THE KNOWN EFFECTS OF PULP AND PAPER

MILL EFFLUENTS AND THEIR CONSTITUENTS

ON ESTUARINE AND MARINE ENVIRONMENTS

IN CANADA: A BRIEF REVIEW

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# TABLE OF CONTENTS

		PAGE					
1.	INTRODUCTION	1					
	1.1 Overview of Inputs and Impacts	1					
2.	BIOCHEMICAL OXYGEN DEMAND	2					
	<ul><li>2.1 BOD Loading</li><li>2.2 Fate</li><li>2.3 Biological Effects of Reduced Dissolved Oxygen</li></ul>	2 2 3					
	2.3.1 Port Alice, British Columbia 2.3.2 Port Alberni, British Columbia 2.3.3 Gold River, British Columbia 2.3.4 L'Etang Estuary, New Brunswick 2.3.5 Other Locations in British Columbia 2.3.6 Other Locations in the Atlantic Provinces	3 4 5 5 5 6					
3.	TOTAL SUSPENDED SOLIDS	6					
	<ul><li>3.1 Loading</li><li>3.2 Fate</li><li>3.3 Biological Effects of Effluent Solids</li></ul>	6 6 7					
4.	TOXICITY: ACUTE LETHALITY	8					
	<ul><li>4.1 Loading</li><li>4.2 Biological Effects of Effluent Toxicity</li></ul>	8 8					
5.	IMPACT OF ORGANOCHLORINE SUBSTANCES	10					
	<ul><li>5.1 Loading</li><li>5.2 Fate</li><li>5.3 Biological Effects</li></ul>	10 10 11					
6.	SUMMARY	14					
7.	RECOMMENDATIONS						
В.	REFERENCES	16					
	TABLES 1 TO 3						
	FIGURES 1 TO 6						

#### 1. INTRODUCTION

This summary document, based largely on an extensive report (Colodey et. al. in prep.) and numerous specific reports and reviews, (such as McLeay 1987, and Sprague and Colodey 1989), summarizes from a national perspective the serious environmental effects of pulp and paper mill effluents discharged to estuarine and marine waters in Canada.

The impact of each federally regulated parameter is addressed: Biochemical Oxygen Demand (BOD), Total Suspended Solids (TSS) and Toxicity. Known field impacts in Canadian marine waters are described briefly. In addition, the effects of organochlorine substances present in some effluents are described.

Where readily available, some information on freshwater impacts has been included; however, this has not been exhaustive and could be supplemented by persons with access to current information from Quebec, Ontario and the Western Provinces.

#### 1.1 Overview of Inputs and Impacts

Data for each of six parameters from B.C. coastal mills in 1987 is summarized in Table 1a. Data from Atlantic region mills for 1988 are in Table 1b.

Pacific coastal waters received 336 t/d of BOD and 135 t/d of TSS in 1987, in effluents that at source were usually acutely toxic to fish. Atlantic coastal waters received 254 t/d of BOD and >127 t/d of TSS in 1988, in effluents that at source were either non-acutely toxic to fish or very toxic to fish.

The locations of the 10 mills discharging to estuarine and marine areas on Canada's west coast are depicted in Figure 1. Measurable impact has been documented at every coastal mill. Changes in mill process (e.g. sulfite recovery, or switching from sulfite to kraft) and changes in discharge mode (from surface outfall to submerged diffuser) have led to environmental improvement at many locations on the coast. In contrast, other mill changes, such as increased loading due to production increases, or reduced assimilative capacity via natural factors (such as reduced river runoff) are threatening the fishery resources at other locations.

Mill locations in the Atlantic Provinces are shown in Figure 2. Measurable impacts have been documented at a minimum of eight of the mills. Most of the mills are older ones, that in their original design, have not provided much treatment for their effluents prior to discharge (Waldichuk 1988). However, total daily discharge loadings from Atlantic mills have been reduced 60% for BOD and 51% for TSS from 1969 to 1984 (Eaton et. al. 1986). This has been achieved through a combination of process changes, in-plant controls and external treatment, mainly with treatment ponds (Waldichuk 1988).

Along the St. Lawrence River and Estuary, 29 mills were in operation in 1982 and collectively discharged about 703 t/d of BOD and 336 t/d of TSS, a decrease from levels discharged in 1973: 891 t/d BOD and 684 t/d TSS (Statistics Canada 1986). In the Saguenay Fjord, pulp wastes have been detected through measures of enhanced organic material in the sediments and have provided a historical record of the industries' activities (Smith 1988, In: MEQ in Canada 1989). Evidence of impact of these mills was not available for this summary.

#### 2. BIOCHEMICAL OXYGEN DEMAND

#### 2.1 BOD Loading

Pulp and paper mill effluent is a complex mixture of many substances including carbohydrates, lignins, organic acids and alcohols. Oxygen is reduced when bacteria consume or degrade these materials, BOD being a measure of this oxygen demand. The majority of effluent oxygen demand can be met either in a treatment system, or if the effluent is not treated, the demand will be met by consumption of oxygen from the receiving waters. Each day, the ten B.C. mills discharge over 330 tonnes of oxygen demanding wastes to coastal waters in 2 million cubic metres of effluent (Table 1a). Despite reductions of up to 56% between 1975 and 1980 at B.C. coastal mills (Kay 1989), the BOD load from all but one pulp mill exceeds the BOD loading of municipal sewage Sixteen mills in the from B.C.'s second largest coastal city, Victoria. estuarine and marine 254 t/day to the Provinces discharge Atlantic environments (Table 1b). Only one B.C. coastal mill has secondary treatment for about half its effluent, while 8 Atlantic mills (6 coastal, 2 inland) have secondary treatment.

#### 2.2 Fate

The impact of mill effluent BOD on dissolved oxygen levels is moderated by the assimilative capacity of the receiving environment at each location. For example, at Elk Falls, B.C., where the effluent is rapidly mixed and dispersed by strong currents in Discovery Passage, there is little or no change in dissolved oxygen levels, even at the outfall. At the other extreme, where effluent is discharged into relatively quiescent coastal estuarine inlets which have restricted circulation (Port Alice, Port Alberni, Gold River, L'Etang Estuary), wide-scale, low dissolved oxygen levels are chronic and become serious during certain times of the year. especially when inlet flushing rates are reduced due to low river runoff. Unfortunately, this is also a time of fisheries sensitivity due to migrating salmon stocks.

The Lake Utopia (semi-chemi-mechanical) Paper Mill, discharging into the L'Etang Estuary in southwestern New Brunswick, caused serious dissolved oxygen depletion in the upper estuary throughout the 1970's and 1980's. Although mill production levels decreased from 250 t/d to 200t/d, dissolved oxygen conditions in the receiving water remained the same - (often severely depleted)

from 1975 to 1985 (Wildish <u>et</u>. <u>al</u>. 1986). Recent modifications to convert the upper retention ponds to <u>fresh</u> water have resulted in reduced hydrogen sulphide production in the upper estuary, and perhaps reduced BOD in lower parts.

Mill effluent not only exerts an oxygen demand, it can also reduce oxygen supply in receiving waters. Effluent colour can shade phytoplankton, thus reducing re-oxygenation via photosynthesis, and thereby reduce primary productivity in the estuary (Parker and Siebert 1972). This has been demonstrated near mills at Crofton, Port Alberni, Powell River (Anderson 1983) and at Port Mellon and Woodfibre (Stockner et. al. 1975).

It is important to note that some significant improvements have been made to BOD loadings by some mills. For example, prior to the installation of secondary treatment at the Fraser's Inc. mill in Edmunston, New Brunswick, and the Georgia-Pacific mill in Woodland, Maine, depressed levels of dissolved oxygen in the Saint John River and the St. Croix River, respectively, were observed. Since the implementation of secondary treatment at these mills (1980 for Edmundston, 1977 for Woodland), significant increases in receiving water oxygen levels have been measured (Dafoe et. al. 1987, Eaton 1989).

#### 2.3 Biological Effects of Reduced Dissolved Oxygen

Lethal and sublethal responses of fish to decreases in dissolved oxygen include changes in behaviour, growth, swimming, respiration, fecundity, disease resistance, and feeding (Davis 1975, Birtwell 1989). These demonstrated sublethal effects are more difficult to document in wild populations, but may be more widespread and significant than dramatic, serious events like fish kills, which may only rarely be documented. It also should be noted that the toxicity of kraft mill effluent increases when dissolved oxygen levels decrease (Marier 1973, Sprague 1985). Specific examples of effects related to BOD loading are given below.

#### 2.3.1 Port Alice (British Columbia)

Following recovery of sulfite liquor in 1977, the zone of serious BOD impact in Neroutsos Inlet was reduced from 20 km from the mill, to about 2 km (Kay However, during September and October 1985, the zone of potentially lethal (to salmon) dissolved oxygen concentrations (DO <3 mg/L) extended over 5 km up and down-inlet from the pulp mill and included water depths from 0-20 m at some locations, Figure 3 (from Birtwell 1989). Measured dissolved oxygen levels fell below 1 mg/L (Colodey and Pomeroy 1986), and were thus inadequate for even some species of invertebrates (Miller et. al. 1988). No direct fish mortalities were observed during this time, although there were anecdotal accounts of non-salmonid mortalities, including herring and ratfish (Colodey However, fish mortalities and stressed fish were and Pomeroy 1986). demonstrated using in situ juvenile chinook salmon bioassays (Kruzynski, DFO, pers. comm. in Birtwell 1989). Distribution of migrating salmon caught by DFO test-fishing nets was positively correlated to ambient dissolved oxygen

concentrations (Colodey and Pomeroy 1986). The changes in maturation of these salmon were described by Birtwell (1989), who concluded that poor water quality blocked and hindered first-run salmon from entering their natal streams. He notes that fish entered the creek to spawn during increasing dissolved oxygen levels in inlet waters following strong winds and heavy rainfall.

In 1986 there were 14 reported fish kills in Neroutsos Inlet ranging from 26 ratfish up to 10,000 dead herring, or on another occasion 90% of all intertidal life in a 10 km distance. In five cases, mortalities are clearly related to low ambient dissolved oxygen (dissolved oxygen less than 1 mg/L), while others may have been due to various factors such as effluent toxicity, or upwelling of hydrogen sulfide gas from the decomposition of deposited fibres and wood wastes (Colodey file reports). Stucchi (1988) reviewed the physical oceanography, dissolved oxygen and BOD loading in Neroutsos Inlet and concluded that a substantial reduction in the daily BOD loading is required before Davis (1975) Level B dissolved oxygen levels can even be approached. (At Level B the average member of a species starts to exhibit symptoms of oxygen distress.) Improved treatment facilities to achieve 80% reduction in BOD are currently in the pilot stage of development at Port Alice.

#### 2.3.2 Port Alberni (British Columbia)

The situation at Port Alberni has been studied for many years since the modelling and BOD evaluation of the proposed sulfite mill by Tully in 1949. Waldichuk (1974) has reviewed the response of Alberni Inlet to increasing BOD loading as kraft mill production increased. He notes that there was no oxygen problem when the mill was producing 220 short tons per day of unbleached kraft pulp because of the low volume of waste. Dissolved oxygen problems were encountered when BOD loading rose in conjunction with increases in mill production. Although total production is now about 1300 tonnes per day, it should be noted that BOD loading has decreased from about 40 t/d in 1970, to present averages of about 9 t/d, due to installation of an aerated stabilization basin in 1970 which treats about half the mill's effluent. However, despite partial secondary treatment, weekly loadings of 14,500 kg/d were exceeded on 3 occasions in 1985 (Dyer 1986).

In <u>situ</u> bioassays at Alberni Inlet have demonstrated that acutely lethal conditions occur due to low dissolved oxygen levels and to effluent toxicity (Birtwell and Harbo 1980, Birtwell and Kruzynski 1989). Use of the water column by juvenile and adult salmonids is restricted, at Alberni Inlet, to the upper water layers (with highest effluent concentration) due to the low levels of oxygen in deeper waters. Recent studies on the impact of mill BOD (Colodey et. al. 1988, Birch 1989) have concluded that the Alberni Inlet is receiving more oxygen demanding wastes than can be balanced by oxygenation processes, leading to a polluted situation with low ambient dissolved oxygen. Mill studies corroborated the observations of declining dissolved oxygen levels within the inlet (Seacounsult 1989). Davis' Level A criteria are not even met in the surface water layer for 50% of the data points collected from 1977-1988 as shown in Figure 4 (Colodey et. al. 1988). Below 2 m median conditions were

such that a large portion of the population would suffer a severe deleterious effect if exposed beyond a few hours (Davis 1975, Level C).

#### 2.3.3 Gold River (British Columbia)

Effluent from the mill at Gold River is discharged to an inlet which is very deep and has restricted circulation. The dissolved oxygen minima in the upper 20 m water layer, often coincides with effluent concentration maxima at each station sampled (Colodey 1986, 1988 data). This lowered dissolved oxygen layer (1.5-3.0 mg/L) can extend up into shallower depths (e.g. 2 m), thus lowering the amount of usable fish habitat within the inlet (Colodey 1989 data).

#### 2.3.4 L'Etang Estuary, New Brunswick

Following the plant opening in 1970, the benthic changes in the Lower L'Etang Estuary associated with mill discharges stabilized by 1972-1975 (Wildish et. al. 1986). Water quality in the Lower l'Etang is adequate for aquacultural purposes as presently practiced. The multiple effects of aquaculture and effluent loading have resulted in some eutrophication, reduced diversity, and phytoplankton blooms and die-backs. The Upper l'Etang, however, became incapable of supporting benthos and fish shortly after mill start-up, due to the organic loading and resulting anoxia (Poole et. al. 1978, Wildish 1983). Extensive descriptions exist of the major effects on the upper part of this small estuary, including the development of anoxic or hypoxic bottom waters, hydrogen sulfide production, the rapid elimination or gradual disappearance of indigenous species, and colonisation by hypoxic-tolerant benthic organisms. Not to be forgotten are the long-term impacts of the mill's effluent on the bacteriological water quality, in terms of shellfish harvesting standards (Blaise and Legault 1973, Baxter 1972, Menon, unpubl. man.). L'Etang is closed to all bivalve shellfish harvesting. High levels of bacteria were detected in the treated effluent in 1979 (Menon and Baxter, This problem has been well documented in other Canadian unpubl. data). situations, freshwater and marine.

#### 2.3.5 Other Locations in British Columbia

At other sites localised, minor depressions in dissolved oxygen have been noted in the vicinity of effluent outfalls at Crofton (Colodey 1989 data, Colodey and Tyers 1987, Sullivan 1980), Port Mellon (Cross 1989), and Woodfibre (Western Pulp 1988). No dissolved oxygen depressions have been noted near the outfalls at Harmac (Sullivan 1980, Colodey 1989 & 1986 data), or Powell River (Sullivan 1980, Colodey 1983 data). Dissolved oxygen levels at Porpoise Harbour have improved since the sulfite mill was shut down (1976) and the diffuser brought on-line (1978) for kraft mill effluent (Kay 1989).

#### 2.3.6 Other Locations in the Atlantic Provinces

At the mouth of the Nepisiguit River in Bathurst, New Brunswick, surface water dissolved oxygen concentrations are reduced when effluents are temporarily backed up by rising tides and low river flows. No significant oxygen depletion occurs when the tide is falling and effluents are flushed out into the harbour (I.E.C. Beak Consultants Ltd. 1985).

At Cornerbrook, Newfoundland, dissolved oxygen was reduced to 75% saturation up to 2 km from the Cornerbrook Pulp and Paper Limited mill located on the Humber Arm (Moores 1989).

#### 3. TOTAL SUSPENDED SOLIDS

#### 3.1 Loading

Suspended solids associated with pulp and paper mill effluent are largely composed of cellulose fibres, wood chips, and bark fines, although a smaller portion of the solids can be inorganic constituents such as boiler ash and calcium carbonate. The range of TSS loading for B.C. coastal mills is from 4 to 29 tonnes per day (EP P&Y 1987 data) (Table 1a). Atlantic mills have loadings ranging from <1 to 30 tonnes per day (Table 1b).

#### 3.2 Fate

The long-term oxygen demand of mill effluent is related to its organic solids which are deposited on the seabed at many locations in British Columbia waters. At Port Alberni, for example, an elevation in oxygen demand was demonstrated for degraded sediments which covered about 40% of the harbour bottom and accounted for about 10% of the total oxygen demand in the inner harbour (Hodgins 1989b). Hodgins states that: "... if the solids mat were to decrease in size through better retention and disposal of solids, then ultimately the benthic BOD would decrease. Studies reported in the literature indicate that this would not be a rapid recovery process."

Suspended solids loading causes the loss of productive benthic habitat when discharged solids settle and smother benthic organisms. A typical black, anaerobic deposit is often sampled near the outfall of most coastal mills. These deposits are often in the form of a jelly-like fibre mat which can be from several centimetres to 15 meters in depth (Pomeroy 1983). The release of methane gas, hydrogen sulphide gas (Waldichuk 1983). and acids (Miller et al. 1979) along with organic contaminants (Section 5 below) are a serious threat to the receiving environment. The release of nutrients to the water column (Pearson 1982), although beneficial, is over-shadowed by benthic habitat degradation. Figure 5a is a photo taken from the PISCES IV submersible of a typical fibre mat area, while Figure 5b shows the loss of benthic habitat by coarse wood material from log-booming operations at Port Alberni.

Present zones of measurable impact range from about 0.5 km to 5.0 km from the mills, covering several square kilometres at most mills. For example, benthic degradation at B.C. coastal mills range from 1 to 8 square kilometres. Insufficient data are available to determine the rate at which solids deposition is encroaching on the benthic habitat, although it has been suggested that at some locations mats are in equilibrium (e.g. Harmac: Ellis and Ostrovsky 1983). Where fibre accumulations have been adequately sampled, we can conclude that their size is relatively stable and fluctuates from year to year (e.g. Crofton: Colodey and Tyers 1987).

In the Atlantic Provinces, the results of a monitoring program on the Humber Arm, Newfoundland, indicated that the major impact from the Cornerbrook mill effluents was the smothering of benthos by wood fibre for a distance of 2 km north and northeast of the point of discharge (LeDrew and Bennett 1989, Moores 1989). The active deposition in the Arm of coarse wood material, from logs drives and debarking, is now stopped (LeDrew and Bennett 1989).

Similarly, sediment cores taken in Liverpool Harbour, Nova Scotia, indicated that Organic Sediment Index values were elevated at stations up to 1 km from the outfall of the Bowater Mersey mill (Beak 1971), and a major benthic deposit of wood fibre has been present off the Stora Mill at Point Tupper, Cape Breton, for at least 20 years (Machell et. al., Unpubl. MS; M. Bewers, pers. comm.). As in British Columbia waters, it is expected that effects on benthos, even after deposition stops, will occur over many decades.

#### 3.3 Biological Effects of Effluent Solids

Resource species, as well as smaller invertebrates used as food sources, may be replaced by fewer kinds of less desirable, pollution-tolerant species when organic deposits change the characteristics of the sediment. Degraded sediment from Port Alberni and Port Mellon are toxic to amphipod crustaceans in laboratory bioassays (Chapman and Barlow 1984). Changes to invertebrate species diversity and abundance in response to increasing sediment organic content have been described for Porpoise Harbour, Port Alberni, Port Mellon, Port Alice, Harmac, Ocean Falls by McGreer (1984). Fish kills have been caused by upwelling of toxic gases from benthic deposits at Powell River (B.C. MOE; B. Moore, pers. comm.). Even at Elk Falls, where strong currents disperse the effluent, there are degraded sediments (wood chips and indication of pulp). Near the outfall, benthic species response is reduced abundance and diversity in comparison to the reference location (Morrow Engineering 1985).

The recovery rate following cessation or reduction of solids deposition is a function of site-specific sedimentation rates and larval supply, both of which are mediated by local current regimes. It may be decades until natural sediments cover the historical inputs (up to 15 m thick) which have accumulated at many sites, such as at Ocean Falls in British Columbia (Pomeroy 1983, Fournier and Levings 1982).

Reduction in TSS output through improved levels of effluent treatment is required to prevent further degradation and begin the recovery period as

quickly as possible. Since a large portion of hydrophobic organic chemicals (like dioxins) are associated with suspended solids (Muir  $\underline{et}$ .  $\underline{al}$ . 1985, Servos and Muir 1989), solids control could also be expected to  $\overline{limit}$  the release of particulate associated contaminants. Settling suspended particulate matter (SPM) is of great importance for the introduction of these contaminants to a variety of organisms, where uptake is mainly by ingestion of SPM (Broman  $\underline{et}$ . al. 1989).

#### 4. TOXICITY: ACUTE LETHALITY

#### 4.1 Loading

Acute bioassay tests are used, in accordance with federal regulations, to evaluate the lethality of the mixture of toxic compounds found in mill effluents.

One review (Sprague and Colodey 1989, using data from McLeay 1987) calculated that untreated kraft mill effluent had a 96 h LC50 of 16% (killed 50% of the test fish when exposed to 16% effluent for 96 h). Only one B.C. coastal mill has secondary treatment for about half its effluent, and as a result B.C. marine discharge mills in toto have rather toxic effluents (median LC50 = 32%, Table 1a), in relation to B.C. freshwater discharge mills where secondary treatment is required (all LC50 = 100%). On the east coast, 7-? mills have secondary treatment and mills have final effluent toxicities ranging from 96 h LC50's of 2% to >100% (Table 1b).

In the early 1980's, 3 mills in northern New Brunswick were studied extensively (Boudreau et. al. 1988). "Consolidated-Bathurst effluent samples, mixed from four sewers in proportion to flow, were found to have 96 h LC50's to Atlantic salmon parr of 32-56%, 10-32% and 0-10%. Miramichi Pulp and Paper effluent samples had LC50's of 56-100% and greater than 100%. Effluent samples from Atholville Pulp had LC50's of 10-18% and 10-32%" (Boudreau et. al. 1988).

Well designed and operated secondary treatment facilities are an effective way to eliminate effluent toxicity of kraft mill effluent, largely by the decomposition of resin and fatty acids (Sprague and Colodey 1989). Secondary treatment can also be expected to reduce the loading of some chlorinated organic compounds (Gergov et. al. 1988, Fleming et. al. 1990, inter alios).

In the Atlantic Region by 1984, eight mills of 13 being modernized, produced effluents which met the toxicity requirements of the Pulp and Paper Effluent Regulations, and were non-acutely toxic to test fish (Waldichuk 1988).

#### 4.2 Biological Effects of Effluent Toxicity.

The aquatic toxicity of pulp and paper mill effluents has been extensively reviewed by McLeay (1987) and Sprague and Colodey (1989). Recently, in situ

field bioassays at Port Alberni and Port Mellon, British Columbia, demonstrated toxicity of partially treated and untreated BKME to several species of salmon (Birtwell and Kruzynski 1989). The reader is also referred to Section 5.3 for more information on effluent toxicity, particularly as it relates to organochlorine substances.

Even when effluent passes the acute bioassay test and is deemed "non-acutely toxic", it is possible that it can cause other detrimental effects. Examples from the freshwater environment illustrate the point. A study near the Proctor and Gamble bleached kraft mill at Grand Prairie (Alberta), which discharged treated non-toxic effluent, showed that downstream fish had accumulated chlorinated effluent compounds and had liver damage when compared to upstream fish (AEC 1987). A recently published study by Munkittrick (1989) demonstrated that fish exposed to bleached kraft mill effluent had elevated liver enzyme activity, coupled with reduced steroid hormone levels. These metabolic changes were associated with reduced sexual development in male and female whitefish. These effects on liver enzyme activation have also been demonstrated in fish exposed to BKME on the Fraser River (Servizi et al. 1990), Thunder Bay (Smith and Rokosh 1989), St. Maurice River (Hodson et al. 1990), Finland (Lindstrom-Seppa 1989), and Sweden (Andersson et al. 1988).

The above discussions on toxicity have focused primarily on the toxicity of whole effluents to fish. However, all ecosystem components must be protected For example, Anderson (1983) has demonstrated that from effluent toxicity. copepods, which are an important part of the food chain of many species, are more sensitive to kraft mill effluent (LC50 = 12%) than juvenile salmonids (LC50 = 25%). The toxic effect of bleached kraft mill effluent has also been demonstrated using sea urchins (Cherr et. al. 1989). Pulp mill effluent chlorate levels rise when chlorine dioxide is substituted for chlorine gas. chlorate has been shown to be toxic to brown algae (Fucus vesiculosus) at 10-20 ug/L when ambient nitrate levels were low (Lehtinen et. al. 1988). Chlorine dioxide, itself, is two to four times more toxic than total residual chlorine to freshwater fish (Wilde et. al. 1983), and has a 96 h LC50 of 20 ug/L for juvenile fathead minnows. However, effects of chlorine dioxide on marine organisms occur only at relatively high concentrations: kelp germination and sea urchin malformations at 250 mg/L (Hose et. al. 1989).

Wu and Levings (1980) demonstrated that blue mussels and barnacles suffered reproductive impairment and slower growth rates when transplanted near the Port Mellon kraft mill outfall. The authors note that the results of their study correlated well with a previous study at the location which found low densities of barnacles and mussels near the outfall (Levings and McDaniel 1976).

In the Atlantic Provinces, three mills in New Brunswick created extensive effluent plumes in the receiving estuaries. During rising tides, the Nepisiguit River had concentrations ranging from 1 to 10% effluent across most of its width and greater concentrations near the sewers. Under similar tidal conditions, the Miramichi River had 1 to 5% effluent across its entire width. The Restigouche River, however, had areas on its north side where effluent could not be detected at any part of the tidal cycle (Boudreau et. al. 1988).

Behavioural studies in the laboratory with Atlantic salmon parr showed that effluents from three mills (Consolidated-Bathurst, Miramichi Pulp and Paper, Atholville Pulp) at high concentrations (10% and 100%) increased their locomotor activity. Behavioural (aberrant?) responses occurred in 10% effluent concentrations for all mills, and concern was expressed that salmon migration might be affected in the receiving estuaries (Boudreau et. al. 1988).

In a follow-up 1985 study by IEC Beak at the Consolidated-Bathurst mill in Bathurst, New Brunswick, it was shown that juvenile Atlantic Salmon would avoid mill effluent at a concentration of 3.5%. Under low flow river conditions, a plug of effluent at, or above that concentration was predicted to form in the freshwater layer at hightide and to flow downstream until low tide was reached (I.E.C. Beak 1985).

#### 5. IMPACT OF ORGANOCHLORINE SUBSTANCES

#### 5.1 Loading

The impacts and toxicity of pulp mill related organochlorine substances have been recently reviewed (Colodey 1989, Sprague and Colodey 1989). About 200 low molecular weight chlorinated compounds have been identified in kraft bleach plant wastes (Reeve and Earl 1988, Suntio et. al. 1988, McKague et. al. 1989). This represents only a portion of the total number of chlorinated compounds represented as AOX (Adsorbable Organohalogens). A single large kraft mill may discharge around 4 tonnes of organically bound chlorine or about 50 t/d chlorinated organic substances (Sprague and Colodey 1989). AOX data for B.C. coastal mills (Table 2) indicates organically bound chlorine loadings of 1-26 t/d, compared to Atlantic Region mills which discharge from 1-5 t/d (Table 1b).

As described by Waldichuk (1988), the locations of pulp mills using chlorine bleaching in the Atlantic Region are shown in Figure 6. Kraft pulp mills in Newcastle, Saint John, Mackawic and New Glasgow each produce about 580 tonnes of pulp per day and use a total of about 200 tonnes of chlorine. The three sulphite mills located at Atholville, Edmunston and Point Tupper produce a total of 1000 tonnes of pulp per day and use about 55 tonnes of chlorine. The untreated effluents from these mills have the potential for introducing several thousand tonnes of chlorinated organics into the receiving waters annually.

#### 5.2 Fate

Once discharged into the environment, these compounds can become sorbed to particles (Broman et al. 1989, Servos and Muir 1989), dispersed and contaminate a large area. Chlorinated organics from pulp mills on the Fraser River were detected in water and fish in the Fraser Estuary up to 750 km downstream (Carey and Hart 1988, Rogers et. al. 1988a, Rogers et. al. 1988b, Servizi et. al. 1988). The linear dispersion distances of dioxins,

chloroguaiacols and chlorocatecols from B.C. coastal mills is shown in Table 3. Just as dispersion distance depends on site-specific variables, recovery of contaminated sediments by burial with clean sediment will also be related to local sedimentation and erosion rates, and the degree to which the sediments are modified by local communities (bioturbation). It has been demonstrated that levels of 2,3,7,8-TCDD (dioxin) in contaminated sediment from Newark Bay, N.J., have declined considerably over the past 25 years; however, some areas with low net sediment accumulation and sediment mixing can cause surficial sediments to have the highest contaminant concentration (Bopp et. al. 1988). These sediments would therefore be available as a biological contact surface.

Other studies predict that food-chain uptake is the main bioaccumulation route for polychlorinated dibenzo-p-dioxins in aquatic environments, especially when water concentrations are low (Muir and Servos 1988, Servos and Muir 1988, Muir and Yarechewski 1988, Muir et. al. 1988).

Such food-chain models are recommended as the preferred method for calculating site-specific dioxin body burdens (Dudley and Wagner 1989). Although 2,3,7,8-TCDD fate has been studied in freshwater model ecosystems (Isensee and Jones 1975, Corbet et. al. 1983, inter alios), results are not available for marine and estuarine systems. Freshwater studies indicate that bioaccumulation of 2,3,7,8-TCDD occurred primarily through the food chain and secondarily through contact with contaminated sediment, when rainbow trout were exposed in the laboratory. The water-exposure route did not appear to make a significant contribution in this case (Batterman 1989).

The environmental fate and biological effects of these compounds is further complicated because some higher molecular weight compounds (e.g. chloroguaiacols) can be degraded and transformed into other more toxic compounds (e.g. chloroveratroles: Neilson et. al. 1984, Neilson 1989, Paasivirta 1987). Chloroveratroles and chloroanisoles bioaccumulate and have been linked to tainting in some species of fish (Paasivirta 1988).

A study of the fate of organochlorine compounds from the 750 ton/d bleached kraft mill at Saint John, NB (Bacon 1978, Bacon 1980, Bacon 1983) showed the presence of dichlorophenols, trichlorophenols, trichloroguaiacols and tetrachloroguaiacols in sediments, clam (Mya arenaria) extracts, tomcod and flounder liver, and confirmed the earlier results of Brownlee and Strachan 1977. Pentachlorophenol was markedly present in all animals examined, and may have been related to the use of wood preservatives. A wide range of concentrations (not given) were detected, all at presumably sublethal levels. Flounder, smelt and tomcod accumulated the compounds. in liver and fat (flounder), viscera (tomcod) and liver (smelt). respectively. Resin acids and fatty acids were only detected, in clams and tomcod from one location.

#### 5.3 Biological Effects of Effluent Organochlorine Compounds

The biological effects of chlorinated compounds found in pulp and paper effluents on marine and estuarine biota are not well known. Lab studies on the uptake and depuration of chlorinated phenols and guaiacols in mussels

agreed with field results which indicated the presence of these contaminants in biota from Saint John Harbour and the Northumberland Strait near New Glasgow. The uptake and depuration of these compounds were also studied using a small estuarine fish (Fundulus heteroclitus). Histological damage was demonstrated when fish were exposed to either 0.5 ppm trichlorophenol (TCP), or trichloroguaiacol (TCG) or 2.5 % bleachery effluent. Liver damage was only completely reversible in fish exposed to TCP and TCG, but not in fish exposed to 2.5 % bleachery effluent (Bacon 1980).

Recent studies on Fraser river juvenile chinook salmon exposed to treated BKME support the importance of the food chain uptake of dioxin, demonstrate liver enzyme activation (EROD induction 2.5 times control levels) and tentatively identify liver granulomas, as healing bacterial kidney disease (Servizi et. al. 1990; J. Servizi, pers. comm.). Fish growth and seawater acclimation were not affected by effluent exposure. Lab exposure of fish to treated BKME led to contaminant uptake (chlorophenols, chloroguaiacols, EOCL= extractable organochlorine) in proportion to effluent exposure. Field studies (Rogers et. al. 1989) indicated similar levels of organochlorine contamination in feral fish, but much higher levels of EROD induction (55 times control).

The effect of BKME on fish in the St. Maurice River (Quebec) show a similar pattern of response to effluent exposure as Fraser River fish. Liver AHH enzyme activity was elevated in exposed white suckers 5-10 fold at sites up to 95 km downstream. Changes in enzyme induction match changes in liver somatic index (LSI), hematocrit, serum glucose, serum protein and fin-ray asymmetry, and were all clearly related to effluent exposure. A decrease in gonad somatic index (GSI), for both sexes at the 95 km site indicate that sexual maturation was retarded, a suggestion supported by changes in serum hormone levels measured for both sexes. Too few northern pike were sampled to be conclusive, but they seem to demonstrate a similar pattern for AHH induction Similar reproductive changes were described by and LSI as white suckers. (1989) in Section 4.2 above. The authors conclude Munkittrick Scandinavian studies of BKME are applicable in Canada, at least in some cases (Hodson et. al. 1990; J. Carey, pers. comm.).

Another study has examined the effects of blue crabs consuming radio-labelled 2,3,7,8-TCDD (dioxin) contaminated clams (Cristini et. al. 1989). It was determined that the digestive gland (hepatopancreas) of the crab accumulated the highest concentrations of TCDD, which is consistent with data from B.C. organochlorine surveys (Dwernychuk 1988, EP data). Some 2,3,7,8-TCDD was detected in the fecal material which the authors interpreted as excretion, but may be a result of incomplete assimilation. In a separate experiment, crab were fed dioxin and furan contaminated (137 ppt) clams from Newark. N.J. (Cristini et. al. 1989). The physiology of these crabs was disrupted in that they had fewer molts and slower limb regeneration compared to crabs from the control area.

A clam transplant study (Cristini and Cooper 1988) was used to evaluate the bioavailability and physiological effect of dioxins and furans on the clam (Mya arenaria). Exposed clams (Elizabeth and Newark, NJ) accumulated dioxins and furans, had reduced length, width, and shell-meat ratios, exhibited

significant lesions in the digestive tract and hepatopancreas, and had lowered adenylate energy charge. The authors concluded that the environmental exposure to dioxins and furans could alter physiological processes. A later study demonstrated shell-thinning and a variety of lesions of the gill, kidney and digestive gland (Cooper et. al. 1989).

The subtle nature of the mode of action of some chlorinated compounds is perhaps best illustrated by the delayed mortality to rainbow trout fry exposed to 38 part per quadrillion 2,3,7,8-TCDD: fish died after a 28 day depuration phase which followed the 28 day exposure phase (Mehrle et. al. 1988). Other examples of delayed mortality in fish exposed to 2,3, $\overline{7}$ ,8- $\overline{TCDD}$  are cited by Muir and Yarechewski (1988).

The bioaccumulation of some of these compounds (dioxins and furans) to levels higher than public health standards has resulted in the restrictive use or closure of over 67,000 hectares of productive fisheries habitat in British Columbia. Although half-lives for some dioxin and furan congeners have been calculated for some species of freshwater fish like fathead minnows, trout (Niimi and Oliver 1986, Muir and Yarechewski 1988, Muir et. al. 1989), carp (Kuehl et. al. 1987) and yellow perch (Kleeman et. al. 1986), similar data for marine species of fish and invertebrates are unavailable. It is known that invertebrates accumulate dioxins and furans in marine environments with little or no selectivity due to metabolism, and that fish can metabolize non-2,3,7,8 congeners greater than 2,3,7,8 congeners (Norstrom and MacDonald 1989).

The prudent approach is to limit the entry of chlorinated compounds into the environment by instituting effluent controls on the chlorinated compounds as a class of substances, since the fate and biological effects of these compounds in largely unknown.

#### 6. SUMMARY

It is clear from the above documented summary, and the extensive recent reviews by McLeay (1987), and Sprague and Colodey (1989), that untreated pulp and paper mill effluents from Canadian mills are most often acutely toxic at source. They have had, and in most cases continue to have, marked deleterious effects on receiving waters due to toxicity, and high BOD and TSS loadings. Impacts on benthic and intertidal organisms, contamination of biota by a wide range of organochlorine compounds, and changes in colour, primary productivity and dissolved oxygen in the receiving waters, all have been demonstrated.

The sublethal effects of the lowered oxygen levels and suspended solids on the water column and bottom communities of organisms are understood, whereas the potential effects of major organochlorine contamination of the water, sediments and biota are not fully understood, especially under natural conditions.

A number of documented cases of effects (contamination, toxic effects, habitat disruption) on estuarine and marine organisms or their communities exist from both Pacific and Atlantic coasts. They need to be updated in many cases in light of current biomonitoring techniques and changes in mill processes and effluent treatment.

There is evidence of improved water quality for dissolved oxygen levels where effluents have been treated successfully.

Concern exists due to the expected long-term impacts of large accumulations of bark and other solids on inshore benthic habitats.

The long-term effects of pulp mill-generated organochlorine discharges on the receiving environment are largely unknown, but based on their properties, known persistence, results of toxicity studies in the laboratory, known bioaccumulation under natural conditions, and the findings of recent North American and Scandinavian studies which linked enzyme activation to reproductive changes in fish, such effects are a major concern.

There is no question as to the magnitude and seriousness of the biological impacts of pulp and paper mill effluents; they have been documented in Canadian waters for over three decades. Major new regulatory initiatives are completely justified to reduce the impacts of pulp and paper mill effluents and to allow for environmental recovery.

#### 7. RECOMMENDATIONS

Studies are needed to quantify the responses and describe the health of fish exposed to BKME in the marine and estuarine environments, using current diagnostic health and ecotoxicological techniques.

The sublethal effects of effluents and organochlorines can be partly assessed by new laboratory tests proposed under the changes to the federal regulations. However, hazard assessments and environmental effects monitoring at each mill site are needed to complete the accurate assessment of potential sublethal effects and length of time of bioaccumulation of organochlorine compounds. There is a clear need to reduce acute toxicity of effluents, especially in situations where the effluent is discharged to areas of shallow water or constricted areas through which migrating fish must pass.

In conjunction with field biomonitoring and hazard assessments, long-term bioassays are needed for assessing the effects of specific chlorinated compounds alone and in combination from pulp and paper effluents.

There is an urgent need to track Canadian mill effluent chlorate levels, as chlorine dioxide substitution rates increase, and also to evaluate its toxicity with local species as well as with standard test species.

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TABLE 1A

EFFLUENT AND PRODUCTION DATA FOR B.C. COASTAL MILLS IN 1987,
RANKED BY INCREASING PRODUCTION<sup>1</sup>

BOD	TSS	LC50	FLOW	TER <sup>2</sup>	PRODUCTION	
t/d*	 t/d	<b>%</b>	$m^3 \times 10^3/d$		t/d	
85	. 4	33	175	530	462	
24	11	11	76	691	620	
					647	
					699	
					1043	
					1103	
					1294	
					1751	
					1910	
52 	25	23 	250	1087	2039	
336	135	<del>-</del>	1971	6711	11568	
9-85	4-29	11-75	76-320	272-1655	462-2039	
	85 24 16 19 22 37 9 28 44 52	85 4 24 11 16 7 19 9 22 9 37 12 9 10 28 29 44 19 52 25	85 4 33 24 11 11 16 7 24 19 9 54 22 9 75 37 12 11 9 10 64 <sup>3</sup> 28 29 59 44 19 30 52 25 23	t/d* t/d % m³ x 10³/d  85	t/d* t/d % m³ x 10³/d  85	

<sup>1</sup> EP, P&Y Region 1987

<sup>2</sup> Toxicity Emission Rate Calculated by (100\*1/LC50) \*FLOW
 (see McLeay 1987)

<sup>3 4/5</sup> Tests Passed LC50 of 80%

<sup>\*</sup> Tonnes/day

TABLE 1b - EFFLUENT AND PRODUCTION DATA FOR THE ATLANTIC PROVINCES' COASTAL AND INLAND MILLS<sup>1</sup>

Mill	BOD	TSS  t/d	AOX  t/d	LC50	Flow	TER <sup>2</sup>	Production t/d
Atholville Pulp Inc.	20		2	20	09	300	340
Consolidated Bathurst	35	ហ	N/A	32	40	125	730
Fraser Inc. (F)	12	10	S	3.4	100	294	820
Irving Pulp and Paper	19	10	2	9	7.7	1280	750
Irving Tissue	N/A	, 1	N/A	>100	N/A	N/A	50
Lake Utopia Paper	17	2	N/A	12	9	50	300
Miramichi Pulp & Paper-Nelson	7	7	N/A	2	7	350	300
Miramichi Pulp & Paper-Newcastle	S	13	٣	>100	65	< <b>6</b> 5	790
NBIP Forest Products Inc.	24	12	N/A	38	61	305	930
Rothesay Paper Inc.	22	15	N/A	9	45	750	880
St. Anne-Nackawic (F)	м	ю	1	>100	29	<i>19</i> >	610
Bowater Mersey Paper Co. Ltd.	29	7	N/A	30	41	137	610
Canadian Keyes Fibre	N/A	<b>,</b> 1	N/A	>100	N/A	1	40
Canexel	4	4	N/A	19	m	16	310
Scott Maritimes Ltd.	ß	ю	1	>100	84	· 84	570
Stora Forest Products Ltd.	52	19	7	7	8.7	4350	920
Abitibi Price IncGrand Falls (F)	27	13	N/A	38	145	380	670
Abitibi Price IncStephenville	7	ю	N/A	o	33	370	450
Corner Brook Pulp & Paper Inc.	17	30	N/A	20	94	450	770
Totals	296		16		1015		10,840
Ranges	252	<1-30	1-5	2->100	3-145	<65-4350	40-930

1 - EP, Atlantic Region, 1988, Pulp and Paper Mills Sector Report 2 - Toxicity Emission Rate Calculated by (100 x 1/LC50) x Flow (see McLeay 1987) (F) - Inland Mill

TABLE 2 - AOX DATA FOR BC COASTAL MILLS

Mill	AOX (kg/adt) <sup>1</sup>	Loading (t/d) <sup>2</sup>
Crofton	4.4-7.1	8.4-13.7
Elk Falls	6.0	12.2
Port Mellon	5.8	3.8
Harmac	5.5-7.1	5.7-7.4
Port Alberni	5.8	7.5
Powell River	9.0-14.9	15.8-26.1
Prince Rupert	0.5-3.1	0.5-3.5
Port Alice	5.2	2.4
Woodfibre	3.7	2.3

<sup>1</sup> CPPA Data, Sept. 15, 1989

<sup>2 1987</sup> Production Data

TABLE 3

# DISTANCES (KM) FROM MILL OUTFALLS WHERE TRICHLOROGUAIACOL (TCG), TETRACHLOROGUAIACOL (TECG), TRICHLOROCATECOL (TCC),

TETRACHLOROCATECOL (TECC),

## AND 2,3,7,8-TETRACHLORODIBENZO-DIOXIN (2,3,7,8-TCDD)

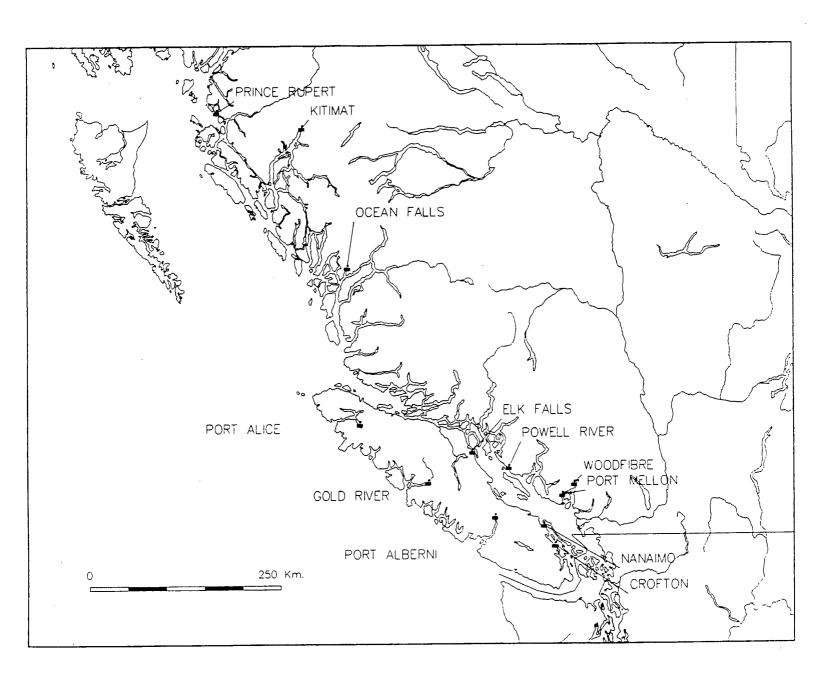
### HAS BEEN DETECTED IN SEDIMENT AND BIOTA

(DATA FROM DWERNYCHUK 1989)

Location	TCG	TeCG	TCC	TeCC	2,3,7,8-TCDD				
	Sediment		Sediment		Sediment	Crab	Shrimp	Mussels	
Prince Rupert	>6	>6	>6	>6	>6	>6	2	<1	
Woodfibre	5	10	8	10	4	>16	<1	<1	
Port Mellon	6	8	5	>10	>10	>22	1	<1	

FIGURE 1

LOCATION OF MILLS DISCHARGING TO ESTUARINE AND MARINE AREAS
ON CANADA'S WEST COAST (FROM KAY 1986).



LOCATIONS OF PULP AND PAPER MILLS IN THE FOUR ATLANTIC PROVINCES AND THE AMOUNT OF BOD (BIOCHEMICAL OXYGEN DEMAND) AND SUSPENDED SOLIDS CONTRIBUTED TO RECEIVING WATERS IN 1984 (FROM EATON ET. AL., 1986).

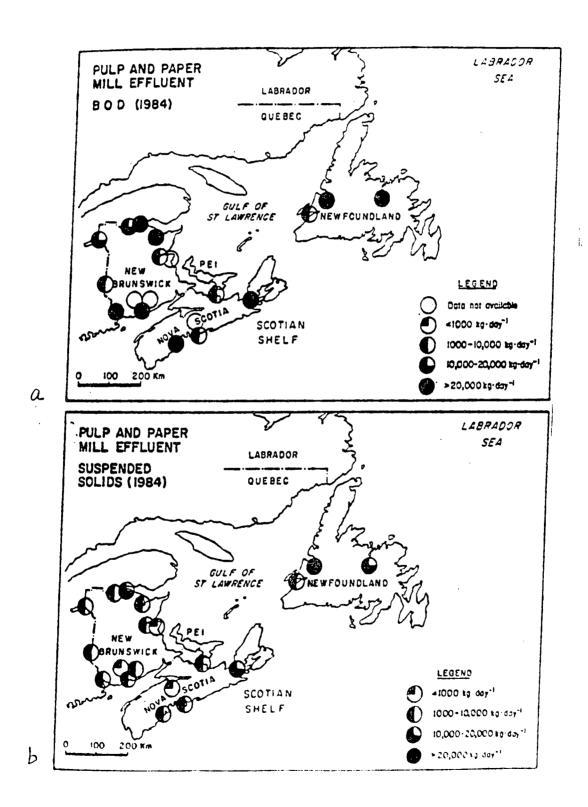


FIGURE 3 - AREA OF HYPOXIC WATER IN NEROUTSOS INLET, B.C.

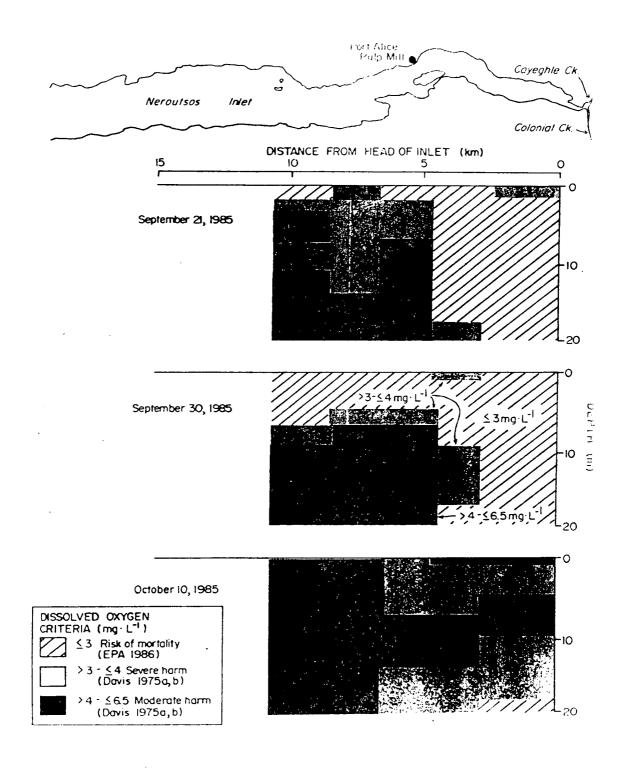
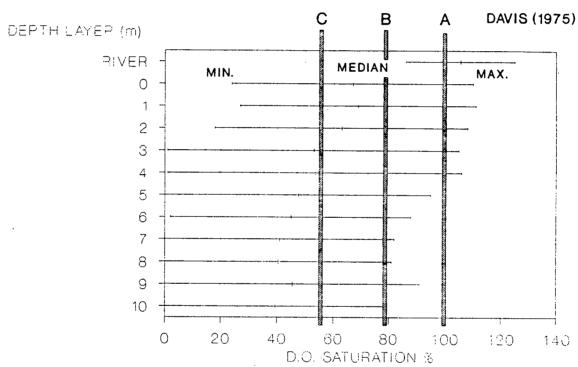


FIGURE 4

FREQUENCY OF DISSOLVED OXYGEN VALUES BELOW LEVELS A, B AND C (DAVIS, 1975) IN ALBERNI INLET, B.C.

# ALBERNI INLET HOLM IS. 1977-88



from Colodey et al. 1988

FIGURE 5a PLATE 1. APPEARANCE OF TYPICAL FIBRE DEPOSIT - NORTHUMBERLAND CHANNEL OFF HARMAC PULPMILL

FIGURE 5b PLATE 2. WOOD DEBRIS AND DEVELOPMENT OF WHITE BACTERIAL SLIME FOUND UNDER SORTING AND STORAGE AREAS - ALBERNI INLET IN VICINITY OF PORT ALBERNI PULPMILL

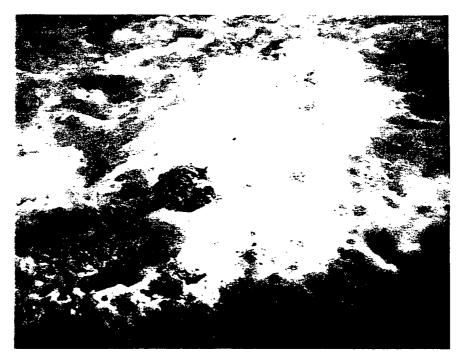
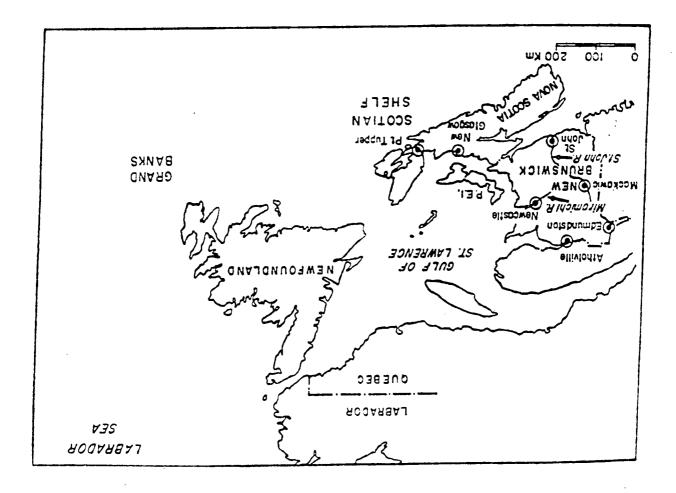


PLATE 1 Appearance of typical fibre deposit -Northumberland Channel off Harmac pulpmill

PLATE 2 Wood debris and development of white bacterial slime found under sorting and storage areas
- Alberni Inlet in vicinity of Port Alberni pulpmill





LOCATIONS OF PULP MILLS IN THE MARITIMES THAT USE CHLORINE BLEACHING (FROM EATON  $\overline{\text{ET}}$ . 1986).

The Known Effects of
Pulp and Paper Mill...

PP40.10