevated operating temperatures on a biological treatment system.
   - The model can be used to optimize the operation of a high temperature MBR for the treatment of evaporator condensate for reuse. Based on the model, the optimal operating temperature for the treatment of evaporator condensate for reuse was determined to be 60 $^{o}$C. However, care must be taken not to exceed this optimal temperature. At temperatures above 60 $^{o}$C, the efficiency of the system declines significantly. Some provisions for maintaining the temperature in the MBR at or below 60 $^{o}$C may be required.

3. The rate of methanol removal in the MBR was significantly impacted by the presence of trace organic contaminants in evaporator condensate. As the ratio of the amount trace organic contaminants to methanol in the feed to the MBR increased, the specific methanol utilization coefficient decreased. This ratio is likely to increase if the evaporator condensate stream is mixed with other waste streams from the pulp mill before treatment. Considering that the removal of methanol is a primary treatment objective, the evaporator condensate should be treated for reuse separately from other waste streams to maximize the rate of methanol removal.

4. A high temperature MBR successfully removed all of the contaminants of concern from the evaporator condensate.

- Regardless of the variations in the characteristics of the evaporator condensate to be treated, the quality of the treated effluent was constant.
- Over 99 % of the methanol, RSC (hydrogen sulphide and methyl mercaptan) and suspended solids and approximately 93% of the organic contaminants, measured as TOC, were removed during high temperature biological treatment using an MBR.
- Only a residual amount of refractory organic material remained in the treated evaporator condensate. It is unlikely that this residual amount of organic material would affect the pulping process or pulp quality (Niemela et al. 1999).
- There were no suspended solids in the treated effluent.

5. High temperature biological treatment is economically more attractive than the currently favored treatment technology (i.e. steam stripping).
   - Capital and operating cost analysis indicated that the treatment of evaporator condensate for reuse using a high temperature MBR is substantially less expensive than treatment using steam stripping. Treating the evaporator condensate for reuse would only increase the cost of pulp by approximately 0.04 % per ADMT produced. This cost appears to be insignificant considering the previously outlined benefits of treating and reusing the evaporator condensate.
   - The operating cost can be partially offset by the savings associated with recovering the heat content of the treated evaporator condensate.
   - The observed growth yield was measured to decreased as the operating temperature increased. Therefore, at higher temperatures, less excess sludge will be produced, potentially resulting in lower waste sludge handling and disposal costs.

## REFERENCES


ACGIH - American Conference of Government and Industrial Hygienists. 1999. Threshold Limit Values for Chemical Substances and Physical Agents and Biological Exposure Indices. Cincinnati. USA.

Annola T.L., Hynninen P. and Henricson K. 1995. Effect of condensate use on bleaching. Paper and Timber. 77(3): 111-115.

Barton D.A., Lee J.W., Buckley D.B. and Jett S.W. 1996. Biotreatment of kraft mill condensates for reuse. In Proceedings TAPPI Minimum Effluent Mills Symposium. Atlanta. USA. pp. 277-287.

Bérubé P. R. 2000. High Temperature Biological Treatment of Foul Evaporator Condensate for Reuse. Ph.D. Thesis. Department of Civil Engineering. The University of British Columbia. Vancouver. BC.

Bérubé P. R. and Hall E.R. 1996. Review of closed cycle technologies for kraft pulp and paper mills. Presented at First Forest Products Management Conference. Edmonton. Alb.

Bérubé P. R. and Hall E.R. 1999. Effects of kraft evaporator condenate matrix on methanol removal in a high temperature membrane bioreactor. Water Science and Technology. 40(11/12): 327-335.

Bérubé P. R. and Hall E.R. 2000a. Effect of temperature on methanol removal kinetics from synthetic kraft pulp mill condensates using a membrane bioreactor. Water Research. (in press).

Bérubé P. R. and Hall E.R. 2000b. Fate and removal kinetics of contaminants contained in evaporator condensate during treatment for reuse using a high temperature membrane bioreactor. In Proceedings PAPTAC Annual Meeting. Montreal. PQ. pp. B67-B72.

Bérubé P. R. and Hall E.R. 2000c. Treatment of recovery cycle condensate using a high temperature membrane bioreactor: Determination of maximum operating temperature and system cost. Pulp and Paper Canada. 101(3): 54-58.

Bérubé P. R. and Hall E.R. 2000d. Cost comparison between a high temperature membrane bioeactor and a steam stripper for the treatment of foul evaporator condensate for reuse (in preparation).

Blackwell B.R., MacKay W.B., Murray F.E. and Oldham W.K. 1979. Review of kraft foul condensates. TAPPI Journal. 62 (10): 33-37.

Durham V.E. 1991. High purity boiler condensates can serve as energy, reduce cost. Pulp and Paper. 65(1): 87-91.

Karels A., Soimasuo M. and Oikari A. 1999. Effects of pulp and paper mill effluents on reproduction, bile conjugates and liver MFO activity in fish at Southern Lake Saimaa, Finland. In Proceedings 6th IAWQ Symposium on Forest Industry Wastewaters. Tampere. Finland. pp. 123-132.

Jain A. K. 1996. Impact of water system closure on HAP and VOC emissions from process vents. In Proceedings TAPPI Minimum Effluent Mills Symposium. Atlanta. USA. pp. 69-78.

Jappinen P., Kangas J., Silakoski L. and Savolainen H. 1993. Volatile metabolites in occupational exposure to organic sulphur compounds. Archieves of Toxicology. 67: 104-106.

Jett S.W. 1995. Effect of shower water methanol content on emission from process vents. In Proceedings NCASI Southern Regional Meeting. pp. 66-75.

Kangas J., Jappinen P. and Savolainen H. 1984. Exposure to hydrogen sulphide, mercaptans and sulphur dioxide in pulp industry. American Industrial Hygiene Association Journal. 45(12): 787-790.

Leech M. and Chung L.T.K. 1982. Gas concentrations and occupational health in kraft mills. TAPPI Journal. November: 95-98

Mannisto H.,Mannisto E. and Krogerus, M. 1996. Current environmental performance of the pulp and paper industry. In Proceedings TAPPI Minimum Effluent Mills Symposium. Atlanta. USA. pp. 9-15.

NCASI. 1994a. Kraft Mill Condensate Treatment System Conceptual Design And Order-Of-Magnitude Cost Estimate. Technical Memorandum Npe39530.A0.

Ncasi. 1994b. Volatile Organic Emissions From Pulp and Paper Mill Sources: Part I- Oxygen Delignification Systems. Technical Bulletin No. 675.

Ncasi. 1994c. Volatile Organic Emissions From Pulp and Paper Mill Sources: Part Ii- Lime Kilns, Smelt Dissolving And Miscelaneous Causticizing Area Vents. Technical Bulletin No. 676.

Ncasi. 1994d. Volatile Organic Emissions From Pulp and Paper Mill Sources: Part Iv- Kraft Brown Stock Washing, Screening and Reject Refining Sources. Technical Bulletin No. 678.

Ncasi. 1994e. Volatile Organic Emissions From Pulp and Paper Mill Sources: Part V- Kraft Mill Bleach Plants. Technical Bulletin No. 679.

Ncasi. 1994f. Volatile Organic Emissions From Pulp and Paper Mill Sources: Part Vii- Pulp Dryers And Paper Machines at Intergrated Chemical Pulp Mills. Technical Bulletin No. 681.

Ncasi. 1998. Pulp And Paper Mill In-Plant and Closed Cycle Technologies – A Review Of Operating Experience, Current Status, and Research Needs. Technical Bulletin No. 557.

Niemela N., Saunamaki R and Rasimus R. 1999. Studies on the use of black liquor evaporation condensate at different bleaching stages. PAPRICAN Conference Report CR507: Heilights of Cellucon ’98. pp. 18.

Pekkanen M., Kiiskila E. 1996. Options to close the water cycle of pulp and paper mills using evaporation and condensate reuse. In Proceedings TAPPI Minimum Effluent Mills Symposium. Atlanta. USA. pp. 229-243.

Sebbas E. 1987. Reuse of kraft mill secondary condensates. TAPPI Journal. 70 (7): 53-58.

Venkatesh V., Lapp W.L. and Parr J.L. 1997. Millwide methanol balances: Predicting and evaluating HAP emissions by utilizing process simulation techniques. TAPPI Journal. 80 (2): 171-175.

Vice K. and Carroll R. 1998. The Cluster Rule: A summary of phase I. TAPPI Journal. 81 (2): 91-98.

Vora V. and Venkataraman B. 1995. The case for steam stripping of foul condensate streams. Papermaker. December: 36-39.