Cadmium Pollution of Belledune Harbour, New Brunswick, Canada

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CADMIUM POLLUTION OF BELLEDUNE HARBOUR, NEW BRUNSWICK, CANADA

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This is the one hundred and twenty-eighth Technical Report from
the Biological Station, St. Andrews, N.B.

PREFACE

The presence of the New Brunswick Mining and Smelting Corporation Ltd. (Smelting Division) operation in Belledune, New Brunswick, has, since its construction, represented a potential source of heavy metals to the Baie de Chaleur. Biological monitoring studies, both by the company and by the federal Departments of Environment and Fisheries, have maintained a continuing watch on the influence of smelter pollutants on the marine environment. The results of these studies showed elevated levels of heavy metals in the immediate area, but the effects were localized and the effects on the marine resources were not considered significant.

The Brunswick Smelting report documenting the 1979 biological monitoring activities in the Belledune area was made available to the Department of Fisheries and Oceans April 18, 1980. This report indicated that the concentrations of heavy metals, in particular cadmium, have been increasing at an accelerated rate in several marine resources. The area affected, as delineated by concentrations of cadmium in mussels "upstream" and below Belledune Harbour, was expanding. Most significantly, the levels of cadmium in lobsters in Belledune Harbour were becoming dangerous from a public health standpoint.

With the lobster season destined to open May 1st, rapid action was required. The Director General, Fisheries Management - Maritimes Region, declared that emergency procedures be instituted to investigate the situation and to develop recommendations regarding the exploitation of the lobster resource from the Belledune area. The Bedford Institute of Oceanography offered its assistance in the investigations. The Fisheries and Environmental Sciences Division of the Maritimes Resource Branch was identified as the lead organization to carry out the required scientific investigations on behalf of the Department.

The immediate research objectives were (1) to determine the geographic extent of cadmium contamination of lobster stocks in Belledune Harbour and adjacent vicinity and to delineate where elevated cadmium levels in lobster return to "baseline"; (2) to develop scientific evidence necessary to establish a "safe" level for cadmium in lobster; (3) to determine the extent to which the sediments were contaminated with cadmium and the effect of local currents and oceanic circulation on the distribution of cadmium in the area; (4) to evaluate the effect of cadmium on the other marine resources in the area. In addition to the above investigations, there remained the operational requirements to meet with fishermen's groups, to establish a controlled fishing zone where all lobsters would have to be specially processed, to encourage implementation of the planned improvements to the effluent control facilities at the smelter, and to develop, with Health and Welfare Canada, appropriate "tolerance" limits for cadmium in lobster.

This report represents the scientific data base that was developed in response to this emergency. It represents an important benchmark on the presence and distribution of cadmium in the Belledune area (against which future pollution control actions can be measured) and provides a better understanding of the fate and biological effects of cadmium on the lobster. The use of the information provided by this report and the continuing research efforts by Noranda Research on behalf of Brunswick Smelting and the effluent treatment facilities should ultimately lead to an improved marine habitat in the Belledune area. Strategies for the management of the Belledune lobster fishery in 1981 are already being planned; with sufficient lead time, there should be no undue disruption in the 1981 lobster fishery in the Belledune area.

It is important to take this opportunity to acknowledge the cooperation and assistance received from Brunswick Smelting and the Noranda Research Centre. The cooperation received from the Environmental Protection Service of the Department of the Environment and the New Brunswick Department of Environment is also acknowledged. A special note of gratitude is extended to the staff of the Area Manager, Northern New Brunswick, to D. G. Robinson of the Invertebrates and Marine Plants Division, and to the scientists of the Bedford Institute of Oceanography for assistance and contributions. Finally, I acknowledge the major efforts made by Division staff at the Halifax Laboratory and at the Biological Station for their professionalisms and dedication during these emergency investigations.

The Editors and I wish to acknowledge the assistance of Ms. Ruth Garnett for technical editing and Mrs. Brenda McCullough for typing and assembling the report.

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ABSTRACT

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Results of investigation of cadmium (Cd) pollution in Belledune Harbour, New Brunswick, Canada, in April-June 1980 are described. The investigation was carried out by the Department of Fisheries and Oceans in response to studies by Brunswick Mining and Smelting Corporation Limited (Smelting Division) that indicated an increase of Cd concentration in blue mussel (Mytilus edulis) and the American lobster (Homarus americanus) between 1977 and 1979.

Brunswick Smelting lead (Pb) smelter at Belledune is the source of Cd in Belledune Harbour and in the immediate vicinity. The smelter processes Pb concentrate and Pb/Cd dust into refined Pb by traditional smelting and refining techniques. Sources of Cd are emissions, surface runoff, slag pond discharge, and leaching of slag and Cd baghouse dust, containing Cd at about <0.01 and 9% respectively. The baghouse dust is recycled until the concentration of Cd reaches about 10% when it is bled off and shipped elsewhere for refining. Cd is readily leachable from both the slag and baghouse dust by seawater.

The exchange of water between Belledune Harbour and coastal flow in the Bay of Chaleur to the southeast appears to be limited to diffusion, but this may be changed by easterly or northeasterly winds.

The concentration of Cd in water within Belledune Harbour (two samples) was 125 µg/L near the smelter outfall, located at the east end, and 25.5 µg/L in the center of the harbour, and decreased to 0.1-2.0 µg/L outside the harbour. The concentration of Cd in sediment was 9-61 µg/g dry weight with the highest values in the western part and 0.4-1.0 µg/g outside the harbour. A band of contaminated sediment extended diagonally to the southeast to a distance of about 12 km from the mouth of the harbour. Belledune Harbour water and sediment were highly contaminated by zinc as well.

The concentration of Cd in biota had a similar pattern. Outside the harbour Laminaria digitata, Fucus vesiculosus, and Ascophyllum nodosum contained 0.4-1.8, 1.3-30, and 0.3-18 µg Cd/g wet weight, respectively. Cd content of Mytilus edulis, Modiolus modiolus, Cancer irroratus gill, coxal muscle, cheliped muscle, hepatopancreas, Littorina littorea, and Buccinum undatum was 7.9-112, 15.2-55.6, 1.7-53.5, 0.2-29.2, 0.1-8.1, 2.3-394, 1.1-44, and 0.4-138 µg/g dry weight, respectively. The concentration of Cd in polychaetes, flatfish viscera and muscle was 0.2-3.6, 0.3-2.0, and 0.02-0.4 µg/g dry weight, respectively. Lobsters within the harbour were highly contaminated. The geometric mean concentration of Cd in the hepatopancreas was 176 (N=29), 62 (N=28) and 24 µg/g wet weight (N=30) in lobsters from the western and eastern parts and just outside the harbour, respectively. At approximately 17 km from the mouth of the harbour to the southeast the concentration was 11.6 µg/g wet weight (N=31). Geometric means of Cd concentration in lobster hepatopancreas from various localities in eastern Canada range from 2.8 to 17.2 µg/g wet weight.

Studies of adenylate energy charge, ATPase activity, and histology do not indicate any adverse effects of Cd on lobsters from Belledune Harbour.

A review of the literature indicates that marine fauna accumulates Cd from seawater even if concentration of Cd is as low as 0.5 µg/L, and the excretion rate of Cd appears exceedingly low. Excretion of Cd from lobsters could not be achieved by feeding or injecting chelating agents. The availability of Cd to rats from contaminated hepatopancreas was only about 50% of that from a Cd-contaminated, casein-based diet.

Key words: cadmium, lead, zinc, heavy metals, trace elements, mining, smelting, American lobster, bivalve molluscs, crabs, Belledune

RÉSUMÉ

Uthe, J. F., and V. Zitko [Editors]. 1980. Cadmium pollution of Belledune Harbour, New Brunswick, Canada. Can. Tech. Rep. Fish. Aquat. Sci. 963, v + 107 p.

Les auteurs présentent les résultats d'une étude réalisée en mai et juin 1980 par le ministère des Pêches et Océans, portant sur la pollution par le cadmium du port de Belledune et basée sur l'information transmise par Brunswick Mining and Smelting Corporation Limited (Smelting Division) indiquant une augmentation de la concentration du cadmium chez la moule bleue, Mytilus edulis, et le homard, Homarus americanus, entre 1977 et 1979.

Brunswick Smelting exploite une jonderie de plomb à Belledune en bordure du port. La fonderie transforme un concentré de plomb et une poussière enrichie de plomb-cadmium au moyen des techniques traditionnelles de fusion et de raffinage. Le cadmium retrouvé dans le port et dans les environs immédiats provient des diverses opérations de la fonderie et plus particulièrement des émissions, du ruissellement de surface, des déversements du bassin de rétention des scories et finalement de la lixiviation des scories et des poussières de la chambre des filtres. Ces scories et ces poussières ont une tenuer respective en cadmium de <0,01 et de 9%. Les poussières de la chambre des filtres sont ensuite recyclées jusqu'à ce que la teneur en cadmium atteigne 10%; elles en sont alors retirées puis expédiées à d'autres raffineries. Des expériences ont démontré que le cadmium est lixiviable des scories et de la poussière au contact de l'eau de mer.

Les échanges d'eau entre le port et le courant côtier de la Baie Des Chaleurs, lequel est orienté vers le sud-est, semblent se limiter à une diffusion. Cependant, il est possible que des périodes de vents d'est ou du nord-est, modifie ce modèle.

Les concentrations de cadmium dans l'eau de mer (d'après l'analyse de deux échantillons) étaient de 125 µg/L près de l'effluent de la fonderie située dans la partie est du port et de 25,5 µg/L au centre de port hors duquel elles baissaient à 0,2-2,0 µg/L. Dans les sédiments, les concentrations variaient entre 9 et 61 µg/g dans le port, les maxima étant observés dans la partie ouest pour diminuer entre 0,4 et 1,0 µg/g à l'estérieur du port. Une bande de sédiments contaminés, orientée vers le sud-est, s'étend sur 12 km à partir de l'entrée du port. Les eaux et les sédiments du port étaient également fort contaminés par le zinc.

Pour ce qui est des concentrations de cadmium dans le biotope marin, on remarquait la même tendance. A l'extérieur du port, des échantillons de Laminaria digitata, Fucus vesiculosus, et Ascophyllum nodosum contenaient respectivement 0,4-1,8; 1,3-30 et 0,3-18 µg Cd/g de poids humide. Les concentrations en cadmium, exprimées en µg/g de poids sec du tissu analysé, ont été déterminées chez Mytilus edulis (7,9-112), Modiolus modiolus (15,2-55,6), Cancer irroratus (branchies: 1,7-53,5, muscles coxaux: 0,2-29,2, muscles des chélipèdes: 0,1-8,1, hépatopancréas: 2,3-394), Littorina littorea (1,4-44), Buccinum undatum (0,4-138), les polychètes (0,2-3,6), et dans les muscles (0,02-0,4) et viscères (0,3-2,0) des poissons plats. Les homards du port étaient fortement contaminés. Ainsi les moyennes géométriques des concentrations de cadmium dans l'hépatopancréas des homards échantillonnés dans la partie ouest et est du port étaient respectivement de 176 (N=29) et de 62 (N=30) µg/g de poids humide à l'extérieur du port dans sa périphérie immédiate. A 17 km de l'entrée du port, vers le sud-est, la concentration de cadmium était de 11,6 µg/g de poids humide (N=31). Chez les homards échantillonnés en divers endroits de l'est du Canada, les moyennes géométriques des concentrations en cadmium des hépatopancréas varient de 2,8 à 17,2 µg/g de poids humide.

L'étude de la charge énergétique des adénylates, de l'activité ATP-asique et des coupes histologiques d'hépatopancréas indique que le cadmium, aux concentrations observées, ne semble pas avoir d'effets nocifs chez le homard du port de Belledune.

D'après une étude bibliographique, la jaune marine concentre du cadmium même lorsque ce dernier est présent dans l'eau de mer à des niveaux aussi bas que 0,05 µg/L, et semble ne l'excréter que très lentement. Ainsi des expériences d'ingestion et d'injection d'agents chélateurs chez le homard n'ont pas réussi à provoquer une excrétion du cadmium. De plus, des expériences ont démontré que des rats nourris avec des hépatopancréas contaminés n'assimilent qu'environ la moitié du cadmium qu'ils assimilent d'un régime ca séinique enrichi de cadmium.

FISHERIES CONCERNS ASSOCIATED WITH THE CADMIUM PROBLEM AT BELLEDUNE

by

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The discovery of high cadmium (Cd) levels in lobsters taken from the Belledune Harbour area this year poses a significant resource management problem as well as environmental protection ramifications. While the most urgent problem that had to be addressed when the cadmium contamination became known was to document the extent and nature of the contamination, in view of the imminent opening of the lobster fishing season, the biological and ecological implications remain a continuing major concern to this department. Physiological and behavioral effects on lobsters and other marine species from high cadmium exposure will require further research and investigation before the extent of resource implications can be established. The more immediate and obvious effects, such as loss of income to fishermen due to area closures and potential impact on local and foreign markets, can be addressed in an operational fashion but long-term biological impact on marine organisms remains and is a much more complex and difficult problem to address.

The Belledune cadmium incident, being a localized point source problem, facilitated the application of stringent control measures to be applied within a limited area. Although a control zone extending outside the harbour was ultimately established, lobster fishing within this control zone was permitted and all catches within this area were subjected to special processing procedures under the supervision of departmental personnel.

The department was seriously handicapped in determining what level of cadmium was permissible from a human health aspect because no specific levels have been stipulated. Only after extensive consultation with Federal Health and Welfare personnel were permissible levels in lobster tomalley and muscle determined. These levels were based on extensive pre-analyses of lobsters taken from inside and outside Belledune Harbour and previously established background levels for the area. Based on this information a final control zone was established but not until the lobster fishing season was well under way. If permissible levels of cadmium, based on human health requirements, had been established earlier, the delay and frustration would have been minimized.

The lag period encountered for transmission of scientific data on the Belledune cadmium problem from industry to regulatory agencies and from regulatory agencies to the department resulted in a short lead time in which the department was able to react before the fishing season opened. This created serious internal administrative difficulties and remains a major concern to the department. Improved procedures for more timely exchange of environmental data having fisheries implications, and corrective measures to be adopted to prevent a similar situation from occurring in the future, are matters that require serious discussion with regulatory agencies and industry officials. A much more timely transfer of new information relative to possible fisheries implications is warranted.

The Brunswick Smelting plant situated adjacent to Belledune Harbour has been identified as the prime contributor of the elevated cadmium levels within and immediately outside the harbour. Recently, additional measures have been implemented by the owners to reduce cadmium and other heavy metals being released into Belledune Harbour. The success of these corrective measures can only be assessed after implementation but there is good reason to believe a significant improvement can be expected. A potential hazard exists with respect to existing high levels of cadmium in marine sediments and its availability to marine organisms. A solution to this continuing hazard is necessary although the options available to the company are rather limited.

INDUSTRIAL DISCHARGES OF CADMIUM AT BELLEDUNE

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INTRODUCTION

Since 1966 Brunswick Mining and Smelting Corporation Limited, Smelting Division (Brunswick Smelting) has operated a lead/zinc smelting and refining plant at Belledune. In 1971 the refining of zinc was terminated, but the lead smelting continued. The plant produces some 60,000 metric tons (MT) of lead, 3,600 MT of copper matte, lesser amounts of silver and lead-antimony alloys and 175,000 MT of sulfuric acid per year. This paper describes the operation of the smelting and refining and of the fertilizer plant at the same location, and their discharges to the Bay of Chaleur.

THE LEAD SMELTING AND REFINING PROCESS AT THE BELLEDUNE SMELTER

The plant at Belledune originally produced both lead (Pb) and zinc (Zn) by the Imperial Smelting Process (ISP) but, in 1972, the Belledune smelter was modified to produce only refined Pb and associated byproducts by traditional Pb smelting methods. The production of Pb increased by a factor of 2.5. This conversion was due to increased coke costs associated with the high temperatures required for Zn production, and to improved milling methods which permitted the separation of the complex Brunswick Mines No. 12 ore into separate Pb and Zn concentrates. The changeover and modifications were completed in 1972-73.

The process flow diagram has been presented by Dugdale and Hummel (1977); however, there have been some process modifications since then, mainly in the refining area. The smelting and refining process consists of several stages: charge preparation, sintering, blast-furnace smelting, and finally, refining to remove or recover metallic impurities and produce pure Pb.

THE Pb CONCENTRATE

In ores, cadmium (Cd) is closely associated with Zn and small amounts of Zn are present in Pb concentrates (Lymburner 1974) from the mining flotation processes. The boiling point of Cd is 767°C (Considine 1974) in contrast to the boiling point of Pb which is 1740°C (Considine 1974). Above these temperatures the metals will be present as vapors or "fumes."

Brunswick Mining produces a lead-zinc-copper-silver ore of which 80% is sulfides and 20% is quartz, calcite and silicate. Brunswick Mining concentrator produces Cu, Zn and Pb concentrates by a differential flotation process (Neumann and Schnarr 1971). Brunswick Smelting currently receives 225,000 MT of Pb concentrate from this source, plus minor amounts of purchased high-grade concentrate. The ~~past and~~ present analyses of mine concentrate are shown in Table 1.

Since 1976 approximately 4500 MT per annum of Cottrell dust (approximately 1-2% Cd) from Noranda Horne mine in northern Quebec has been brought in by rail, unloaded by a rotary car dump and stockpiled on the Belledune site in a covered building. The Horne dust and the above concentrate are used in the charge preparation.

CHARGE PREPARATION

At this stage of the process, Pb concentrates are mixed with fluxes (sand and lime), sinter fines, dusts, drosses, sludges, etc. (which contain Pb and other metals). This mixture is then formed into pellets and conveyed to the sintering machine.

SINTERING PROCESS

The sintering process is carried out on a Lurgi (Dwight-Lloyd type) sintering machine. A layer (3.8 cm thick) of the pelletized mixture is laid onto the moving bed/grate of the machine and is subsequently ignited by burners above the bed. The rest of the "charge" is then loaded on top of this burning layer to form a bed up to 30.5 cm thick. The travelling grate then enters the updraught "windbox" section of the machine where air is blown through the bed. This provides the oxygen for the burning process. The bed burns from the bottom to the top, releasing sulfur dioxide (SO_2) and traces of sulfur trioxide (SO_3) - resulting in removal of almost all of the sulfur and volatiles from the concentrate. At this point any Cd not "fixed" in the sinter is volatilized and carried off with the SO_2 and dust. At Brunswick Smelting, SO_2 from the sinter machine is drawn through an electrostatic precipitator to remove most (98%) of the Pb dust prior to gas cleaning in the Acid Plant. The precipitated Cd-bearing dust is returned to the process at the rate of 1-2 MT per hour to recover Pb.

The hot sinter coming off the end of the machine is broken by a claw breaker into pieces

Table 1. Composition of Pb concentrates.

	Composition, %		
	Brunswick Mining No. 12 Pb concentrate	Pb concentrate (Brunswick Smelting data)	
Component	(Neumann & Schnarr 1971)	Jan-Mar 1979	1980
Pb	38	32	30
Zn	10	7	7
S	27	33	34
Fe	14	23	23
Cd	0.02	0.014	0.014

approximately 10.2 cm diameter and fed into the blast furnace. The fines are removed and returned to the sintering process charge preparation stage.

BLAST-FURNACE SMELTING

The sinter is mixed with coke and fed to the blast furnace. Table 2 shows the typical Brunswick Smelting sinter analysis.

A series of complex chemical reactions occur in the blast furnace, yielding the following products:

1) Pb bullion, which still contains the impurities, Cu, arsenic (As), antimony (Sb), bismuth (Bi) and some silver (Ag). This bullion must be refined further to produce a higher purity Pb final product.

2) Slag, which is composed of the silicates of iron, calcium, magnesium and almost all of the Zn, present originally in the concentrate. At Brunswick Smelting no attempt is made to recover the Zn. This slag is then granulated by impacting a stream of molten slag with a jet of fresh water and transported with a stream of seawater to the slurry pumps. The granulated slag formed is black, glassy and consists of sand-like particles or larger lumps. The granulated slag and water are then pumped to the slag pile where the water part of the slurry runs off.

3) Baghouse dust, which is recycled to the sinter plant. However, when the Cd content approaches 10%, a portion is stockpiled for future sale.

REFINING OPERATIONS

The impure Pb bullion is refined by a series of drossing (floating on the surface) operations. The prime objective of drossing is to remove metallic impurities. Drossing occurs in large, steel, hemispherical kettles up to 3.66 m in diameter and capable of holding up to 209 MT of molten material. The kettles are heated externally and have associated with them equipment for stirring, skimming and transferring the products. In the first drossing process, the temperature of the Pb bullion is lowered close to the melting point of Pb (327.4°C). At this temperature the solubility of Cu in Pb is low and the Cu forms a crust on top of the melt. The Cu crust is skimmed off the top, resulting in reduction of the Cu content of the bullion from several percent to less than 0.5%. The liquid lead is then transferred to a second kettle where another Cu drossing occurs lowering the Cu content of the Pb to less than 0.01%. The Cu drosses are then processed further to form Cu matte.

Next the Pb is softened to remove Sb, As and tin (Sn). This is accomplished by a process known as oxygen softening (previously oxidative slagging with sodium hydroxide and sodium nitrate was used in this softening process). An air-oxygen mixture is injected into the molten Pb and the As, Sb and Sn are oxidized and form a crust which is skimmed off. Ag and Au are then removed by treatment of the molten Pb with Zn metal. Zn combines with Ag and Au to form alloys that are insoluble in Pb and forms a crust that is skimmed off and treated to produce Ag. The trace amounts of Zn remaining in the molten Pb are removed and recovered by a vacuum distillation. Bi is then removed in a two-step process with additions of calcium and magnesium and a lowering of the molten Pb temperature. The crust of intermetallic compounds is skimmed off and the Bi recovered.

The molten Pb is now of high purity and is cast into pigs (25.4 or 45.4 kg) and jumbos (0.91 MT). The refining operations at different Pb smelters can vary in the type of processes used (due to the variation in the concentrations of the impurities and which byproducts are desired). These operations are complex, with various chemicals being added in combination with heating, cooling and transferring between kettles of the molten Pb.

FERTILIZER PLANT OPERATION

To use the SO_2 and SO_3 byproducts released in smelting, Brunswick Smelting operates a sulfuric acid plant. Sulfuric acid is conducted by a short pipeline to the Belledune phosphate fertilizer plant. In the plant a Florida phosphate rock (127,000 MT in 1970) is ground in a ball mill and then treated with the sulfuric acid. Phosphoric acid is produced in the reaction and is then reacted with ammonia to produce diammonium phosphate (fertilizer).

Table 2. Composition of sinter.

Component	Belledune sinter (Brunswick Smelting data) (%)	
	Jan-Mar 1979	1980
Cu	0.6	1.0
Pb	32	30
Fe	20	22
S	1.0	1.0
Zn	8.0	7.0
Cd	0.38	0.30

RELEASE OF Cd TO THE ENVIRONMENT

The significant discharges or emissions of Cd to the environment are to the air, by surface runoff due to precipitation events, by cooling water discharges, from slag pond discharges, from the fertilizer plant discharges, and from the leaching of solids (dusts, slags) by salt, fresh or rainwater.

AIR EMISSIONS

Since 1968, dust from the blast furnace, sinter plant and lead refinery has been collected in baghouses and recycled to the sinter plant. The total baghouse capacities have been increased from 150,000 a.c.f.m. in 1968 to 1,000,000 a.c.f.m. in 1976 with the construction of two larger baghouses in 1973 and 1976. It should be noted that used baghouse dust tubes are also recycled back to the blast furnace. Because of the recycle of dust, the Cd concentration in all dusts (particularly that from the blast furnace) gradually increases. When the Cd concentration in the blast-furnace dust reaches about 10% (has been known to exceed 20% in the past), it is bled off and sealed in containers, for sale and refining elsewhere or is piled in the yard (about 180 MT at present). This Cd-laden dust is the major output source of Cd (of about 545 MT produced last year, the company sold approximately 363 MT).

Cd in forest ecosystems around lead smelters in Missouri has been documented by Wixson et al. (1977) and by Gale and Wixson (1979). Elevated Cd concentrations were found in forest litter and the decomposed litter along stream beds, with appreciable contributions of dissolved Cd to watershed runoff. Cd emissions were discussed by Dugdale and Hummel (1977). Losses to the atmosphere were estimated to be 3,391 kg Cd/yr from baghouses and scrubbers. Figure 1 shows the distribution pattern of several heavy metals in surface soil samples in the area of the smelter complex along with some indication of wind direction. If one considers the background Cd concentration in soils to be between 1 and 2 µg/g, then the effects due to the smelter appear to be localized within the first few kilometers. This same pattern of localization appears to hold for heavy metal concentrations in vegetation. Undoubtedly, there is a similar pattern of deposition on the surface of the estuary. Quantifying this deposition is difficult but for Pb it has been estimated at 9 kg/ha x month (Dugdale and Hummel 1977) and may be in the order of 0.2 kg/ha x month for Cd at the smelter complex, decreasing rapidly within the first few kilometers from the site. Although the air pollution controls are the best available for this type of industry, heavy metals reaching the surface of the water from fugitive baghouse and scrubber dusts may be in a readily bioavailable form. These air emissions may be a significant source of Cd. Smaller amounts of Cd-containing dust may be released to the atmosphere from the rotary car dump facility where the concentrate from Brunswick No. 12 and the Horne dust are unloaded from rail cars.

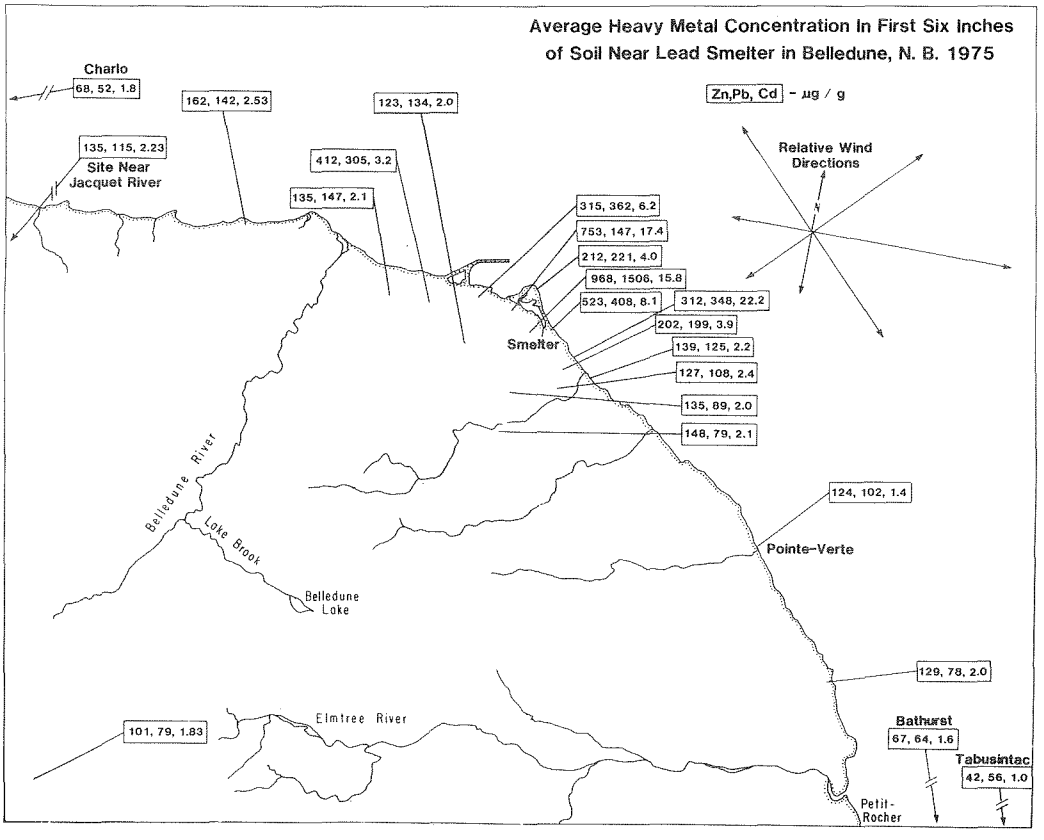


Fig. 1. Average concentrations of Zn, Pb and Cd in µg/g in the first 6 in. of soil near the lead smelter in Belledune, N.B., 1975.

SURFACE WATER RUNOFF

A study of the drainage patterns near the smelter was made for Brunswick Mining and Smelting in 1979 by Montreal Engineering Company Limited. Drainage from approximately 809 ha passes around and through the smelter property. East and west ditches divert runoff from all but 28 ha of the smelter proper. The latest change (made this year) was to divert the runoff from 10 ha south of Highway 11 to the west diversion ditch.

In the past years the smelter runoff drained into the bay at several points along the property, but primarily through the slag pond and a contaminated ditch on the east side. In 1980 modifications were made that have diverted this drainage to the northeast corner of the site, where a new treatment facility has been constructed.

Heavy metal concentrations in runoff can be estimated for the contaminated ditch on the east side of the smelter. The concentrations of Pb, Zn, and Cd are 0.8, 5.3 and 0.1, and 6.6, 35.2, and 1.6 mg/L during dry weather and heavy precipitation, respectively (1979 data, Brunswick Smelting). Accordingly, surface runoff can contribute significant amounts of Cd to the Bay of Chaleur.

COOLING WATER

Typically, 45,460-54,550 L/min of salt water are used for cooling. This water is passed through the plant by means of two channels on the northwest side of the property. Since it does not contact the process waters within the plant, it contributes very little to the heavy metal loading of the harbour. However, the saltwater inlet is very close to the process water outlet and there is some entrainment of the final effluent in the cooling water intake. Before entering Belledune Harbour, the cooling water combines with the slag pond discharge. By the end of 1980 the cooling water will no longer combine with the process water.

PROCESS WATERS AND SLAG POND DISCHARGE WATERS

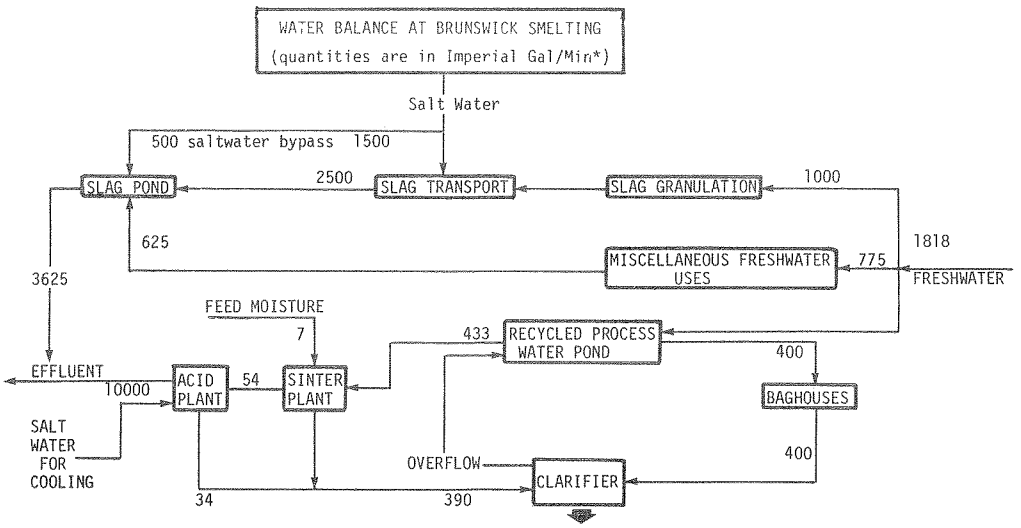
Figure 2 is a simplified water balance diagram for the smelter. Fresh water from the various processes in the plant, as well as the salt water used to transport the slag to the slag pile, combine in the slag pond, which in turn discharges into the main cooling water channel. Heavy metal concentrations in process water range from 1.5-4.2 mg/L for Pb, 3.2-16.4 mg/L for Zn, and 0.6-38.0 mg/L for Cd at the discharge of the slag pond (1979 data, Brunswick Smelting). After combining with the cooling water, the concentrations range from 0.4-2.3 mg/L for Pb, 0.3-9.2 mg/L for Zn, and 0.3-3.0 mg/L for Cd. Based on the volume of water used in the process this is also a significant source of Cd to the environment. Treatment of these process waters to remove heavy metals is now planned.

Figure 3 is a schematic diagram showing the sulfuric acid plant. The acid stream washer water is diluted sulfuric acid and is added to the reprocess water pond. As acid will solubilize Cd and as there is probably an overflow in this pond, it stands to reason that more Cd will be added back into the process by recycling this water (then subject to loss of high Cd water) or to ditches via overflow.

LEACHING OF SLAG AND DUSTS

Leonard et al. (1977) reported the concentration of Cd at various steps in the refining process. Some of their findings are shown in Table 3.

Also in this reference was a leaching study performed with secondary blast-furnace slag, furnace dust and scrubber sludge. Only one test was performed for each, the pH was not specified and higher



* NOTE - the diagram is an approximation and does not exactly balance in part due to losses to the atmosphere, etc.

Fig. 2. Water balance diagram for Brunswick Smelting operation at Belledune.

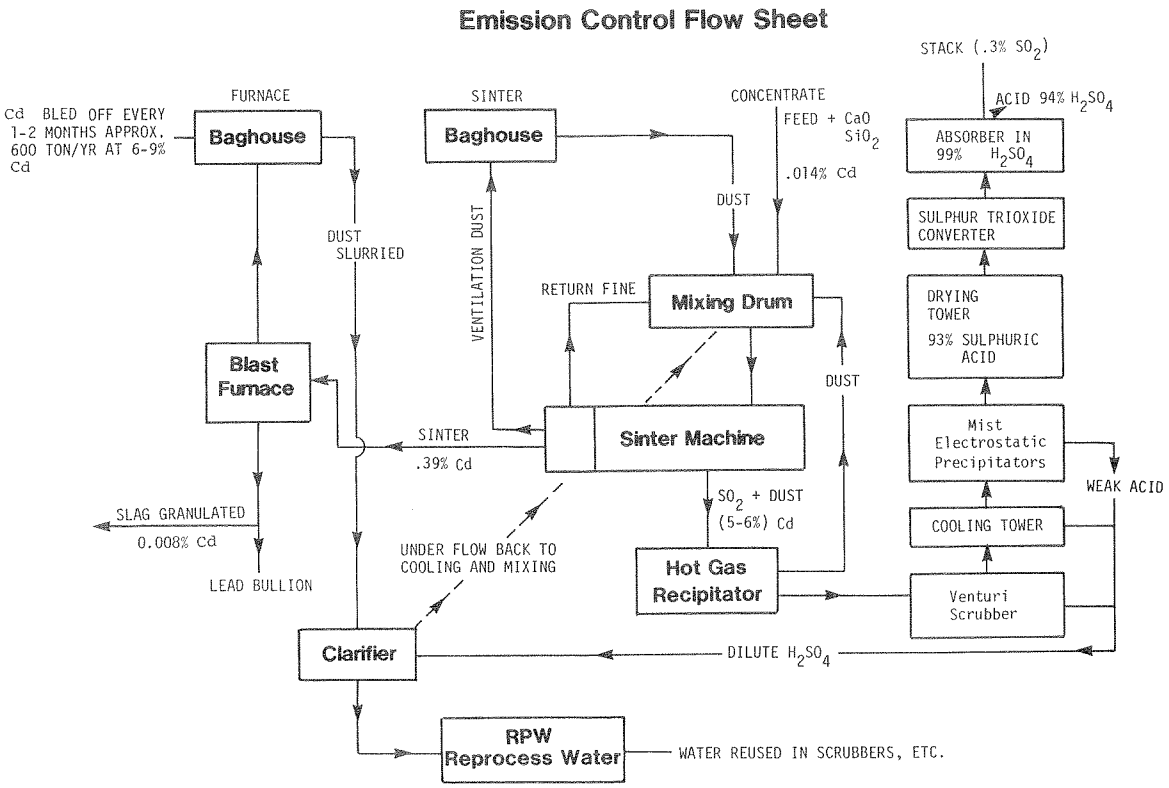


Fig. 3. Emission control schematic diagram showing dust recycling and SO_2 removal processes in the smelter complex.

or lower pH's were not examined. Less than 0.01 ppm Cd was leached from the slag, 5 ppm Cd from the sludge and 230 ppm Cd from the furnace dust in a 76-h test. This does not explain the results observed in Table 3 for Plant B slag where extensive leaching is evident.

In regard to leaching, samples of the Belledune slag, baghouse dust and Cottrell dust (Horne mine) were analyzed and tested for leaching of Cd by Sergeant and Ray (this report).

HORNE MINE DUST

The conversion from a Pb/Zn smelter to a Pb smelter, the increased throughput since 1974 and the processing of the Cottrell dust (Horne mine) may be resulting in increased Cd emissions. Dugdale and Hummel (1977) show that the concentration of Cd in mussels increased from 3.4 ppm wet weight in 1972 to 11.0 ppm wet weight in 1975, after the plant conversion. Whether this increase was due to the process change or to buildup of available Cd since plant start-up is open to conjecture. Dust from the furnace top ranges as high as 12.9-21.8% Cd (Dugdale and Hummel 1977), and whether the Cottrell dust has caused this to increase further or to be reached faster is not known.

FERTILIZER PLANT DISCHARGES

The effluent of the fertilizer plant contains gypsum, fluorides and iron as well as very small concentrations of the heavy metals. Pb ranged from 0.003-1.1 mg/L; Zn from 0.17-1.1 mg/L; Cd from 0.07-1.6 mg/L (Environmental Protection Service data). Discharge water flow is approximately 9,090 L/min so the yearly contribution to the Bay of Chaleur would be low (assuming continuous operation of the plant).

In Sweden there is concern over additions of Cd to soils in fertilizer (Nilsson 1980). It has been estimated that one third to one half of the Cd additions to soil occur through the use of phosphate fertilizers. With this in mind, one cannot discount surface and non-process water runoff as a significant source of Cd at this site. See also Lymburner (1974) on superphosphate fertilizers.

Table 3. Cadmium (Cd) concentrations at various stages in U.S. Pb industries and Cd solubility in water exposed to process materials (Leonard et al. 1977).

Material analyzed		Cd concentration (ppm)
Plant A		
Fresh blast-furnace slag		10
Old blast-furnace slag		88
Sinter scrubber sludge		900
Lagoon dredgings		700
Baghouse dust		14000
Plant B		
Fresh blast-furnace slag		1150
Old blast-furnace slag		73
Plant A (secondary refining process)		
Scrubber sludge		340
Wastewater treatment sludge		10
Filtrate from a solubility test using:		
Blast-furnace slag		0.03
Lead fuming slag		0.01
Blast-furnace dust		8.0
Sinter scrubber sludge		9.1
Lagoon dredgings		11.0
Primary lead Smelting and refining	Slag	166
	Sludge	6900

POLLUTION CONTROL MEASURES

By late 1980 the following pollution control measures will be in place:

1. All clean water drainage will have been diverted around the plant.
2. Conventional treatment for sewage wastes will be operational.
3. All contaminated surface water drainage from the site will be collected in one place for treatment.
4. All contaminated waters will be treated by pH adjustment, coagulation and settling.
5. Underflow (slurry of sludge) from this treatment facility will be recycled. Much of the overflow will also be recycled reducing flows in the effluent considerably.
6. Slag will be handled by cooling after granulation and trucked to a new slag area south of the highway. The drainage water from this area will be treated along with other process waters.

Once these pollution abatement measures are in place, the Cd discharged into the environment should drop significantly. However, the storing of dust materials in the open at the site should also be discouraged (in particular, storage of 10% Cd dust in the open) as this will only be wind-blown.

ACKNOWLEDGMENTS

We thank Dr. V. Zitko for providing us with many of the references, Dr. J. Uthe for his comments on an earlier draft, and Brunswick Smelting for the release of their data.

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DISTRIBUTION OF CADMIUM IN MARINE BIOTA IN THE VICINITY OF BELLEDUNE

by

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INTRODUCTION

The occurrence of high cadmium (Cd) levels in the tissues of lobsters and mussels taken from the vicinity of the Brunswick Smelting lead smelter at Belledune, N.B., was brought to our attention in late April 1980. The lobster fishing season was due to open on May 1 and a study was initiated immediately to determine the extent of Cd contamination in lobsters in the area (Uthe et al., this report).

We conducted a survey during May 5-16, 1980, to determine the extent of Cd contamination in marine biota other than lobsters. The survey included sampling stations in the area bounded by Beresford and Little Belledune, N.B. Included in the biota sampled were several commercially important plant, bivalve, crustacean and fish species. Also, sediment samples were collected at each sampling site.

The main fisheries in the Belledune area include lobster trapping at depths of 3-11 m within 1.5 km of shore, scallop dragging at 12-18 m in areas offshore of Beresford, N.B., and Belledune, and herring fishing with gill nets or seines. There are no extensive beds of clams, oysters, quahogs or Irish moss. Crabs and shrimp are not fished commercially in the area.

SAMPLING AND ANALYSIS

COLLECTION

Samples of plants, animals, and sediments were collected at 11 shore and 22 offshore stations (Fig. 1). The A stations were sampled from shore at low tide and B stations (8-11 m) were sampled by SCUBA divers. The C stations (11-20 m) were sampled by scallop drag, and D stations (29-34 m) were sampled by ponar grab. A 14-m vessel owned by C. Doucette was used for sample collections.

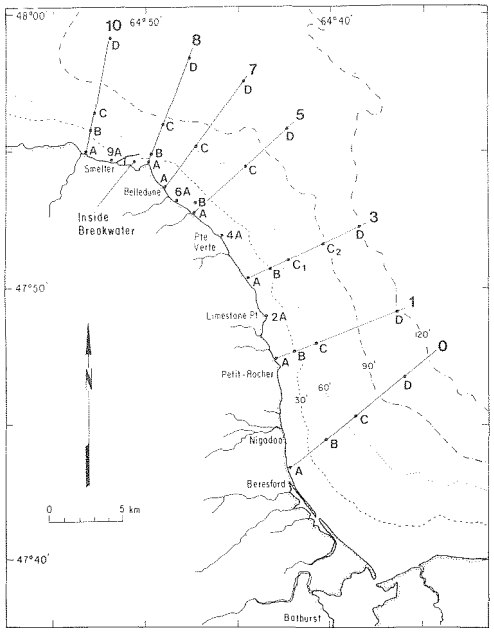

Fig. 1. Locations of sampling transects and stations in the survey area adjacent to the Brunswick Mines smelter.

Scallops, mussels, crabs, barnacles, and macrophytic algae were the taxa collected most extensively. Biota with limited distribution were not analyzed (Table 1). Immediately upon collection, all individual biota and sediment samples were frozen in Whirl-pac ^{R} bags.

SAMPLE PREPARATION

Biota samples were thawed and washed with deionized water to remove grit and salt before dissection. Scallops (Placopecten magellanicus, Chlamys islandicus) were dissected into viscera (containing stomach, gill, gonad, hepatopancreas, heart and kidney), adductor muscle, and mantle. Samples taken from flatfish (Hippoglossoides platessoides, Limanda ferruginea and Pseudopleuronectes americanus) were dorsal muscle and viscera (stomach, intestines and gonad). Cancer irroratus and Hyas araneus crabs were dissected into hepatopancreas, gill, cheliped (pincer) muscle, and coxal (shoulder) muscle. Whole Pagurus sp. crabs and barnacles (Balanus balanoides) were prepared. For all other shellfish, only soft tissues were used. Fucus vesiculosus and Ascophyllum nodosum algae were divided into stem and frond sections.

Polychaetes, periwinkles, barnacles, scallop adductor muscle, and C. irroratus and H. araneus crab tissues were freeze-dried only. Flatfish muscle, macrophytic algae, Mytilus edulis and Modiolus modiolus mussels, cockles, Aporrhais occidentalis, Pagurus crabs, and scallop mantles were freeze-dried and milled to a powder. Scallop and flatfish viscera, quahogs and whelks were homogenized with deionized water by using a Sorval homogenizer and then freeze-dried. Sediment samples were thawed, dried at 60°C, and sieved through 250-µm and 63-µm stainless-steel sieves.

Biota samples (0.2-0.5 g) were ashed for 12 h at 450°C and taken up in 1 mL nitric acid for Cd analysis. Sediment samples were digested with 1 mL nitric:hydrochloric acid (3:1; v/v), filtered and made to volume for analysis. The acids used were of Aristar quality (B.D.H. Chemicals Ltd., Poole, U.K.). Cd was measured by using a Perkin-Elmer Model 503 atomic absorption spectrometer and Model 500 graphite furnace. Accuracy and precision were checked by using N.B.S. reference materials (#1643a - water, #1566 - oyster tissue). The results are expressed on a dry weight basis, except where stated otherwise. Animal sizes and either the wet weights and dry weights or wet weight-dry weight relationships for biota samples are tabulated in Appendix I.

RESULTS AND DISCUSSION

The general distribution of bottom sediments in the survey area, as determined by SCUBA observation and subjective examination of bottom grabs, is illustrated in Fig. 2. The distribution of the biota in the survey area appears to be related to the bottom type. For instance, scallops are generally located in the areas with gravel and clay sediment and cockles were collected in muddy areas. Except for fish and crabs, few species were seen by divers in the silt zone surrounding the lead smelter.

Table 1. Species collected in the survey area. Numbers of specimens collected are in brackets and P denotes a pooled sample. Species marked with asterisk were not analyzed.

Species \ Transect	0	1	2	3	4	5	6	7	8	9	10
Molluscs											
1) Placopecten magellanicus	C(3)			C(1)		B(1) C(3)		C(3)	C(3)		C(3)
2) Chlamys islandicus									C(2)		C(2)
3) Mytilus edulis	A(2)	A(7P)	A(5P)	A(5P)	A(4P)	A(5P)	A(5P)	A(4P)		A(10P)	A(5P)
				B(3)							B(3)
											C(2)
4) Modiolus modiolus		B(2)				C(1)					C(2)
5) Clinocardium ciliatum		D(2)		D(2)		D(2)		D(2)			
6) Mercenaria mercenaria						B(2)					
7) Littorina sp.		A(7P)				A(6P)	A(7P)			A(6P)	A(2P)
8) Aporrhais occidentalis	C(1)	C(1)		D(1)				C(1)	C(1)		
								D(1)			
9) Buccinum undatum								C(2)	C(4)		C(1)
10)*Mya arenaria						D(1)					
11)*Astarte sp.				B(3)					C(2)		
12)*Dentalium entale						D(2)			D(4)		D(2)
Crustaceans											
13) Cancer irroratus	B(2)	B(1)		C(1)	C(1)	B(1)		C(1)	B(3)		A(1)
	C(1)								C(1)		B(1)
											C(1)
14) Pagurus pubescens	C(2)			B(1)		B(1)					B(1)
				C(1)							
14) Pagurus acadianus						C(1)					C(1)
15) Hyas araneus		C(1)						D(3)			
16) Balanus balanoides		A(12P)		A(5)		A(15P)	A(16P)	A(17P)		A(14P)	
*Amphipod											A(4)
*Mysid											A(4)
Polychaetes (marine worms)											
Aphrodite hastata				C(2)							
Lepidonotus squamatus				D(1)							
Phyllodoce sp.		D(1)									D(1)
Clymenella sp.		D(1)									D(1)
Nephtyid				C(2)							
				D(1)							
Arabellid	C(1)								D(1)		D(1)
Macrophytic algae (seaweeds)											
17) Laminaria digitata	A	A	A	A	A	A	A		A	A	A
		B		B		B					B
18) Fucus vesiculosus	A	A	A	A		A	A	A		A	
		B									
19) Ascophyllum nodosum				A	A	A				A	A
Fish											
20) Limanda ferruginea	C(1)	C(2)									C(1)
21) Hippoglossoides platessoides	C(2)	C(2)							B(2)		
22) Pseudopleuronectes americanus				C(1)							
23)*Raja serta									B(1)		
24)*Myoxocephalus octodecimspinosus									B(1)		
25)*Gasterosteus aculeatus											A(10)
Starfish											
26)*Solaster endeca	C(1)							C(1)			D(1)
27)*Ophiopholis aculeata	D(3)			D(1)					D(2)		

1)Scallop; 2)Iceland scallop; 3)Mussel; 4)Horse mussel; 5)Iceland cockle; 6)Bar clam; 7)Periwinkle; 8)American pelican's foot; 9)Dog whelk; 10)Soft shelled clam; 11)Waved Astarte; 12)Tusk shell; 13)Rock crab; 14)Hermit crab; 15)Spider crab; 16)Barnacle; 17)Kelp; 18)Bladder wrack; 19)Knotted wrack; 20)Yellowtail flounder; 21)American plaice; 22)Winter flounder; 23)Skate; 24)Sculpin; 25)Stickleback; 26)Sun star; 27)Brittle star.

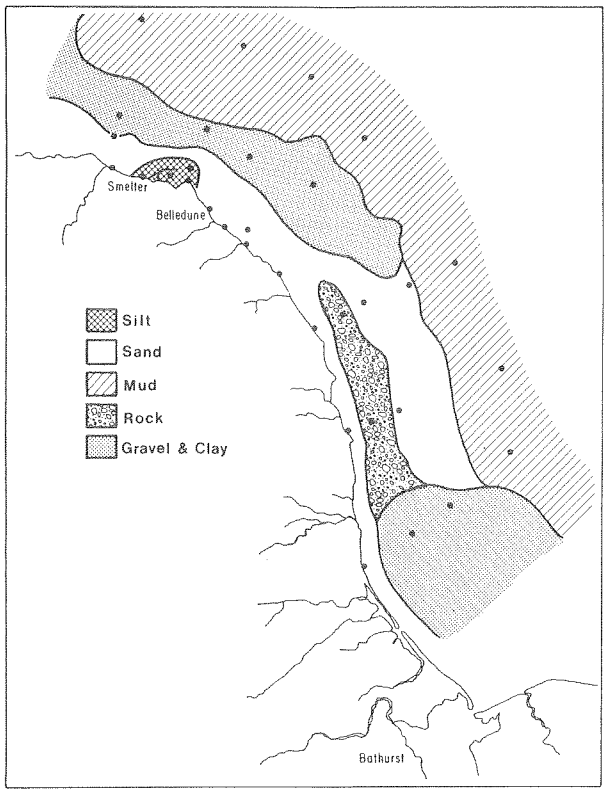

Fig. 2. General distribution of bottom sediment types in the survey area.

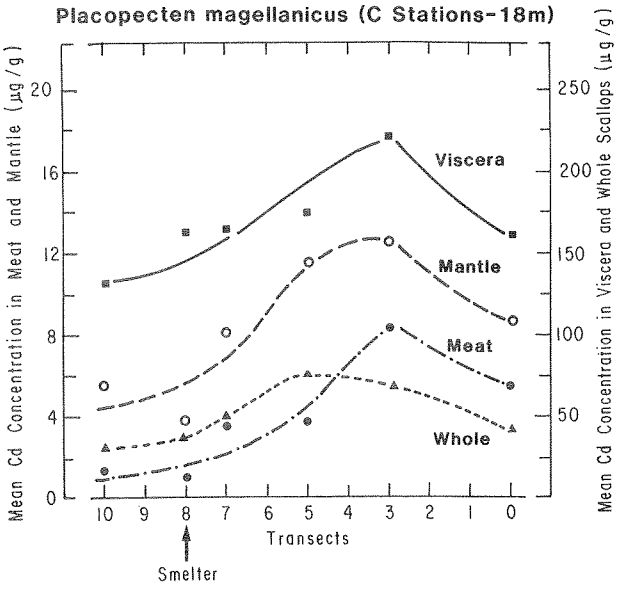

Fig. 3. Mean cadmium (Cd) concentrations in meat, mantle, viscera, and whole animals of P. magellanicus sampled at C stations (18 m).

SCALLOPS

P. magellanicus were collected from seven stations and C. islandicus from two stations. The mean Cd content of adductor muscle, mantle and viscera of the scallops from each station is illustrated in Fig. 3-6.

The means (ranges) of Cd levels (N=17) in P. magellanicus from the survey area were 3.7 (0.5-8.4), 8.5 (3.2-18.0) and 183.4 (87.6-493.2) µg/g for muscle, mantle and viscera, respectively. For comparison, P. magellanicus (N=6) of approximately the same size range, collected from Passamaquoddy Bay, N.B., had lower mean Cd concentrations of 0.8, 1.3 and 47.7 µg/g in muscle, mantle and viscera (Ray et al., unpublished). Pesch et al. (1977) reported average Cd concentrations of 20.9 µg/g for whole, shucked P. magellanicus collected near ocean disposal sites off the U.S. Atlantic coast. For comparison, the mean and range of concentrations in whole scallops in the Belledune area of 50.25 µg/g (23.1-115.6) were estimated from the sum of the Cd burdens in the various tissues. Zook et al. (1976) found 0.1 (.04-0.2) µg Cd/g wet weight in adductor muscle from P. magellanicus collected off the U.S. Atlantic coast. The mean and range of Cd concentrations on a wet weight basis for P. magellanicus muscle in the Belledune area were 0.8 (0.1-1.6) µg/g.

Cd levels in the three tissues of P. magellanicus taken at C stations (Fig. 3) indicate that concentrations are elevated at transects 0, 3, 5 and possibly 7. Estimates of the whole-body concentration follow this general trend, although

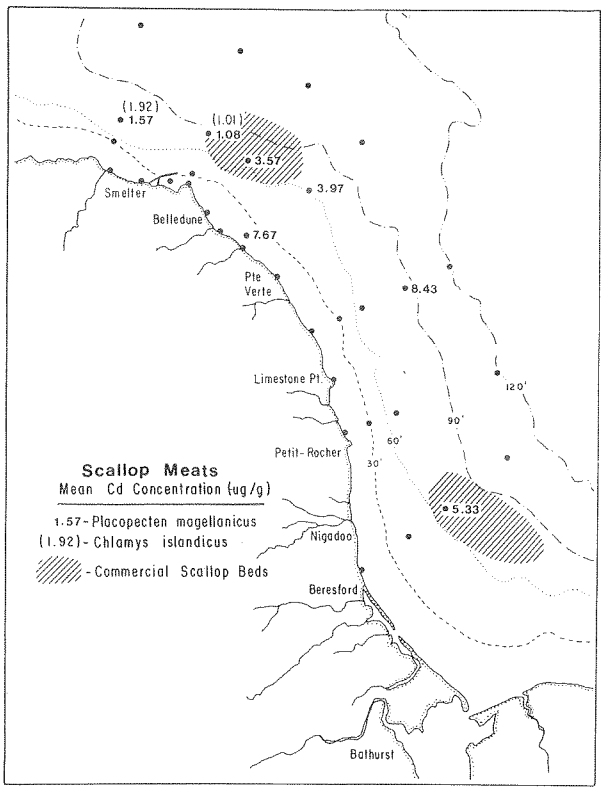

Fig. 4. Mean cadmium (Cd) concentrations in meat of P. magellanicus and C. islandicus samples from the survey area.

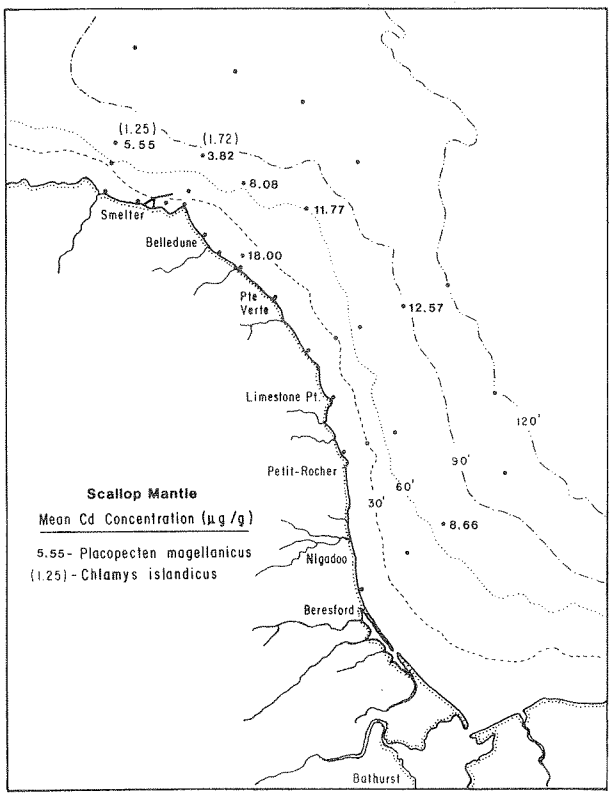

Fig. 5. Mean cadmium (Cd) concentrations in mantles of P. magellanicus and C. islandicus sampled from the survey area.

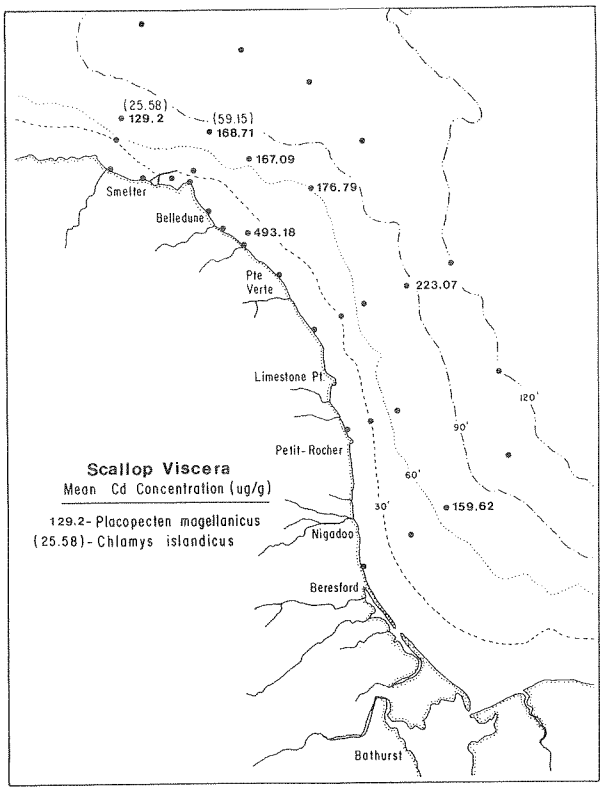

Fig. 6. Mean cadmium (Cd) concentrations in the viscera of P. magellanicus and C. islandicus sampled from the survey area.

variations in the proportion of viscera in the scallops cause the Cd maximum to shift to transect 5.

The data for C. islandicus were limited to two sampling stations. However, adductor muscle from both scallop species from the same sites contained approximately equal levels of Cd, while Cd concentrations in the mantles and viscera of C. islandicus were considerably lower than for P. magellanicus.

MUSSELS

Mytilus edulis were collected from 13 stations and Modiolus modiolus from four stations within the survey area. For some stations, several M. edulis (4-7 animals) were pooled for analysis but, because of variations in the Cd content of subsamples from the pooled tissues, the remaining mussels were analyzed individually. All M. modiolus were analyzed individually. The mean Cd concentrations in the soft parts of M. edulis and M. modiolus from each station are illustrated in Fig. 7 and 8.

The soft tissues of M. edulis and M. modiolus contained a mean and range of 30.1 (2.9-112.2) µg/g and 32.1 (15.2-55.9) µg/g of Cd, respectively. These concentrations are considerably higher than the Cd levels reported in the literature (Table 2) for mussels from unpolluted and moderately polluted marine areas. In M. edulis from shore (A) stations (Fig. 7), the mean Cd concentration decreases with distance to the north or south of transects 6 and 7. M. modiolus at stations 10C, 8C and 5C contained relatively uniform concentrations of Cd (Fig. 8).

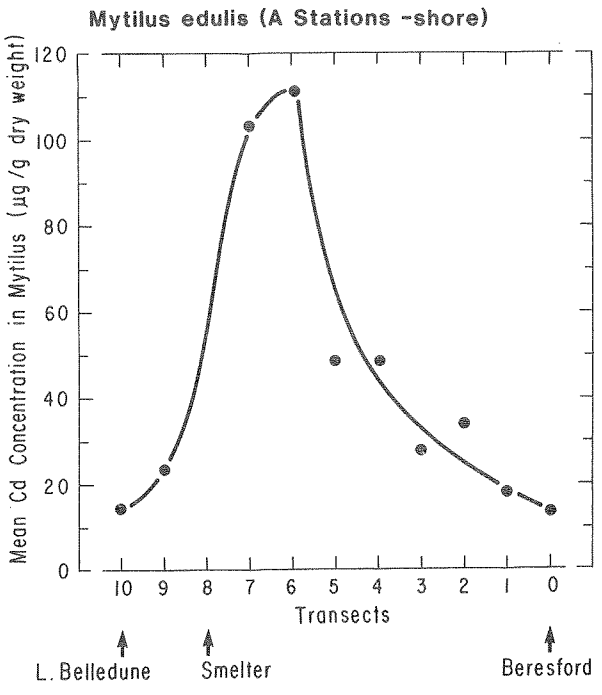

Fig. 7. Mean cadmium (Cd) concentrations in soft parts of M. edulis sampled at A stations (shore).

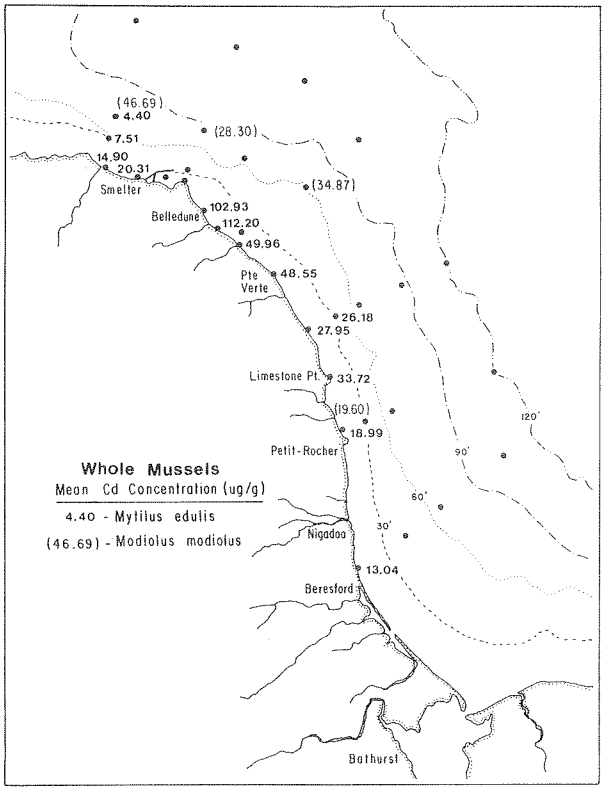

Fig. 8. Mean cadmium (Cd) concentrations in soft parts of M. edulis and M. modiolus sampled from the survey area.

CRABS

The mean concentrations of Cd in hepatopancreas, gill, coxal muscle and cheliped muscle of Cancer irroratus and Hyas araneus, and in whole Pagurus species are illustrated in Fig. 9-14.

Samples of H. araneus and Pagurus sp. crabs were too infrequent to indicate the distribution of Cd within the survey area. Whole Pagurus sp. contained an average of 1.2 µg Cd/g and H. araneus contained Cd concentrations of 4.2, 9.7, 0.6 and 0.8 µg/g in hepatopancreas, gill, coxal muscle and cheliped muscle, respectively.

The more extensive data for C. irroratus indicate that the mean concentration of Cd in hepatopancreas tissue is elevated at stations 5B and 8B, and moderately elevated at station 7C (Fig. 9). The distributions of Cd concentrations in the other tissues (Fig. 10-13) also reflect this general pattern.

The mean and range of Cd levels in C. irroratus tissues from the survey area were 87.1 (2.3-394.3), 17.4 (1.7-53.5), 5.3 (0.2-29.2) and 1.3 (0.1-8.1) µg/g dry weight for hepatopancreas, gill, coxal muscle and cheliped muscle, respectively. The calculated mean and range of concentrations on a wet weight basis in hepatopancreas and coxal muscle were 7.1 (0.2-35.1) and 0.65 (0.03-3.8) µg/g. C. irroratus (N=4) of approximately the same size range from Passamaquoddy Bay, N.B., and from ocean dumpsites off the U.S. Atlantic coast (Table 3) had lower levels in their tissues. Concentrations in C. pagurus from two British sites (Topping 1973; Reynolds and Reynolds 1971) were approximately the

Table 2. Cadmium (Cd) concentrations reported for whole soft tissue of mussels.

Species	Cd concentration - mean or range (ppm)	Location	Reference
Mytilus edulis Modiolus modiolus	5.1(dry wt) 5.8(dry wt)	Irish Sea	Segar et al. (1971)
Mytilus edulis	2.5(dry wt)	Southampton, U.K.	Leatherland & Burton (1974)
Mytilus edulis Modiolus neozelandicus	0.3-1.6(dry wt) 0.04(dry wt)	New Zealand	Nielsen & Nathan (1975)
Mytilus edulis	0.1-0.9(wet wt)	Scotland	Topping (1973)
Mytilus edulis	0.2-18.2(wet wt)	Victoria, Australia	Phillips (1976)
Mytilus galloprovincialis	0.4-5.9(dry wt)	N.W. Mediterranean	Fowler & Oregioni (1976)
Mytilus edulis	1.4-4.2(dry wt)	St. Croix est., Me.	Fink et al. (1976)
Mytilus edulis	0.9-1.8(dry wt) 1.7 (dry wt)	Maine Narrangansett Bay	Goldberg et al. (1978)
Mytilus edulis Modiolus modiolus	2.9-112.2(dry wt) 15.2-55.6(dry wt)	Belledune area	This study

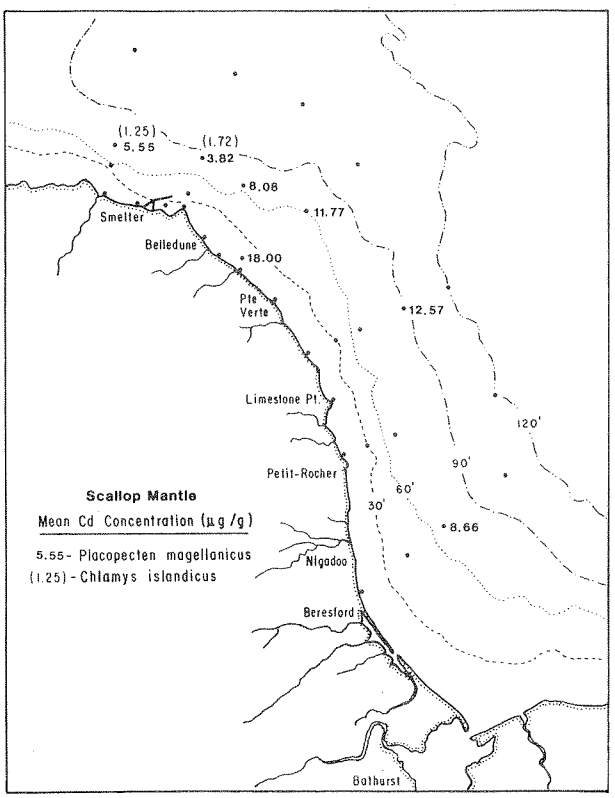

Fig. 5. Mean cadmium (Cd) concentrations in mantles of P. magellanicus and C. islandicus sampled from the survey area.

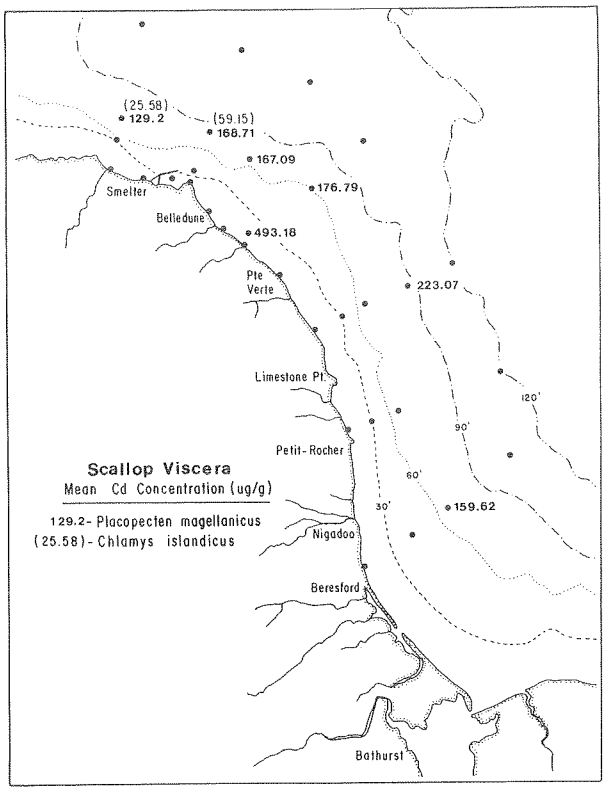

Fig. 6. Mean cadmium (Cd) concentrations in the viscera of P. magellanicus and C. islandicus sampled from the survey area.

variations in the proportion of viscera in the scallops cause the Cd maximum to shift to transect 5.

The data for C. islandicus were limited to two sampling stations. However, adductor muscle from both scallop species from the same sites contained approximately equal levels of Cd, while Cd concentrations in the mantles and viscera of C. islandicus were considerably lower than for P. magellanicus.

MUSSELS

Mytilus edulis were collected from 13 stations and Modiolus modiolus from four stations within the survey area. For some stations, several M. edulis (4-7 animals) were pooled for analysis but, because of variations in the Cd content of subsamples from the pooled tissues, the remaining mussels were analyzed individually. All M. modiolus were analyzed individually. The mean Cd concentrations in the soft parts of M. edulis and M. modiolus from each station are illustrated in Fig. 7 and 8.

The soft tissues of M. edulis and M. modiolus contained a mean and range of 30.1 (2.9-112.2) µg/g and 32.1 (15.2-55.9) µg/g of Cd, respectively. These concentrations are considerably higher than the Cd levels reported in the literature (Table 2) for mussels from unpolluted and moderately polluted marine areas. In M. edulis from shore (A) stations (Fig. 7), the mean Cd concentration decreases with distance to the north or south of transects 6 and 7. M. modiolus at stations 10C, 8C and 5C contained relatively uniform concentrations of Cd (Fig. 8).

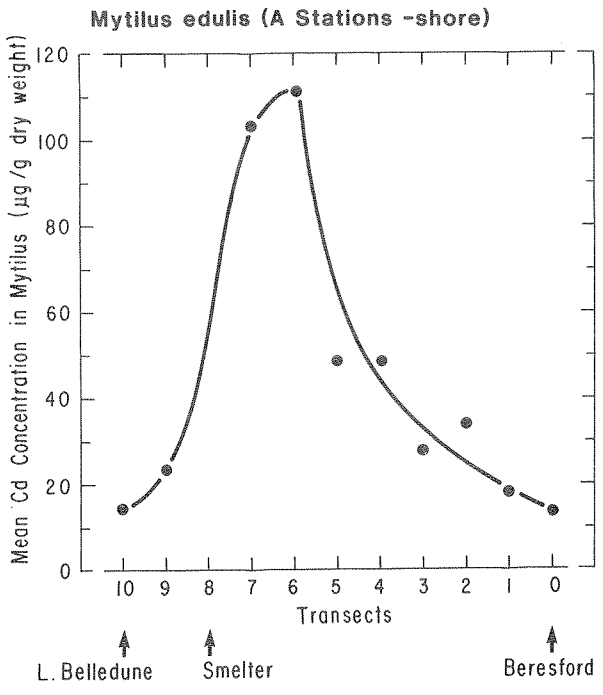

Fig. 7. Mean cadmium (Cd) concentrations in soft parts of M. edulis sampled at A stations (shore).

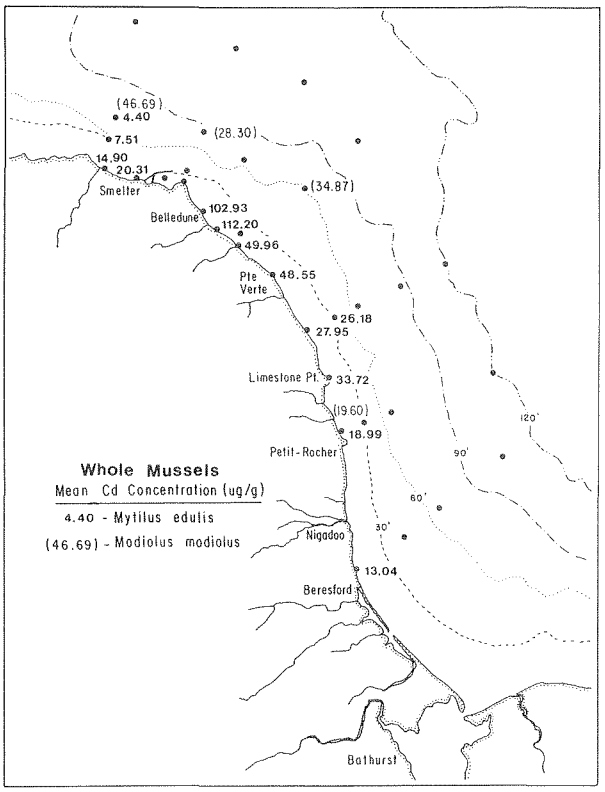

Fig. 8. Mean cadmium (Cd) concentrations in soft parts of M. edulis and M. modiolus sampled from the survey area.

CRABS

The mean concentrations of Cd in hepatopancreas, gill, coxal muscle and cheliped muscle of Cancer irroratus and Hyas araneus, and in whole Pagurus species are illustrated in Fig. 9-14.

Samples of H. araneus and Pagurus sp. crabs were too infrequent to indicate the distribution of Cd within the survey area. Whole Pagurus sp. contained an average of 1.2 µg Cd/g and H. araneus contained Cd concentrations of 4.2, 9.7, 0.6 and 0.8 µg/g in hepatopancreas, gill, coxal muscle and cheliped muscle, respectively.

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Mytilus edulis Modiolus neozelandicus	0.3-1.6(dry wt) 0.04(dry wt)	New Zealand	Nielsen & Nathan (1975)
Mytilus edulis	0.1-0.9(wet wt)	Scotland	Topping (1973)
Mytilus edulis	0.2-18.2(wet wt)	Victoria, Australia	Phillips (1976)
Mytilus galloprovincialis	0.4-5.9(dry wt)	N.W. Mediterranean	Fowler & Oregioni (1976)
Mytilus edulis	1.4-4.2(dry wt)	St. Croix est., Me.	Fink et al. (1976)
Mytilus edulis	0.9-1.8(dry wt) 1.7 (dry wt)	Maine Narrangansett Bay	Goldberg et al. (1978)
Mytilus edulis Modiolus modiolus	2.9-112.2(dry wt) 15.2-55.6(dry wt)	Belledune area	This study

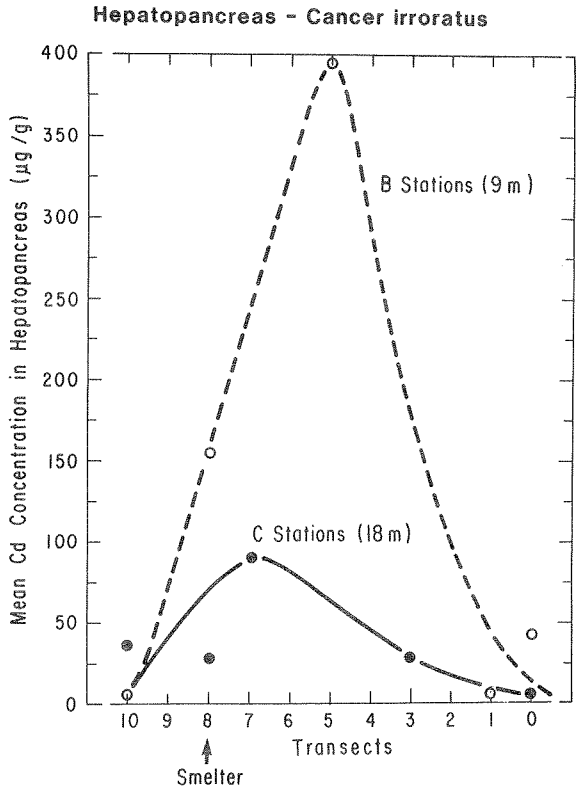

Fig. 9. Mean cadmium (Cd) concentrations in the hepatopancreas of C. irroratus sampled at B (9 m) and C (18 m) stations.

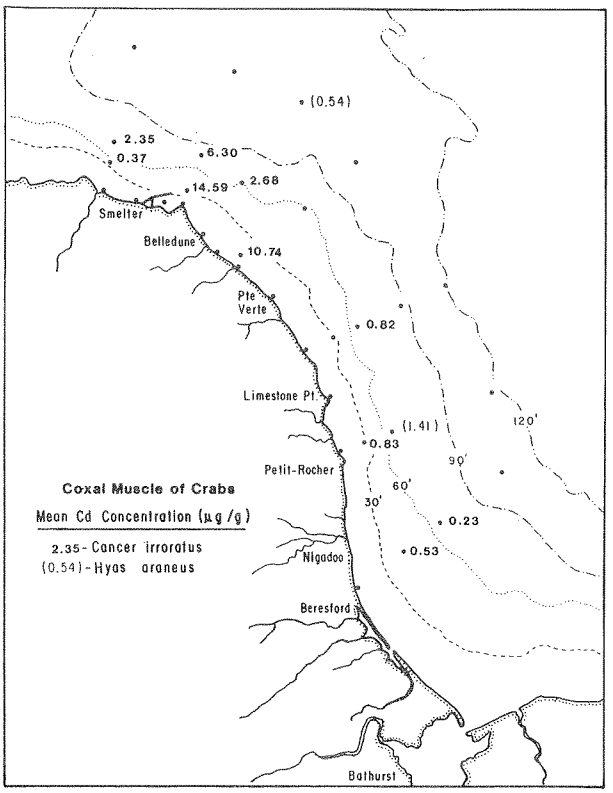

Fig. 11. Mean cadmium (Cd) concentrations in the coxal muscle of C. irroratus and H. araneus sampled from the survey area.

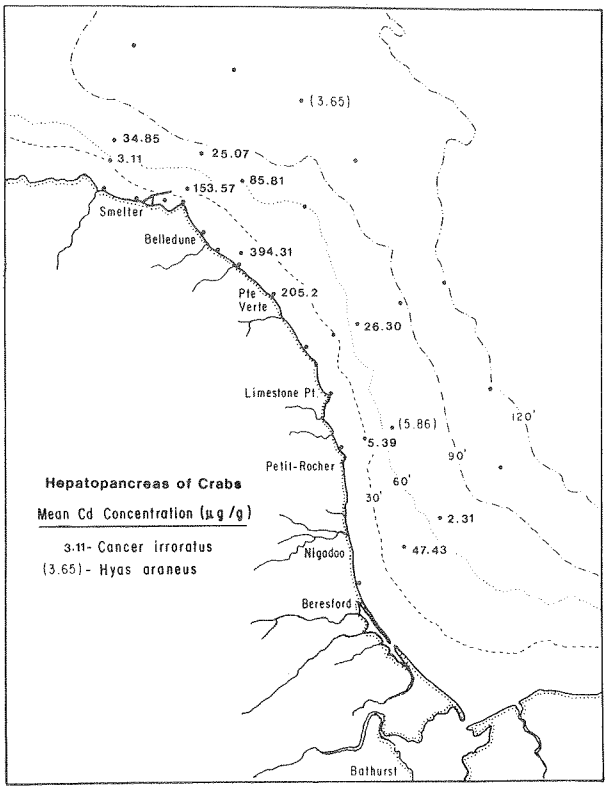

Fig. 10. Mean cadmium (Cd) concentrations in the hepatopancreas of C. irroratus and H. araneus sampled from the survey area.

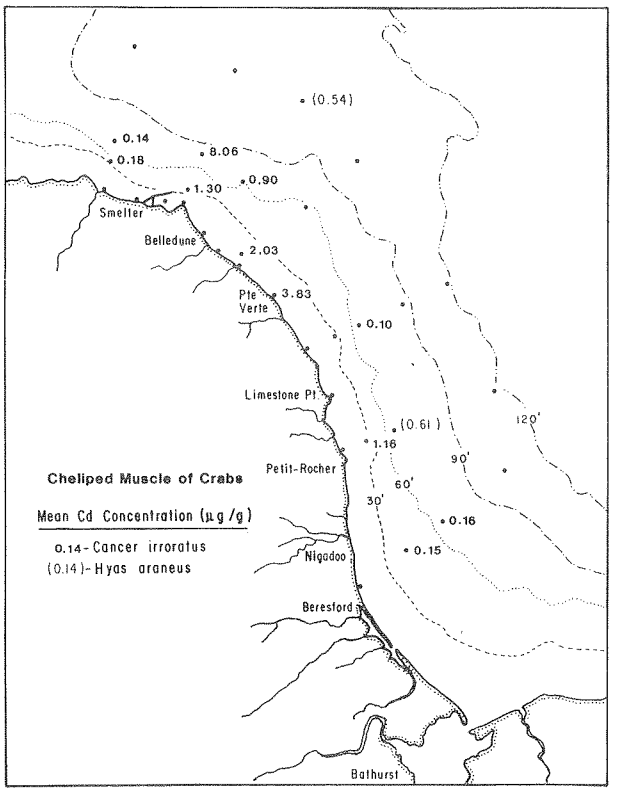

Fig. 12. Mean cadmium (Cd) concentrations in the cheliped muscle of C. irroratus and H. araneus sampled from the survey area.

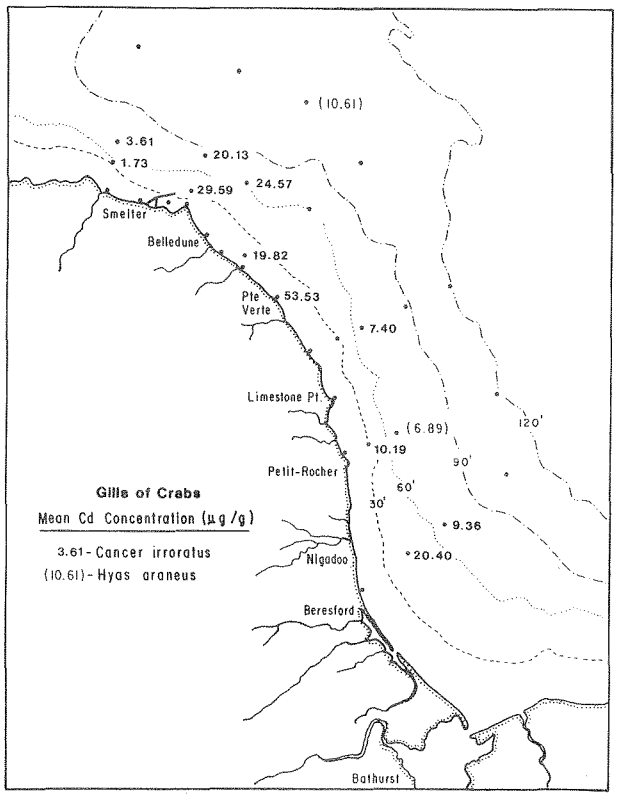

Fig. 13. Mean cadmium (Cd) concentrations in the gills of C. irroratus and H. araneus sampled from the survey area.

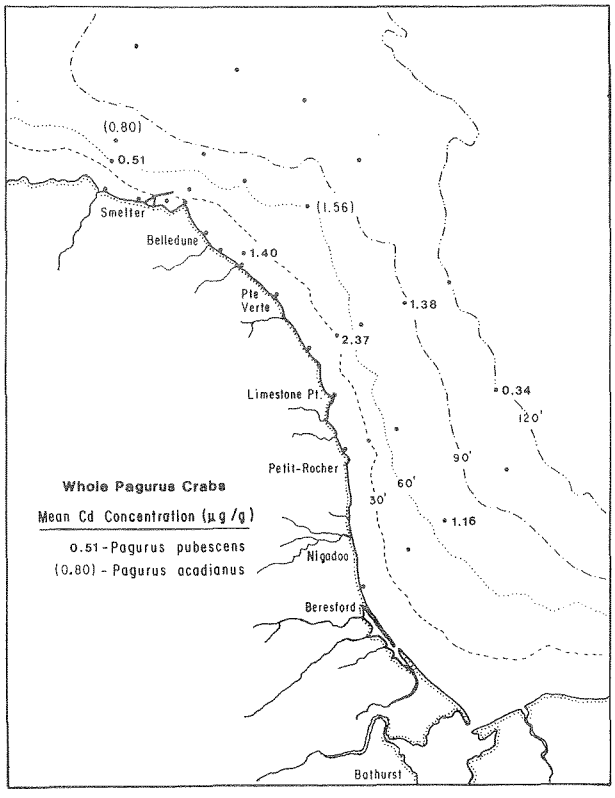

Fig. 14. Mean cadmium (Cd) concentrations in whole P. pubescens and P. acadianus sampled from the survey area.

same as found in C. irroratus from the survey area. Gill tissue from C. irroratus in the present study contained proportionally higher concentrations of Cd in relation to hepatopancreas tissue than did C. pagurus analyzed by Overnell and Trewhella (1979).

PERIWINKLES

Littorina sp. were collected at several shore (A) stations within the survey area. The mean Cd concentrations in soft tissue (Fig. 15) are higher at transects 5 and 6 than at transects 1, 9 and 10.

The mean and range of Cd concentrations in periwinkles from the entire survey area were 17.8 (1.1-44.0) µg/g. This is higher than Cd levels reported for Littorina species from other marine areas (Table 4), with the exception of L. littoralis collected from the polluted Severn estuary (Leatherland and Burton 1974) and Bristol Channel (Nickless and Stenner 1972) in the U.K.

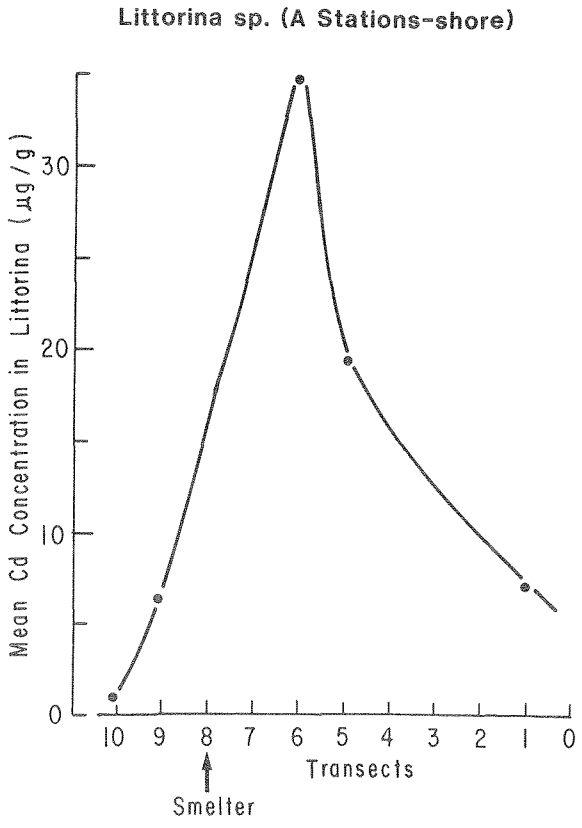

Fig. 15. Mean cadmium (Cd) concentrations in soft parts of Littorina sp. sampled at A stations (shore).

BARNACLES

The highest mean Cd concentrations in Balanus balanoides collected at shore (A) stations were from animals collected at transect 3 (Fig. 16). However, mean concentrations decreased at transect 5 and rose to a second maximum at transect 7.

The mean and range of Cd concentration in B. balanoides from the survey area were 1.5 (0.2-3.9) µg/g. There is little published information on Cd

Table 3. Cadmium (Cd) concentrations reported for tissues of crab species.

Species	Cd concentration - mean or range (µg/g)	Location	Reference
Cancer pagurus - whole	0.4(wet wt)	Northumberland, UK	Wright (1976)
C. pagurus - muscle	3.6-5.2(wet wt)	Scotland	Topping (1973)
C. pagurus - muscle	5.4(wet wt)	Devon, UK	Reynolds & Reynolds (1971)
- hepatopancreas	30.0(wet wt)		
C. pagurus - gill	0.8-1.0(wet wt)	Orkney Is., Scot.	Overnell & Trewhella (1979)
- muscle	0.4-0.9(wet wt)		
- hepatopancreas	2.0-17.8(wet wt)		
C. irroratus - gill	0.7-2.7(wet wt)	American Atlantic	Greig et al. (1977a)
- muscle	0.1-1.0(wet wt)	- dumpsites	
- hepatopancreas	1.1-4.8(wet wt)		
C. irroratus - gill	4.2-24.1(dry wt)	Passamaquoddy Bay,	Ray et al. (unpubl.)
- coxal muscle	0.5-6.6(dry wt)	N.B.	
- cheliped muscle	0.02-0.2(dry wt)		
- hepatopancreas	8.8-115.0(dry wt)		
C. irroratus - gill	1.7-53.5(dry wt)	Belledune area	This study
- coxal muscle	0.2-29.2(dry wt)		
- cheliped muscle	0.1-8.1(dry wt)		
- hepatopancreas	2.3-394.3(dry wt)		
H. araneus - gill	1.3-19.4(dry wt)	Belledune area	This study
- coxal muscle	0.4-1.4(dry wt)		
- cheliped muscle	0.2-0.9(dry wt)		
- hepatopancreas	2.6-5.9(dry wt)		
Pagurus acadianus	0.3-2.4(dry wt)	Belledune area	This study
P. pubescens	0.5-1.5(dry wt)	Belledune area	This study

Table 4. Cadmium (Cd) concentrations reported for tissues of Littorina species.

Species	Cd concentration - mean or range (µg/g)	Location	Reference
L. littorea	0.9(dry wt)	Southampton, UK	Leatherland & Burton (1974)
L. littoralis	178.0(dry wt)	Severn est., UK	
L. littorea	0.03-0.5(wet wt)	Scotland	Topping (1973)
L. littorea	2.0(dry wt)	English Channel,UK	Bryan (1976)
L. littorea	1.1-2.1(dry wt)	St. Croix est. Me.	Fink et al. (1976)
L. littorea	ND-210.0(dry wt)	Bristol Channel,UK	Nickless & Stenner (1972)
L. littorea	0.9-7.3(wet wt)	UK, several areas	Howard & Nickless (1978)
L. littorea	1.1-44.0(dry wt)	Belledune area	This study

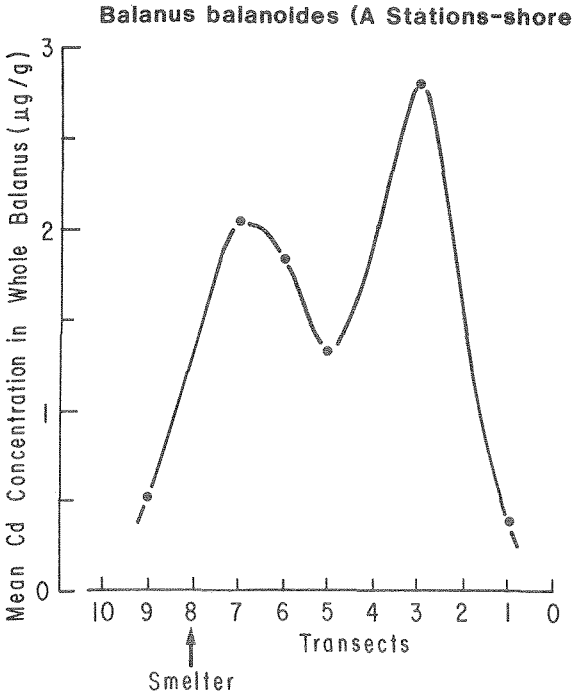

Fig. 16. Mean cadmium (Cd) concentrations in whole B. balanoides sampled at A stations (shore).

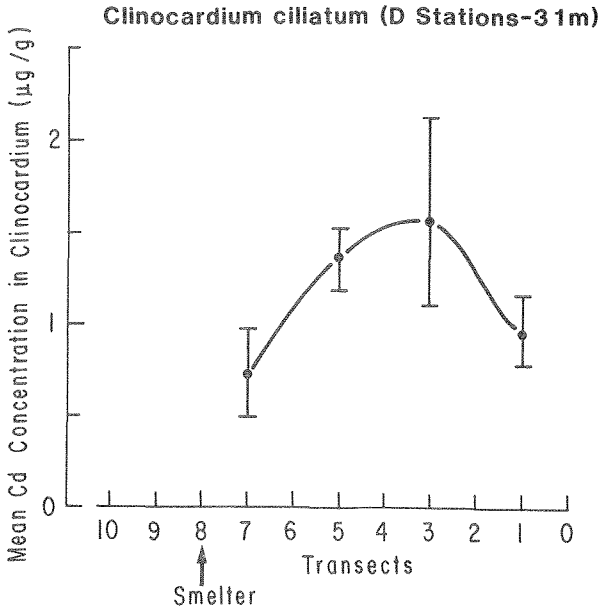

Fig. 17. Mean cadmium (Cd) concentrations in soft parts of C. ciliatum sampled at D stations (31 m).

levels in barnacles from other marine habitats, but Stenner and Nickless (1975) reported concentrations of 4.5-12.1 µg/g dry weight for whole Balanus perforatus and B. amphitrites from the polluted Rio Tinto area of Spain. Semibalanus balanoides concentrated Cd by a factor of approximately 1200 during 30 d exposure to seawater solutions of Cd (Rainbow et al. 1980).

COCKLES

Clinocardium ciliatum were collected at four D stations (31 m). The mean Cd levels in cockles from transects 3 and 5 appear greater than levels in samples from transects 1 and 7 (Fig. 17). The mean and range of Cd concentration for all samples were 1.2 (0.5-2.1) µg/g. This is slightly higher than the mean Cd concentration of 0.8 µg/g dry weight reported for Cardium edule collected from Solway Firth, U.K. (Leatherland and Burton 1974).

MISCELLANEOUS FAUNAL SPECIES

Polychaetes, flatfish, quahogs (Mercenaria mercenaria), edible whelk (Buccinum undatum), and American pelican's foot (Aporrhais occidentalis) were not collected at a sufficient number of sites to indicate the distribution of Cd within the survey area. A comparison between the mean Cd concentrations in these biota (Table 5) and related species from other marine areas (Table 6) indicates that the polychaetes and flatfish are not excessively contaminated with Cd, whereas M. mercenaria from station 5B and the two large gastropod species appear to contain elevated levels of Cd.

MACROPHYTIC ALGAE

The brown algae, Fucus vesiculosus, Ascophyllum nodosum and Laminaria digitata, were primarily collected from shore (A) stations in the survey area. The fronds of F. vesiculosus and A. nodosum, which are zones of rapid growth, were separated from the stem, since the uptake of heavy metals by seaweeds appears related to growth (Bryan 1969). It is not certain whether samples of L. digitata from each station were always taken from the same portion of the kelp blade.

The mean concentration of Cd in L. digitata for the survey area is 0.8 (0.4-1.8) µg/g. No published data have been found for Cd levels in L. digitata from other areas, but Bryan (1969) reported copper concentrations of 1.6 µg/g in L. digitata from the Seaton area of Britain. Since Cd and copper have a similar affinity for alginate extracts of L. digitata (Haug 1961), it appears that concentrations of Cd in the kelp from the vicinity of the smelter may not be excessive. The distribution of Cd concentrations in L. digitata (Fig. 18) does not conform to a discernible pattern, possibly as a result of non-uniformity in the sampling of the kelp blade.

The mean and range of Cd concentrations in F. vesiculosus samples from the survey area were 8.6 (1.7-18.2) µg/g and 10.2 (1.3-30.3) µg/g for stems and fronds, respectively. Similarly, mean and range of concentrations in A. nodosum stems and fronds were 6.2 (0.3-13.7) and 5.5 (0.6-18.0) µg/g. These concentrations fall within the upper range of Cd levels in F. vesiculosus and A. nodosum from other marine environments (Table 7). The Cd concentrations at each station for these species are illustrated in Fig. 19-21. With the exception of 4 out of 14 samples, concentrations were higher in fronds than in stems. Cd levels in F. vesiculosus from shore stations (Fig. 19a) were at a maximum at transect 7. Cd concentrations in A. nodosum are less variable, but they follow the same general pattern (Fig. 19b).

Table 5. Mean cadmium (Cd) concentrations in flatfish, polychaetes and some mollusc species from the vicinity of the Brunswick Mines smelter, N.B.

Species	Mean Cd concentration (µg/g) by dry wt	by wet wt	Stations sampled
Molluscs			
Mercenaria mercenaria	9.8(6.7-12.9)	0.6(0.3-0.8)	5B
Buccinum undatum	39.6(0.4-137.8)	4.3(0.1-16.3)	0C, 0D
Aporrhais occidentalis	14.3(0.2-98.3)	3.2(0.03-22.2)	1C, 3D, 7C, 7D, 8C, 10C
Polychaetes			
Arabellid	1.9(1.1-2.6)	-	0C, 8D
Nephtyid	1.1(0.8-1.6)	-	3C, 3D
Phyllodoce sp.	1.2(0.6-1.8)	-	1D, 10D
Clymenella sp.	0.4	-	3D
Lepidonotes squamatus	2.9(2.2-3.6)	-	1D, 10D
Aphrodite hastata	1.1(0.2-2.0)	-	3C
Flatfish (viscera)			
Hippoglossoides platessoides	0.9(0.3-2.0)	-	0C, 1C, 8B
Limanda ferruginea	0.6(0.4-0.9)	-	0C, 1C, 10C
Pseudopleuronectes americanus	2.0	-	3C
Flatfish (muscle)			
H. platessoides	0.10(0.02-0.40)	0.012(.004-.03)	0C, 1C, 8B
L. ferruginea	0.07(0.05-0.10)	0.005(0.002-0.01)	0C, 1C, 10C
P. americanus	0.05	0.005	3C

Table 6. Cadmium (Cd) concentrations reported for flatfish, polychaetes and some mollusc species.

Species	Cd concentration - mean or range (µg/g)	Location	Reference
Molluscs			
Mercenaria mercenaria	2.1(dry wt)	Irish Sea	Segar et al. (1971)
M. mercenaria	0.1-0.2(wet wt)	U.S. Atl. coast	Zook et al. (1976)
Buccinum undatum	2.2(dry wt)	Irish Sea	Segar et al. (1971)
Buscyon canaliculatum	0.1-0.2(wet wt)	U.S. Atl. coast	Greig et al. (1977)
Polychaetes			
Glycera dibranchiata	1.0-2.0(dry wt)		
Nereis diversicolor	0.2-4.3(dry wt)	St. Croix est., Me.	Fink et al. (1976)
Nereis virens	0.7-2.2(dry wt)		
Nephtys sp.	1.9(dry wt)		
N. virens	0.3-0.5(dry wt)	Passamaquoddy Bay, N.B.	Ray et al. (in press)
N. diversicolor	0.1-3.6(dry wt)	S.W. England	Bryan & Hummerstone (1973)
Flatfish (muscle)			
Limanda ferruginea	ND-0.06(wet wt)	Georges Bank	Zook et al. (1976)
Limanda limanda	ND-0.21(dry wt)	German Bight	Westernhagen et al. (1980)
Pleuronectes platessa	ND-0.16(dry wt)	German Bight	
P. platessa	<.002-.01(wet wt)	Lowestoft, UK	Pentreath (1977)

Table 7. Cadmium (Cd) concentrations reported for macrophytic algae.

Species	Cd concentration range (µg/g dry wt)	Location	Reference
Fucus vesiculosus (frond)	3.8-25.6	Bristol Channel,UK	Fuge and James (1974)
F. vesiculosus (whole)	7.0-13.0	Norway	Melhuus et al. (1978)
Ascophyllum nodosum (whole)	0.4-14.3	Norway	Haug et al. (1974)
F. vesiculosus (whole)	1.8-2.1	Near Liverpool, UK	Foster (1976)
A. nodosum (whole)	1.5-1.8		
F. vesiculosus (frond)	1.1-1.4	UK, several areas	Preston et al. (1972)
F. vesiculosus (whole)	1.3-2.2	St. Croix est., Me.	Fink et al. (1976)
A. nodosum (whole)	0.8-1.2		
Laminaria digitata	0.4-1.8	Belledune area	This study
F. vesiculosus - stems	1.3-18.2	Belledune area	This study
- fronds	1.3-30.3		
A. nodosum - stems	0.3-13.7	Belledune area	This study
- fronds	0.6-18.0		

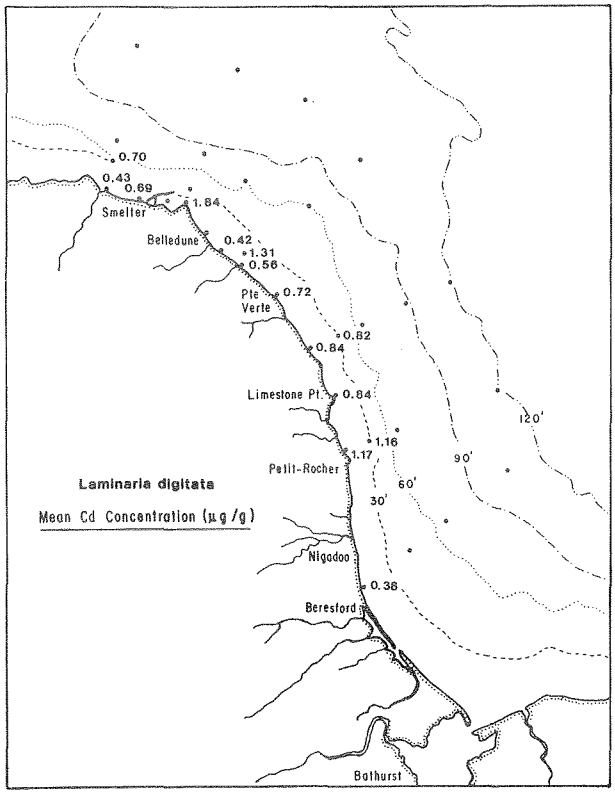

Fig. 18. Mean cadmium (Cd) concentrations in L. digitata sampled from the survey area.

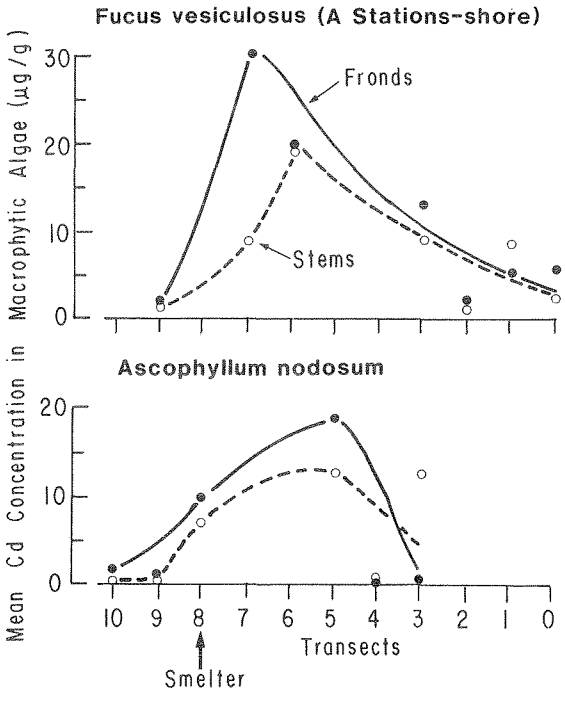

Fig. 19. Mean cadmium (Cd) concentrations in F. vesiculosus and A. nodosum sampled at A stations (shore).

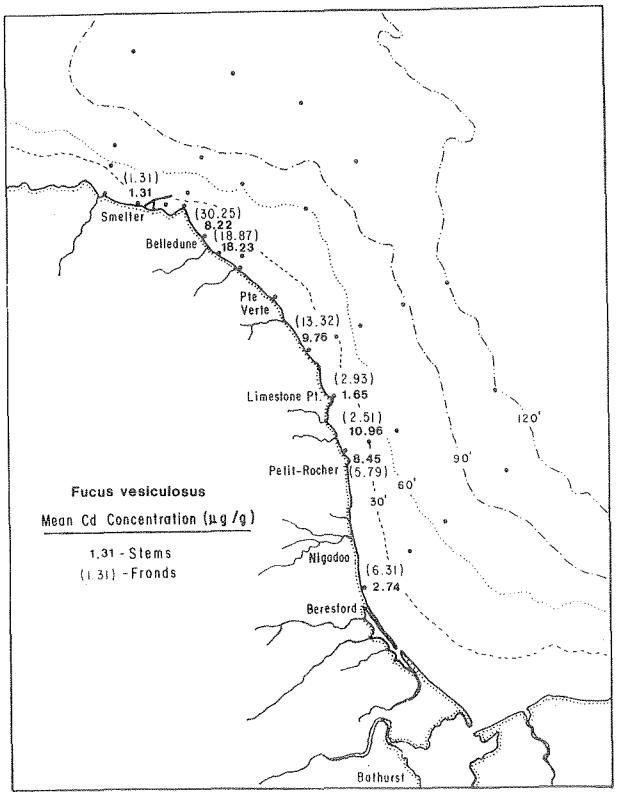

Fig. 20. Mean cadmium (Cd) concentrations in F. vesiculosus sampled from the survey area.

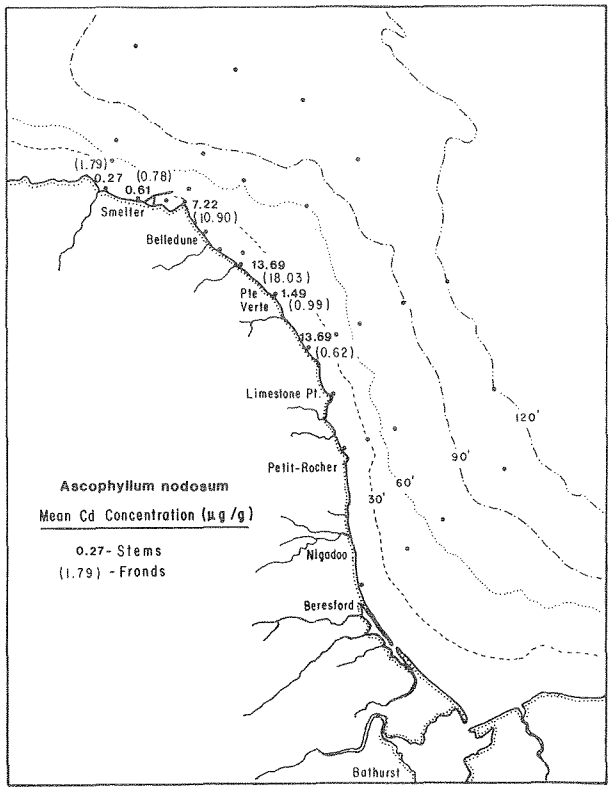

Fig. 21. Mean cadmium (Cd) concentrations in A. nodosum sampled from the survey area.

SEDIMENTS

Each sediment sample was separated into three particle size fractions representing silt-clay (<63 µm), fine sand (63-250 µm) and coarse sand and gravel (>250 µm). Shore (A) sediments consisted of fine and coarse sand, and sediments from B, C and D stations contained varying proportions of all three fractions (Table 8). Silt-clay fractions from C, D and some B stations were analyzed for Cd. The fine sand fractions from A stations and selected B and C stations were also analyzed.

At B and C stations, Cd concentrations in silt-clay fractions (Table 8) reach a maximum at transects 8 and 5, respectively. The same trend is obvious in fine sand fractions from these stations. Similarly, Cd in silt-clay from D stations is slightly elevated at transects 3 and 5.

In samples from shore (A) stations, Cd levels in fine sand (Table 8) are variable, possibly as a result of erosion and deposition processes on the shore. However, there is a trend to higher Cd concentrations at transects 4 through 7.

A comparison of the range of Cd concentrations in sediments from this study (0.1-7.1 µg/g) and from other marine sediments (Table 9) indicates that the sediments in the survey area are moderately contaminated, but not as grossly contaminated as those reported for other areas by Bloom and Ayling (1977) and Bower et al. (1978). This may be partly because there are no fine fluvial sediments in the Belledune area.

The data of Loring et al. (this report) approximate the Cd levels found in sediments from this survey. The slightly lower concentrations in this study may be attributed to the smaller silt-clay fraction (<53 µm) analyzed by Loring.

CONCLUSIONS

The data for Cd concentrations in the silt-clay fraction of the sediments (Fig. 22) indicate a band of contaminated sediment extends diagonally from Belledune Harbour to the southeast across the sampling area. The data further show a decline in the Cd concentration with depth of the water column.

Table 10 shows that minimum concentrations of Cd were generally found in animals at the extremes of the survey area (transects 0, 1 and 10) and maximum concentrations were found in animals from the center of the area (transects 3-8). Results of t-tests show that the differences between minimum and maximum mean Cd concentrations were significant ($P < 0.05$) for P. magellanicus meats and mantles, C. ciliatum, M. edulis, Littorina sp. and B. balanoides (Table 10). Differences between mean concentrations in P. magellanicus viscera, B. undatum and C. irroratus coxal muscle were not significant because of high variability between samples and small sample size. Data for biota in which only one sample was taken per station could not be tested.

The diagonal pattern of contamination, as noted for sediment, also seems to apply to Cd concentrations in biota. As discussed throughout the text, Cd concentrations in the non-motile species (M. edulis, B. balanoides, L. digitata, F. vesiculosus, Littorina sp. and A. nodosum) collected at shore

Table 8. Percent fraction of the whole sediment and cadmium (Cd) concentration in the silt-clay and fine sand fractions.

Station	Silt-clay Fraction (%)	Silt-clay Cd (µg/g)	Fine sand Fraction (%)	Fine sand Cd (µg/g)
0A	-	-	96.4	0.8
1A	-	-	-	-
2A	-	-	48.8	0.6
3A	-	-	1.7	0.2
4A	-	-	0.3	2.2
5A	-	-	4.1	1.5
6A	-	-	2.6	0.9
7A	-	-	1.1	1.1
8A	-	-	-	-
9A	-	-	5.7	0.3
10A	-	-	1.2	0.2
0B	5.2	1.9	8.4	-
1B	-	-	27.5	1.0
3B	-	-	-	-
5B	0.4	1.5	83.7	0.4
8B	12.4	7.1	67.3	5.7
10B	0.2	1.0	89.8	-
0C	0.9	0.9	63.9	0.1
1C	14.5	0.4	50.4	0.2
3C	6.8	1.4	37.5	2.8
5C	7.2	5.6	40.1	6.3
7C	1.8	1.8	14.9	0.3
8C	1.3	1.7	41.0	-
10C	2.7	1.1	15.2	0.4
0D	2.5	0.5	12.9	-
1D	9.3	0.4	22.1	-
3D	2.2	0.8	38.6	-
5D	4.7	0.8	39.8	-
7D	8.9	0.4	36.1	-
8D	8.9	0.4	20.8	-
10D	15.5	0.3	23.8	-

Silt-clay - <63 µm
Fine sand - 63-250 µm

Table 9. Cadmium (Cd) concentrations reported for marine sediments.

Sediment fraction	Cd concentration - mean or range (µg/g dry wt)	Location	Reference
Total	0.6-0.7	Southampton, UK	
Total	3.1	Severn est., UK	Leatherland and Burton (1974)
Total	4.2	Mersey est., UK	
<500 µm	2.2-6.7	Dalhousie Hbr., N.B.	Ray et al. (in press)
<204 µm	ND-4.0	Solway Firth, UK	Perkins et al. (1973)
Total	0.8-1.3	Solway Firth, UK	Taylor (1976)
Total	<0.2-4.2	S.W. England	Bryan and Hummerstone (1973)
Total	ND-4.2	Long Is. Sound, N.Y.	Greig et al. (1977b)
Total	<1.0-15.0	Raritan Bay, N.J.	Greig and McGrath (1977)
<16 µm	0.6-20.0	Rhine-Neuse est., North Sea	Salomons and Mook (1977)
Total	<0.5-1400	Derwent est., Aust. - Zn smelter	Bloom and Ayling (1977)
Total	1-908	Hudson R. est., N.Y. - battery factory	Bower et al. (1978)
Total	0.2-850	Review	Bryan (1976)

Table 10. Ratios and t-tests of minimum and maximum mean cadmium (Cd) concentrations in selected biota and sediments from the survey area.

Species	Min. mean Cd conc. (µg/g)	Station #	Max. mean Cd conc. (µg/g)	Station #	Ratio max/min	t-test result
P. magellanicus						
- meat	1.0	8C	8.4	3C	8.4	Significant
- mantle	3.8	8C	18.0	5B	4.7	Significant
- viscera	129.2	10C	493.2	5B	3.8	Not significant
M. edulis	4.4	10C	112.2	6A	25.5	Significant
Littorina spp.	1.1	10A	35.0	6A	31.8	Significant
B. undatum	2.1	7C	63.7	8C	30.3	Not significant
C. irroratus						
- hepatopancreas	2.3	0C	394.3	5B	171.4	-
- coxal muscle	0.2	0C	14.6	8B	73.0	Not significant
- cheliped musc.	0.1	3C	8.1	8C	81.0	-
- gills	1.7	10B	53.5	4A	31.5	-
P. pubescens	0.3	1D	2.4	3B	8.0	-
B. balanoides	0.4	1A	2.8	3A	7.0	Significant
C. ciliatum	0.7	7D	1.6	3D	2.3	Significant
L. digitata	0.4	0A	1.8	8A	4.5	-
F. vesiculosus						
- fronds	1.3	10A	30.3	6A	23.3	-
- stems	1.3	10A	18.2	6A	14.0	-
A. nodosum						
- fronds	0.3	10A	18.0	5A	6.0	-
- stems	1.0	4A	13.7	5A	13.7	-
Sediment						
- silt/clay	0.3	10D	7.1	8B	23.7	-
- fine sand	0.1	0C	6.3	5C	63.0	-

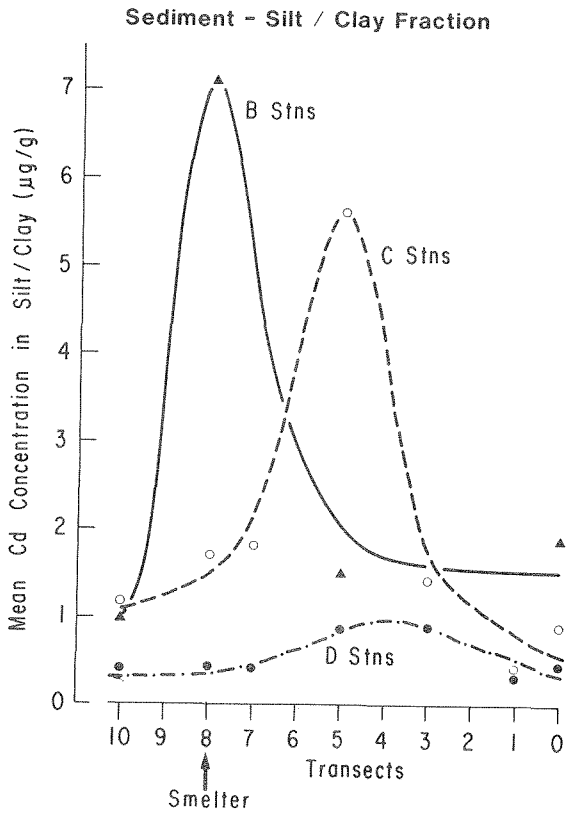

Fig. 22. Mean cadmium (Cd) concentrations in the silt/clay fraction of sediments sampled at B (9 m), C (18 m) and D (31 m) stations.

stations were elevated at transects 5 through 8. For crabs (C. irroratus) taken offshore at 9 and 18 m, maximum Cd concentrations were found at transects 5, 7 and 8. Scallops (P. magellanicus) at 18 m had maximum Cd concentrations at transect 5. Further offshore, at 31 m, C. ciliatum from transects 3 and 5 contained maximum Cd concentrations.

In comparison to data reported for biota from other marine areas, in this survey the filter-feeding bivalves (P. magellanicus, M. edulis, M. modiolus, C. ciliatum, M. mercenaria) contained high levels of Cd. As may be expected, predators upon bivalves (B. undatum, A. occidentalis) also contained very high Cd concentrations. In addition, the herbivorous gastropod, Littorina sp., the filter-feeding crustacean B. balanoides, and the carnivore/scavenger C. irroratus contained high levels of Cd. Of the biota analyzed, only macrophytic algae, polychaetes and flatfish species contained Cd levels near reported "baseline" concentrations. Low concentrations in the latter taxa may be related to their mobility, and low Cd in algae may indicate low levels of dissolved Cd in water in inshore areas.

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Appendix I

Scallops - *P. magellanicus* and *C. islandicus*

Dry weight-wet weight relationships:
Meat: wet wt = 0.3 + 5.0 (dry wt)
Viscera: wet wt = 2.5 + 6.0 (dry wt)
Mantle: wet wt = 0.6 + 9.2 (dry wt)

Station	Valve length (cm)	Dry wt (g) Meat	Viscera	Mantle	Cd (µg/g dry wt) Meat	Viscera	Mantle
P. magellanicus							
0C-1	18.0	3.6	1.2	1.1	7.9	192.0	9.1
0C-2	14.0	6.7	0.9	2.8	6.6	191.3	11.3
0C-3	8.5	0.9	1.6	0.6	1.5	92.6	5.7
3C	13.0	4.2	2.3	2.3	8.4	223.1	12.6
5B	13.5	5.1	1.2	1.9	7.7	493.2	18.0
5C-1	7.5	4.0	1.8	1.1	5.6	185.5	5.8
5C-2	8.3	2.2	2.5	0.3	4.4	223.9	17.6
5C-3	11.0	0.9	0.9	0.5	2.0	121.0	11.9
7C-1	7.5	0.9	0.9	0.3	3.0	111.6	8.1
7C-2	9.0	2.4	1.6	0.8	3.5	156.2	6.7
7C-3	12.0	5.2	1.0	1.3	4.2	233.5	9.4
8C-1	7.0	0.9	0.7	0.3	1.1	117.1	4.9
8C-2	11.0	3.9	0.9	1.1	1.7	159.8	3.3
8C-3	14.0	0.9	0.9	2.3	0.5	229.3	3.2
10C-1	6.0	0.5	0.5	0.1	1.1	87.6	6.6
10C-2	9.0	1.9	1.0	0.7	1.2	108.5	6.8
10C-3	13.5	6.0	1.6	2.0	2.4	191.5	3.2
C. islandicus							
8C-1	7.0	1.9	1.1	0.3	13.3	24.9	1.1
8C-2	8.5	1.9	1.3	0.6	1.0	93.4	2.3
10C-1	6.5	5.2	0.7	0.1	3.4	21.4	1.1
10C-2	7.0	1.1	1.0	0.4	0.5	29.8	1.4

Appendix I (cont'd)

Mussels - M. edulis and M. modiolus

Dry weight-wet weight relationship: Wet wt = 12.6 + 3.9 (dry wt)

Station	Valve length (cm)	Dry wt (g)	Cd (µg/g dry wt)
M. edulis			
0A-1	5.8	0.7	13.9
0A-2	6.3	1.5	12.2
1A	7P (4.3-6.2)[a]	1.9	19.0
2A	5P (6.1-6.7)	2.6	33.7
3A	5P (5.8-7.8)	2.6	30.0
3B-1	7.4	2.5	26.9
3B-2	6.4	1.7	25.5
3B-3	6.9	1.5	26.2
4A	4P (5.3-6.6)	2.1	48.6
5A	5P (5.7-6.4)	2.9	53.4
6A	5P (4.9-6.5)	2.0	112.2
7A	4P (4.7-7.4)	2.6	102.9
9A	10P (3.9-8.2)	2.0	20.3
10A	5P (6.3-8.0)	2.0	14.9
10B-1	6.6	1.9	6.6
10B-2	6.0	1.2	7.0
10B-3	8.2	2.3	9.0
10C-1	6.7	1.4	5.9
10C-2	5.8	0.5	2.9
M. modiolus			
1B-1	9.5	2.9	24.1
1B-2	7.7	1.6	15.2
5C	53.4	5.4	34.9
8C-1	29.0	3.9	24.4
8C-2	63.8	4.1	32.2
10C-1	111.1	15.6	55.9
10C-2	108.8	15.3	37.8

[a] $xP\ (n_1\text{-}n_2)$ refers to the range of valve lengths for a pooled sample of x number of animals.

Appendix I (cont'd)

Crabs - C. irroratus and H. araneus

Dry weight-wet weight relationships:
Hepatopancreas - Wet wt = 2.1 + 3.2 (dry wt)
Gills - Wet wt = 1.6 + 9.6 (dry wt)
Coxal muscle - Wet wt = 1.5 + 4.7 (dry wt)
Cheliped muscle- Wet wt = 0.8 + 4.5 (dry wt)

Station	Carapace length (cm)	Dry wt (g)				Cd (µg/g dry wt)			
		Hepato.	Gills	Cheliped	Coxal	Hepato.	Gills	Cheliped	Coxal
C. irroratus									
0B-1	8.2	0.8	0.4	1.4	0.3	81.4	21.4	0.2	0.6
0B-2	7.6	0.6	0.4	1.2	0.2	13.5	19.4	0.1	0.4
0C	4.8	0.1	0.1	0.1	0.1	2.3	9.4	0.2	0.2
1B	6.9	0.7	0.2	0.4	0.2	5.4	10.2	1.2	0.8
3C	7.6	0.7	0.4	0.9	0.1	26.3	7.4	0.1	0.8
5B	5.0	0.1	0.1	0.2	0.2	394.3	19.8	2.0	10.7
7C	5.3	0.5	0.2	0.6	0.2	85.8	24.6	0.9	2.7
8B-1	5.6	0.3	0.1	0.4	0.2	280.3	23.2	0.7	29.2
8B-2	7.5	1.1	0.3	1.4	0.4	56.1	12.0	2.4	9.9
8B-3	7.4	0.3	0.1	0.3	0.2	124.4	53.5	0.8	4.6
8C	5.5	0.5	0.2	0.5	0.2	25.1	20.1	8.1	6.3
10B	4.9	0.6	0.1	0.7	0.1	3.1	1.7	0.2	0.4
10C	6.8	0.7	0.2	0.3	0.1	34.9	3.6	0.1	2.4
H. araneus									
1C	7.5	0.6	0.1	0.3	0.2	5.9	6.9	0.6	1.4
7D-1	10.8	1.2	0.7	1.6	1.6	5.3	19.4	0.9	0.7
7D-2	9.5	3.7	0.2	2.5	1.5	3.1	1.3	0.2	0.4
7D-3	9.9	1.1	0.1	1.6	0.3	2.6	11.1	0.5	0.6

Polychaetes

Station	Species	Wet wt (g)	Dry wt (g)	Cd (µg/g dry wt)
0C	Arabellid	0.2	0.1	1.1
1D	Phyllodoce sp.	1.6	0.2	1.8
1D	Clymenella sp.	4.4	1.1	2.2
3C	Nephtyid	1.0	0.2	1.6
3C	A. hastata	56.7	8.0	2.0
3C	Nephtyid	1.2	0.3	0.8
3C	A. hastata	50.4	5.4	0.2
3D	Nephtyid	1.3	0.2	1.0
3D	L. squamatus	0.4	0.1	0.4
8D	Arabellid	0.3	0.1	0.3
10D	Phyllodoce sp.	1.4	0.3	0.6
10D	Arabellid	0.4	0.1	2.6
10D	Clymenella sp.	7.0	1.1	3.6

Appendix I (cont'd)

Flatfish

Dry weight-wet weight relationships:
Muscle - Wet wt = 2.8 + 8.1 (dry wt)
Viscera - Wet wt = 5.1 + 4.2 (dry wt)

Station	Species	Length (cm)	Whole wet wt (kg)	Sample dry wt (g) Muscle	Sample dry wt (g) Viscera	Cd(µg/g dry wt) Muscle	Cd(µg/g dry wt) Viscera
0C	L. ferruginea	29.0	0.25	0.3	4.6	0.05	0.44
0C	H. platessoides-1	34.0	0.36	1.0	5.0	0.03	0.26
0C	" -2	22.0	0.18	1.0	1.8	0.12	1.31
1C	L. ferruginea-1	23.0	0.14	0.6	0.8	0.05	0.93
1C	" -2	28.5	0.21	1.0	2.6	0.11	0.60
1C	H. platessoides-1	31.0	0.22	1.2	2.2	0.02	0.77
1C	" -2	27.0	0.18	0.7	1.7	0.06	0.36
3C	P. americanus	31.0	0.40	3.2	3.4	0.05	1.97
8B	H. platessoides	30.0	0.19	0.6	1.2	0.41	1.95
10C	L. ferruginea	29.0	0.28	0.6	4.8	0.07	0.55

Whelk - B. undatum

Station	Wet wt - soft parts (g)	Dry wt (g)	Cd (µg/g dry wt)
7C-1	16.9	2.7	0.4
7C-2	43.5	4.8	3.8
8C-1	10.5	1.4	2.0
8C-2	16.8	2.9	6.3
8C-3	23.4	1.9	137.8
8C-4	21.3	3.2	108.5
10C	17.8	3.3	18.2

American Pelican's Foot - A. occidentalis

Station	Wet wt - soft parts (g)	Dry wt (g)	Cd (µg/g dry wt)
0C	2.6 (2 animals)	0.4	0.2
0D	1.1	0.2	0.3
1C	2.9	0.5	0.4
3D	2.4	0.4	0.2
7C	1.2	0.3	98.3
7D	2.3	0.3	0.2
8C	6.4 (5 animals)	1.0	0.6

Appendix I (cont'd)

Quahog - M. mercenaria

Station	Valve length (cm)	Wet wt - soft parts (g)	Dry wt (g)	Cd (µg/g dry wt)
5B-1	10.4	196.9	12.8	12.9
5B-2	3.8	6.5	0.4	6.7

Crabs - P. pubescens and P. acadianus

Station	Species	Wet wt (g)	Dry wt (g)	Cd (µg/g dry wt)
0C	P. pubescens	2.1	0.5	0.9
0C	"	6.5	1.7	1.5
1D	"	17.4	4.0	0.3
3B	"	1.3	0.3	2.4
3C	"	8.3	2.6	1.4
5B	"	3.6(4 anim)	1.1	1.4
5C	P. acadianus	10.9	3.2	1.6
10B	P. pubescens	2.2(2 anim)	0.6	0.5
10C	P. acadianus	3.8	1.1	0.8

Cockles - C. ciliatum

Station	Valve length (cm)	Wet wt - soft parts (g)	Dry wt (g)	Cd (µg/g dry wt)
1D-1	4.8	22.1	2.2	0.8
1D-2	4.7	13.5	1.3	1.1
3D-1	4.7	15.2	1.4	1.1
3D-2	4.8	13.3	1.4	2.1
5D-1	4.5	9.3	0.9	1.5
5D-2	4.4	10.8	1.0	1.2
7D-1	4.5	11.4	1.5	0.5
7D-2	4.9	12.7	1.3	0.9

Appendix I (cont'd)

Barnacles (*B. balanoides*) and periwinkles (*Littorina* sp.)

Data for dry weight-wet weight relationships not available.

Barnacles			Periwinkles		
Station	Dry wt (g)[a]	Cd (µg/g dry wt)	Station	Dry wt (g)[a]	Cd (µg/g dry wt)
1A	2.5(4)	0.5	1A	3.1(4)	6.5
1A	2.3(4)	0.4	1A	3.3(3)	7.7
1A	2.1(4)	0.3	5A	0.6(2)	8.0
3A	0.4(1)	2.9	5A	0.4(1)	14.2
3A	1.4(2)	1.7	5A	0.4(1)	19.2
3A	1.7(2)	3.9	5A	0.5(1)	26.3
5A	0.4(1)	1.1	5A	0.5(1)	31.1
5A	1.2(4)	1.2	6A	0.2(2)	18.0
5A	1.3(5)	1.4	6A	0.3(2)	44.0
5A	1.5(5)	1.5			
6A	1.4(5)	1.1	6A	0.4(3)	43.0
6A	1.5(5)	2.9	9A	0.3(3)	2.0
6A	1.3(5)	1.7	9A	0.3(3)	10.9
6A	1.7(5)	1.5	10A	0.1(2)	1.1
7A	0.7(1)	2.1			
7A	1.1(6)	1.8			
7A	0.9(5)	2.2			
9A	1.1(6)	0.2			
9A	0.6(4)	0.7			
9A	1.9(5)	0.6			

[a]Data in parentheses refer to numbers of animals used in a pooled sample.

Appendix I (cont'd)

Macrophytic algae

Data for dry weight-wet weight relationships not available.

Station	Dry wt (g)	Cd (µg/g dry wt)
L. digitata		
0A	0.7	0.4
1A	5.0	1.2
1B	0.8	1.2
2A	3.5	0.8
3A	3.8	0.8
3B	2.2	0.8
4A	2.4	0.7
5A	3.5	0.6
5B	1.6	1.3
6A	3.1	0.4
8A	1.7	1.8
9A	1.2	0.7
10A	1.5	0.4
10B	2.6	0.7
F. vesiculosus - stem		
0A	0.9	2.7
1A	0.2	8.5
1B	1.1	11.0
2A	0.4	1.7
3A	0.5	9.8
6A	0.1	18.2
7A	0.9	8.2
F. vesiculosus - frond		
0A	3.9	6.3
1A	3.0	5.8
1B	7.0	2.5
2A	3.9	2.9
3A	8.1	13.3
6A	3.1	18.9
7A	4.0	30.3
9A	0.7	1.3
A. nodosum - stem		
3A	1.4	13.7
4A	5.1	1.5
5A	0.3	13.7
8A	3.5	7.2
9A	1.0	0.6
10A	1.3	0.3
A. nodosum - frond		
3A	7.4	0.6
4A	3.1	1.0
5A	3.9	18.0
8A	12.9	10.9
9A	7.7	0.8
10A	11.4	1.8

A PRELIMINARY SURVEY OF CIRCULATION AND HEAVY METAL CONTAMINATION IN BELLEDUNE HARBOUR AND ADJACENT AREAS

by

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INTRODUCTION

Following a request from the Fisheries Management of the Department of Fisheries and Oceans, units of Ocean and Aquatic Sciences, Atlantic, conducted a preliminary survey of the circulation and heavy metal contamination of sediments, water and suspended matter in the marine environment surrounding Belledune Harbour, N.B. The field work was conducted using the vessel NAVICULA between May 6 and May 8, 1980. Twenty-two grab samples and six water samples were collected at selected sites in Belledune Harbour (Fig. 1a) and a further 13 grab and 12 water samples were obtained from coastal zone transects both to the west and east of Belledune Harbour (Fig. 1b). At each location from which water samples were collected, a sample of suspended particulate matter (SPM) was also obtained. A vertical profiling current meter was used to obtain hydrographic information.

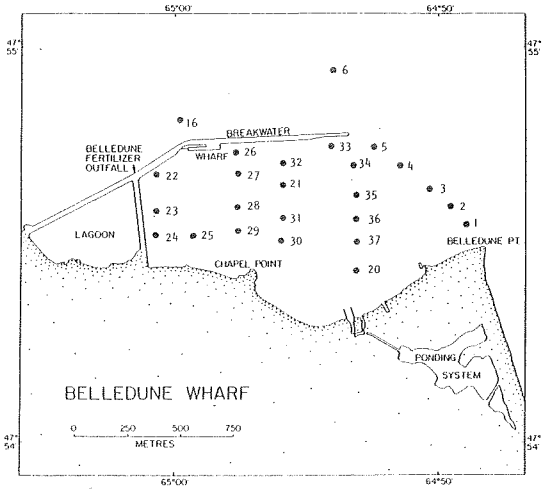

Fig. 1a. Station locations in Belledune Harbour.

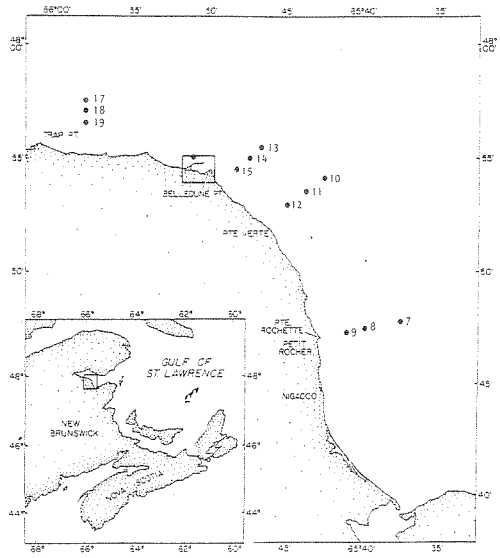

Fig. 1b. Station locations on coastal zone transects.

This document is a preliminary report of the circulation studies, sediment and suspended matter results, size analyses of sediment particles and water analyses, respectively.

PHYSICAL OCEANOGRAPHY

This note summarized the physical oceanographic data obtained by the Bedford Institute in 1964 (Canadian Hydrographic Service 1965) and 1980. For the dispersal and movement of water-borne contaminants, the following processes are thought to be important (no relative scale):

(1) Nature and strength of the coastal circulation;

(2) Response of the harbour to external events;

(3) Circulation within the harbour, source points of contaminants, and exchange with the coastal zone (5 km).

NATURE AND STRENGTH OF THE COASTAL CIRCULATION

There are several current systems in the nearshore zone which dominate the water movements. We will confine ourselves to discussing two: (a) longshore currents produced by an oblique wave approach to the shoreline; (b) coastal currents due to freshwater runoff.

(a) Longshore currents due to waves - this type of current is significant in that it is responsible for the net transport of sand or other beach material along the shore. Belledune Point is subjected to wave action from a sector ranging from NW through north to southeast; however, only the eastern fetch has considerable length. The wave characteristics for this sector range from 2.1 m @ 5 s to 5.8 m @ 9 s. The longshore current for waves coming from the east is shown in Fig. 2. Note the convergence zone on the east side of the Point. It should be noted that the wave action on the eastern side of Belledune Point is usually heavier than on the western side, giving rise to a net littoral drift which is predominantly westward. However, other current systems may modify this pattern.

(b) Coastal currents due to freshwater runoff - fresh water discharged by the Restigouche River (600 m^3/s) and smaller coastal tributaries set up a flow along the southern coast. Current measurements in 1964 and spot measurements at some of the locations shown in Fig. 1 show that typical rates in the surface (0-10 m) layer are of the order 10-20 cm/s, directed to the east, and following the coastline. Large variations in those speeds could however occur, achieving maximum values of 1.6 knots. Figure 3a shows a qualitative picture of the coastal flow determined from parachute drogues released at regular intervals during August 1964. In general, the flow offshore tends to be aligned with the shoreline; some flow separation may occur due to the projection of the breakwater into the stream. If we assume that this

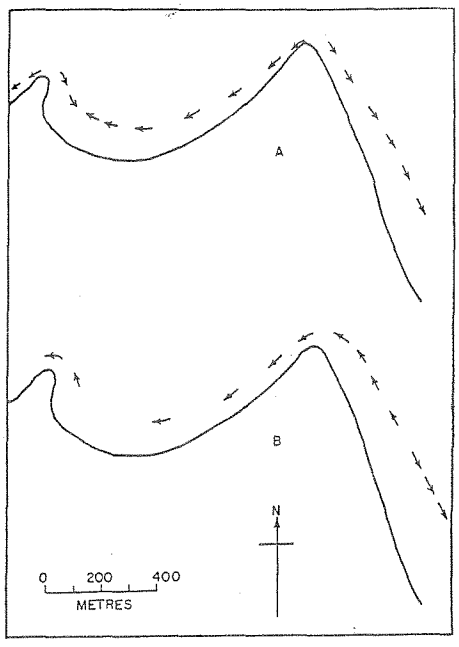

Fig. 2. Direction of longshore current. A - for waves from NE; B - for waves from east.

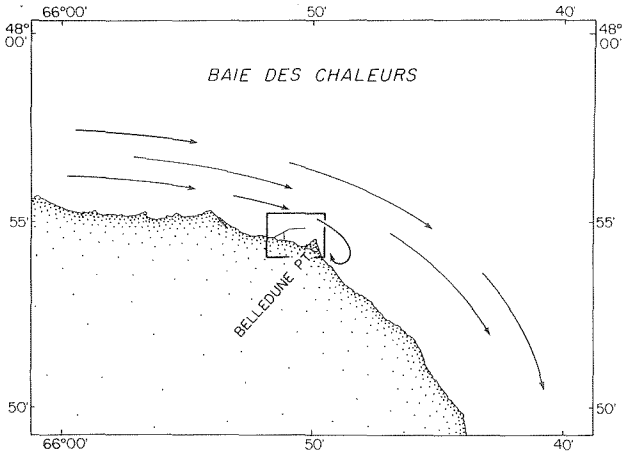

Fig. 3a. Coastal currents due to freshwater runoff in the vicinity of Belledune Point.

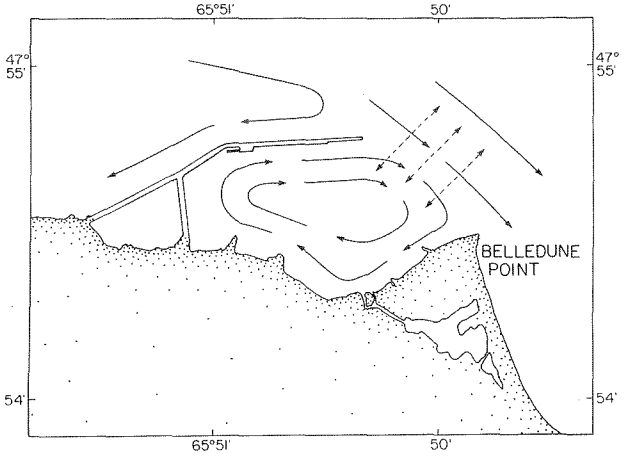

Fig. 3b. Circulation pattern in Belledune Harbour.

flow is decoupled from those waters below 10 m, an estimate of the width of this current can be made. Measurements in May 1980 show a typical density contrast of 2 x 10^{-4}. Assuming a rate of 15 cm/s and thickness of 10 m, the width of this current will be about 2 km.

RESPONSE OF THE HARBOUR TO EXTERNAL EVENTS

During May 1980, a sub-surface pressure gauge was installed at the inner end of the wharf. This gauge measured total pressure (atmospheric + pressure due to fluctuations in mass above the instrument) every 30 s over a time span of 2 d. Figure 4 gives the variation of equivalent water height for the duration. Note that the only significant variations are due to tides. Seiching, which could contribute to the flushing of the harbour, is either absent or insignificant during the sampling period.

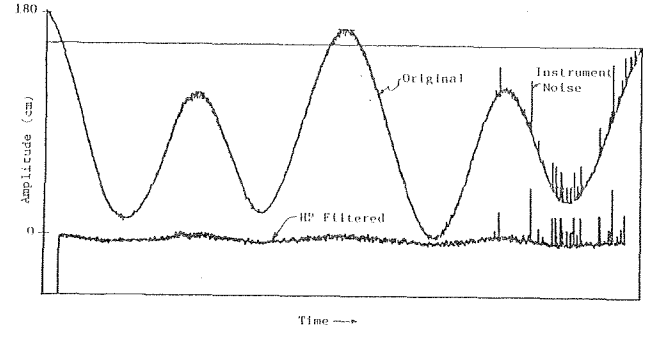

Fig. 4. Water level time history - Belledune Harbour.

CIRCULATION WITHIN THE HARBOUR

The coastal flow due to freshwater discharge is of sufficiently large scale that it does not "see" the opening of the harbour. However, a flow past its mouth could introduce a circulation pattern as shown in Fig. 3b. If this picture is correct, it tends to indicate that the exchange between harbour and coastal flow is limited to diffusive effect only; for all practical purposes the harbour is a closed system. However, this pattern could be altered when easterly or northeasterly winds disturb the coastal flow.

BOTTOM SEDIMENTS

After removal of material >2 mm, a representative part of each sediment sample was oven-dried for chemical analysis. From another portion of the sample, the sand particles (2.0-0.053 mm diam.) and mud size particles (<0.053 mm diam.) were separated by wet sieving, and the fraction <0.053 mm or 53 µm was retained for chemical analysis. The amounts (by weight) of the sand and mud size material in each sample (Table 1) were also used to classify the textural characteristics of the sediments according to the nomenclature of Loring and Rantala (1977) and to estimate the partition of cadmium (Cd) in the sediments (Table 1).

Table 1. Cadmium (Cd) in Belledune Harbour and coastal sediments.

Station no.	Sediment (ppm Cd)	Mud (ppm Cd)	% mud (<53 μm) (in sediments)	Suspended particulate matter (ppm Cd)
Lagoon	12.6	13.0	64.8	
Slag	53.6	-	-	
1 Harbour	not analyzed sand	-	sand	6.6
2 Harbour	9.2	7.9	58.6	
3 Harbour	9.2	15.6	42.8	5.7
4 Harbour	9.5	13.8	33.4	
5 Harbour	13.3	19.6	56.1	15.2
6 Coastal	1.8	4.4	33.8	2.6
7	.1	.4	36.8	6.2
8 Coastal	.2	.6	43.6	3.1
9 Coastal	.5	1.2	48.6	
10 Coastal	not analyzed gravelly sand	1.1	gravelly sand	2.8
11 Coastal	.2	.6	24.8	2.1
12 Coastal	.2	1.6	9.3	3.5
13 Coastal	not analyzed (rocks)		rocks	4.0
14 Coastal	.2	1.1	13.4	4.6
15 Coastal	1.8	8.5	30.3	2.4
16 Coastal	2.8	6.6	42.7	5.0
17 Coastal	.2	.4	52.1	2.9
18 Coastal	.2	.6	32.1	2.9
19 Coastal	.3	.8	37.5	3.4
20 Harbour	16.9	27.8	26.9	96.0
21 Harbour	30.0	40.2	50.8	23.6
22 Harbour	60.8	54.8	83.4	
23 Harbour	49.4	61.5	50.2	
24 Harbour	60.8	63.5	65.2	
25 Harbour	39.6	44.6	44.9	
26 Harbour	54.2	74.0	54.1	
27 Harbour	46.6	38.4	83.5	
28 Harbour	34.0	34.4	86.3	
29 Harbour	21.0	28.3	38.6	
30 Harbour	21.3	28.0	55.4	
31 Harbour	46.6	45.4	69.6	
32 Harbour	41.0	37.2	84.5	
33 Harbour	32.2	34.8	79.9	
34 Harbour	10.6	17.6	43.2	
35 Harbour	14.6	19.2	47.7	
36 Harbour	16.0	23.3	50.2	
37 Harbour	16.4	21.8	47.1	

The harbour sediments contain <5-86% by weight mud and 14->95% sand. Texturally they vary from sand (>95% by weight sand particles) through very sandy (>30% sand) muds, to the sandy (5-30% sand) muds (Stations 21, 22, 27, 28) found just off the lagoon and wharf breakwater.

CADMIUM IN SEDIMENTS AND SUSPENDED PARTICULATE MATTER

Total Cd and zinc (Zn) concentrations were determined by using flameless (Cd) and flame (Zn) A.A. techniques in a dried portion (10-500 mg) of the total sample, in a portion of the 0.053 mm fraction from each sample, and in the SPM.

Total Cd varies from 9-61 ppm in the harbour sediments (Fig. 5). The highest concentration (30-61 ppm) occurs in that part of the harbour west of a line drawn between Chapel Pte and the end of the breakwater. Seaward, Cd concentrations drop to 9-17 ppm off and to the east of the Brunswick Mining and Smelting Limited outfall. Since the highest concentrations occur along the edge of the lagoon breakwater and around to the wharf, the Cd pattern suggests an input of Cd-rich material in this area. The enrichment factor for these sediments is about 100-200 times above a background level of about 0.3 ppm found in the adjacent coastal sediments.

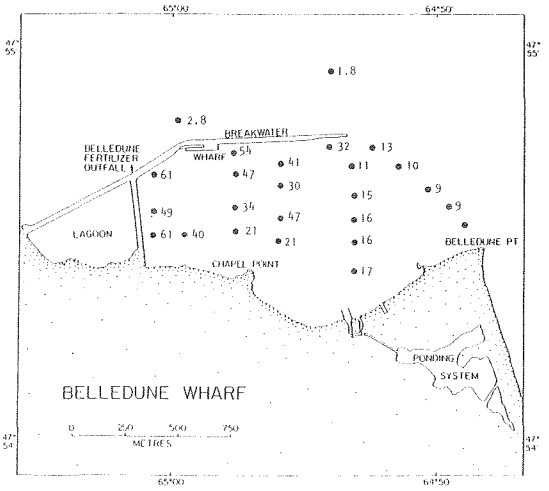

Fig. 5. Cadmium (Cd) in sediments (ppm) in Belledune Harbour.

In the mud fraction (<0.053 mm), Cd concentrations vary from 8-74 ppm (Fig. 6) and have a comparable distribution pattern to those found for the whole sediment. High Cd concentrations (28-74 ppm) occupy the western part of the harbour and decrease seaward (8-28 ppm). The enrichment factor for Cd-rich material is about 50-150 times above a background level of about 0-5 ppm we found in the offshore mud fraction.

Calculation of absolute concentrations of Cd in the sand size fraction (Fig. 7) (2.0-0.53 mm) from the total and <0.053 mm Cd data indicates this total Cd in the sand fraction varies from 4-91 ppm. The highest concentrations (88-91 ppm) of Cd occur just off the wharf and form part of a Cd-rich sand area (52-91 ppm) in the northwestern section of the harbour. Seaward over a distance of 1.5 km the Cd concentrations decrease sharply towards the harbour mouth (52-5 ppm) and follow the same dispersal

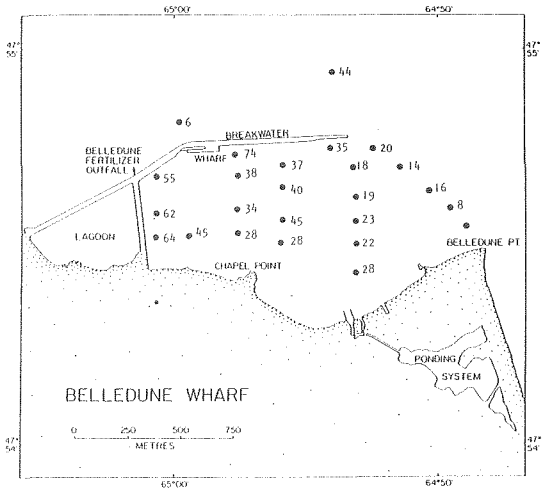

Fig. 6. Cadmium (Cd) in <53 µm fraction of sediments (ppm) in Belledune Harbour.

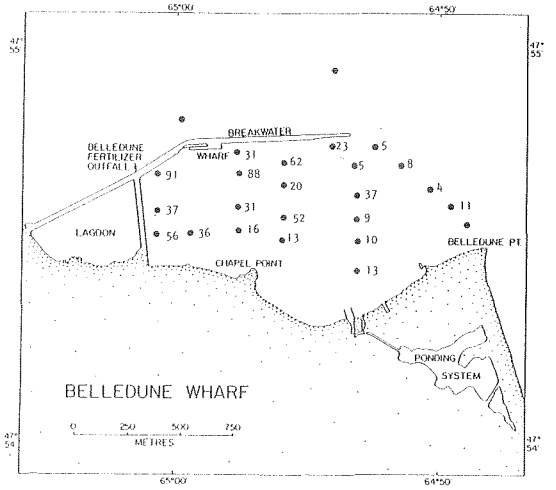

Fig. 7. Cadmium (Cd) in >53 µm fraction of sediments (ppm) in Belledune Harbour.

pattern as the Cd in the total sample and <0.053 mm fraction. The enrichment factor for the Cd-rich sand material is about 150 times above a calculation background level of about 0.05 ppm Cd in the offshore sand fraction and is the same as the enrichment factor found in the mud-size material.

Further calculations of the contributions of Cd in the sand and mud size fractions to the total Cd concentrations show that 13-56% of the total Cd in the harbour sediments occur in the sand size fraction and the remainder (44-87%) of the Cd is contributed by the mud size fraction. Regionally, the highest percentage (30-56% of the total) of Cd contributed by the sand size fraction occurs in the western part of the harbour and the lowest percentages occur seawards. The sand size particles contribute 3.0-19.6 ppm Cd to the total Cd concentration of the samples and the mud size particles contribute ~7.5-48.7 ppm. The highest concentrations of Cd-rich sand contributions (~20 ppm Cd) occur just off the lagoon breakwater and thus decrease gradually seaward with concentrations falling to 5 ppm at the mouth of the harbour. High Cd concentrations in the mud fraction (~30-45 ppm) also occur in the western part of the harbour.

Seaward Cd contribution in the mud decreases sharply from 30 ppm off Chapel Pte to <5 ppm at the mouth of the harbour.

This partition pattern and the absolute Cd concentration in the sand and mud fractions (Fig. 6, 7 respectively) imply that Cd-rich particles are bimodal and represent the input of both relatively coarse and fine Cd-rich material to the harbour. They also indicate that the Cd-rich coarse- and fine-grained material is also mostly trapped at the head of the harbour adjacent to the breakwater with little Cd-rich material being transported more than 1.5 km seaward. Although the exact source has not yet been identified, the coarse particles may reflect spillage of Cd-rich materials (see Sergeant and Ray, this report) and the deposition of the fine-grained particulate Cd, perhaps from the outfall, as suggested by the high Cd concentration (~96 ppm) of the SPM at the outfall at the southeastern corner of the harbour.

Offshore, most of the Cd (70-99%) is located in the mud size fraction and the sand fraction does not contain any significant amounts of Cd-rich sand size particles.

Five spot samples from the harbour contained approximately 6-96 ppm Cd with the highest concentration (96 ppm) occurring at the Brunswick Mining and Smelting Limited outfall (Fig. 8). Other high values (24 ppm) occurred in the center of the harbour and at the seaward end (15 ppm) of the breakwater. At the mouth of the harbour on its southward side, Cd levels are reduced to 6-7 ppm.

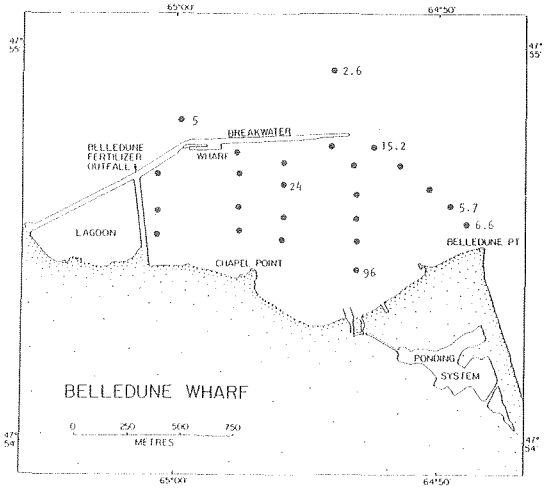

Fig. 8. Cadmium (Cd) in suspended matter (ppm) in Belledune Harbour.

In the three coastal transects to the east of the harbour (Fig. 9), total Cd varies from 0.1-1.8 ppm in the sediments. Except for sediments at station 15 about 3 km seaward of Belledune Pte and Station 16 adjacent to the west side of the breakwater that contain 1.8 ppm and 2.8 ppm Cd (Fig. 8), all the other sediments on this transect and those to the east contain low concentrations of Cd (0.1-0.5 ppm). To the west of the harbour, the sediments along the control transect at Trap Pte are also low in Cd as they contain 0.20-0.30 ppm Cd. These concentrations are considered to be at or near background values.

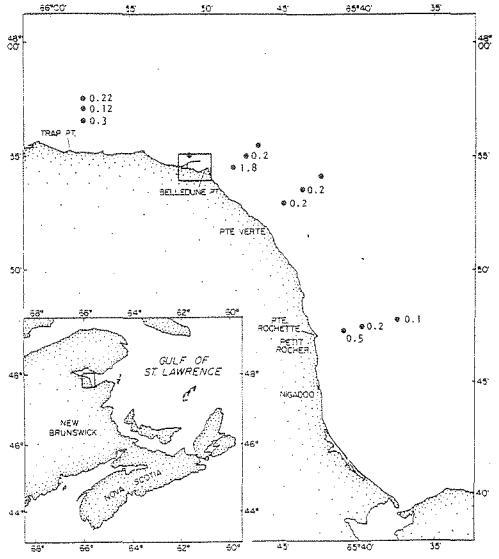

Fig. 9. Cadmium (Cd) in sediments (ppm) on coastal zone transects.

The distribution of Cd in the <0.053 mm fraction also shows the extent of the seaward migration of Cd (Fig. 10). In this fraction Cd concentration varies from 0.4-8.5 ppm Cd. The highest concentration (8.5 ppm) also occurs at station 15. About 2 km seaward of station 15, Cd concentrations decline rapidly to ~1 ppm at station 14, a level which is also found in the fine fraction about 2-3 km off Pte Verte and Rochette and Trap Pte. Beyond these stations Cd levels decline to background levels of 0.4-0.6 ppm in the fine fraction.

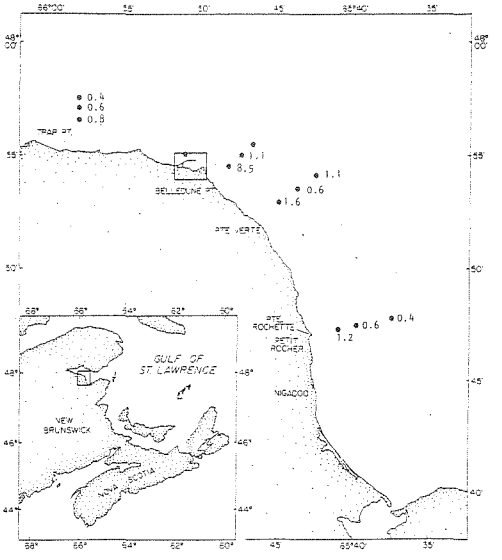

Fig. 10. Cadmium (Cd) in <53 µm fraction of sediments (ppm) on coastal zone transects.

Preliminary analysis of SPM from the coastal waters shows that they contain 2.1-6.2 ppm Cd (Fig. 11). Although these values are much higher than in most coastal sediments, they do not seem to be greatly enriched in Cd when their fine-grained nature is considered. Thus, there is little indication that the sediments, their fine fractions, and the SPM are contaminated with Cd more than 3 km seaward of the harbour mouth or eastward along the coast to Pte Rochette.

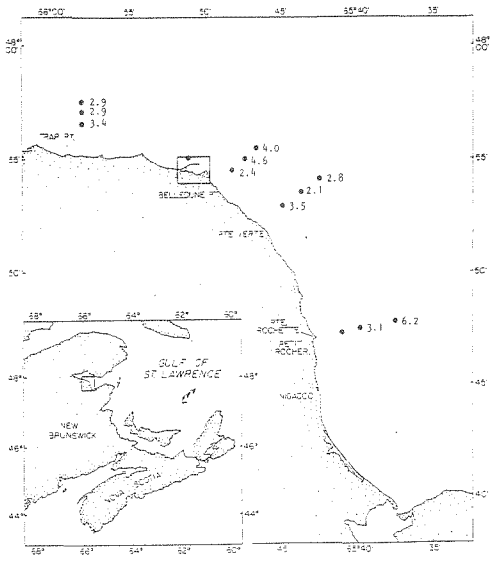

Fig. 11. Cadmium (Cd) in suspended matter (ppm) on coastal zone transects.

ZINC IN SEDIMENTS AND SUSPENDED PARTICULATE MATTER

The tentative results to date show that total Zn concentrations are also very high in the harbour sediments and their fine (<0.053 mm) fractions. Total Zn concentrations (Fig. 12) vary from 925-3800 ppm, with the highest concentrations being found, like those of Cd, in the sediments next to the breakwater in the northwest corner. The lowest Zn concentration occurs close to the Brunswick Mining and Smelting Corporation Limited outfall (Fig. 12). The fine fractions are also enriched in total Zn with concentrations varying from 700 ppm at the seaward end of the harbour to 2350-5250 ppm at the landward (western) end (Fig. 13). This pattern is thus comparable to the Cd distribution (Fig. 6).

The SPM samples contain 380-2115 ppm Zn with the highest concentration (2115 ppm), like Cd, being found at the station adjacent to the Brunswick Mining and Smelting Corporation Limited outfall (Fig. 14). Another high value is found in the center of the harbour (961 ppm) but lower concentrations (380 ppm) occur in the SPM adjacent to Belledune Pte. Thus, there is some indication that Cd-Zn rich material is being injected as particulate matter into the harbour at the Brunswick Mining and Smelting Corporation Limited outfall but becomes quickly diluted by relatively Cd-Zn poor material at the seaward end of the harbour.

Offshore, Zn contamination in the total sediment has not been determined at the present, but the Zn values in the fine (<0.053 mm) fractions (Fig. 15) vary from 117-500 ppm. The highest Zn

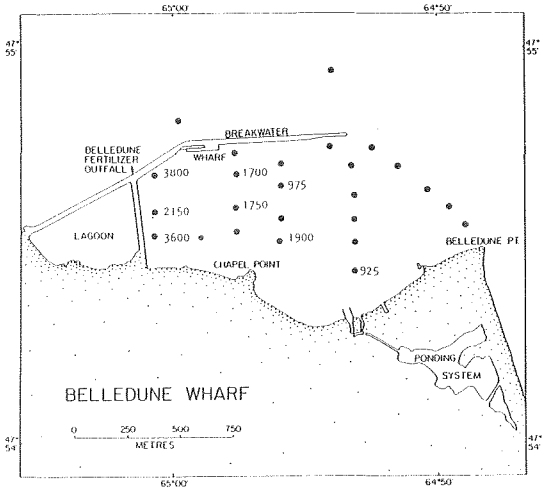

Fig. 12. Zinc (Zn) in sediment (ppm) in Belledune Harbour.

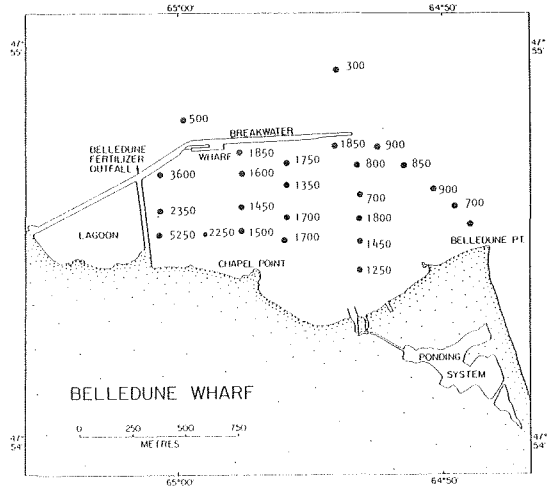

Fig. 13. Zinc (Zn) in <53 µm fraction of sediments (ppm) in Belledune Harbour.

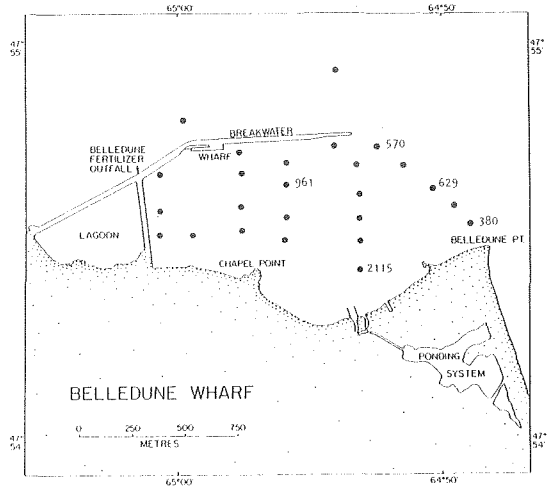

Fig. 14. Zinc (Zn) in suspended matter (ppm) in Belledune Harbour.

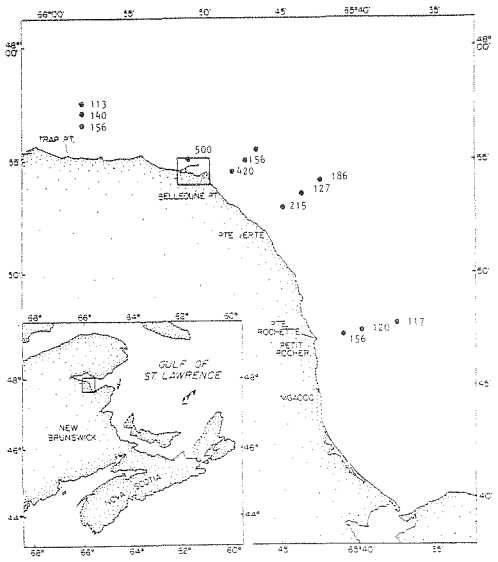

Fig. 15. Zinc (Zn) in <53 µm fraction of sediments on coastal zone transects.

concentrations (420 and 500 ppm), like those of Cd, are found at station 15 about 3 km seaward of Belledune Pte and at station 16 adjacent to the western side of the breakwater. Beyond station 15 and to the east along the coast, Zn concentrations fall quickly to ~120-180 ppm which can be considered to be near background when compared to levels of 113-156 ppm found in the fine fractions along the control transect off Trap Pte.

In the offshore SPM samples (Fig. 16) total Zn varies from 230-503 ppm without a clearcut distribution pattern. Since SPM samples from the Bay of Fundy normally contain 400-900 ppm Zn, the Zn values recorded here appear to be close to expected background concentrations for SPM.

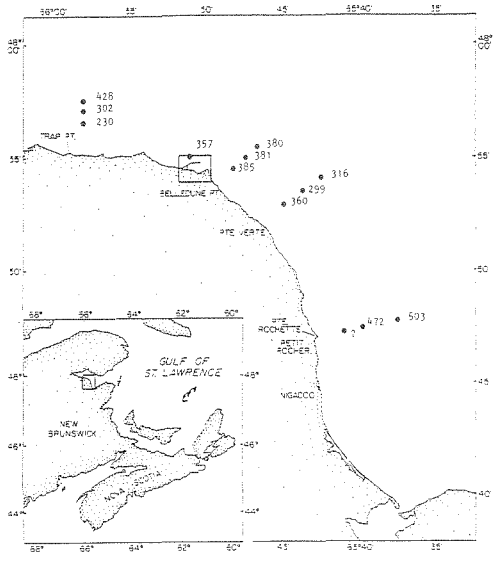

Fig. 16. Zinc (Zn) in suspended matter (ppm) on coastal zone transects.

SIZE FRACTIONATION OF CADMIUM-RICH SEDIMENT FROM BELLEDUNE HARBOUR

For the purpose of determining the Cd concentration associated with the fine-grained, most mobile, size fractions, a bottom sample from Belledune Harbour was fractionated, by settling, into different modal sizes and the grain size of each compared with the Cd content. The experimental procedure consisted of suspending similar amounts of the sediment in super-Q water and from each 3 L suspension withdrawing a pipette fraction from a constant depth after exponentially increasing time intervals. Figure 17 shows the grain-size composition of each sample corrected to a constant initial concentration. In Table 2 is listed the sediment and Cd concentration of each withdrawal sample. From the difference between consecutive samples the nature of individual graded fractions was calculated (Table 3). Size analyses of ore concentrate samples were performed for comparison (Fig. 18).

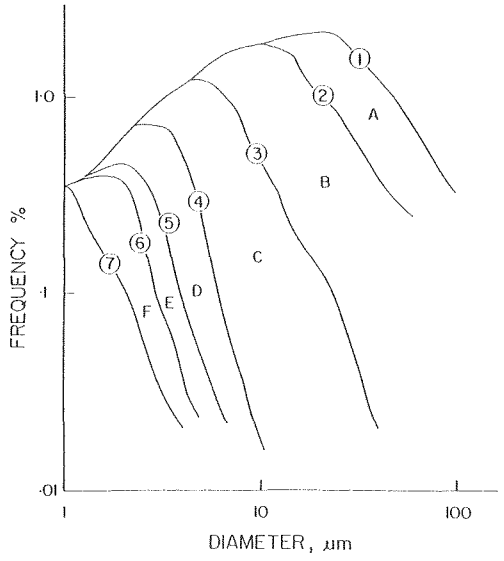

Fig. 17. Size fractionation of Cd-rich sediment. For definition of individual samples and fractions see Tables 1 and 2.

Results indicate that during the first five draw-offs, concentration, grain size and Cd content decreased in a regular, consistent manner; 94.33% of the Cd present in the first draw-off had settled out by the third draw-off, indicating that most of the Cd was associated with particles of 12-20 µm modal diameter. The fraction corresponding to modal sizes of 8 and 5 µm contained only 2.4 and 2.3% Cd, respectively. In the last two draw-off samples the Cd values were higher than found in the last sample and consequently no Cd values could be calculated for the 3.0 and 1.5 µm fractions. This increase is most likely due to flocculation or absorption of colloidal or dissolved Cd onto larger particles. The loss of Cd in this form to the super-Q water is shown by a dissolved Cd content of 0.45 ppb measured in the filtered water of the last draw-off. The one order of magnitude variation in the Cd to surface area ratio is evidence that the Cd does not occur as a coating on larger sediment grains but as discrete Cd-rich particles.

Table 2. Size fractionation of Cd-rich sediment from Belledune Harbour.

Fraction[a]	Settling rate (cm/s)	Correction factor	Total conc. (mg/L)	Cd ppm (wt)	Surface area (μm^2/L)	Cd (mg/L)
1	>4.45E-2	1.45	70.79	70	2.80E10	1.04E-2
2	>1.11E-2	1.1	63.88	54	1.94E10	4.05E-3
3	>6.94E-3	2.1	15.43	38	1.55E10	5.86E-4
4	>1.74E-4	2.1	7.71	43	7.10E9	3.31E-4
5	>8.68E-5	1.8	4.94	39	4.24E9	1.88E-4
6	>4.21E-5	1.6	4.80	91	2.67E9	4.36E-4
7	>2.10E-5	1.0	4.30	58	1.62E9	3.88E-4

[a]See Fig. 1.

Table 3. Size fractionation of Cd-rich sediment from Belledune Harbour.

Fraction[a]		Mode size	Cd mg/L	σ/σ total	Surface area μm^2/L	Cd surface mg/cm^2
A	1.11E-2 - 4.45E-2	20	6.35E-3	61.06	0.86E10	7.33E-1
B	6.94E-4 - 1.11E-2	12	3.46E-3	33.27	0.39E10	8.87E-1
C	1.74E-4 - 6.94E-4	8	2.54E-4	2.44	0.84E9	3.02E-1
D	8.68E-5 - 1.74E-4	5	1.43E-4	1.38	2.85E9	5.01E-2
E	4.21E-5 - 8.68E-5	3	-	-	1.56E9	-
F	2.10E-5 - 4.20E-5	1.5	-	-	1.05E9	-
				98.15		

[a]See Fig. 1.

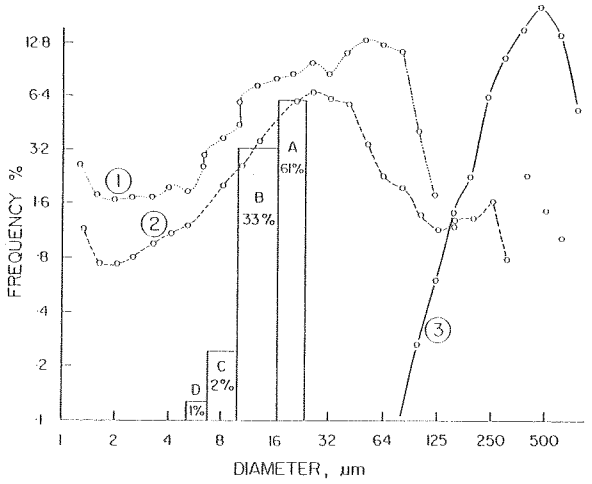

Fig. 18. Grain size analysis of ore concentrate samples from Belledune area. Samples marked: 1. High Cd dust from stock pile; 2. Lead concentrate from Church (RMS); 3. Blast-furnace slag from stock pile. Also shown are relative concentrations of Cd in individual size fractions of fractionated bottom sample.

The ore concentrate samples are dominated by grains larger than 5-8 µm. Two out of three of the samples also contain finer particles. Optical examination showed that the sample labeled "high Cd-rich dust from stock pile" was rich in very fine opaque material similar in appearance to grains seen in the Belledune samples.

In conclusion, Cd in the Belledune sediment samples is present mainly in 5-8 µm and larger grains but fine, partly sub-micron material causes high relative concentrations in the finer fractions which would dominate the suspended matter. Cadmium in the latter may be expected to shift between the dissolved and particulate fraction during normal resuspension and settling action of the sediment.

WATER SAMPLING AND ANALYSIS

Water samples were collected at a depth of 7 m at selected stations by using a General Oceanics 12-L GO-FLO sampling device. Subsamples of the water were collected for salinity and trace metal determinations. The trace metal samples were filtered through preweighed 0.4 µm pore-size Nuclepore filters to remove and collect suspended particulate matter. The filtrate was then acidified with HCl to pH $\leq$2 and stored in 2-L conventional polyethylene bottles (Bewers et al. 1976).

In the laboratory, filters were washed, dried and reweighed to determine the concentration of suspended particulate matter (SPM) per unit volume of water. Aliquots of the filtered water samples were analyzed for Cd, Cu, and Zn by graphite furnace-atomic absorption spectrophotometry following preconcentration by chelation and solvent extraction (Bewers et al. 1976). Separate aliquots of these samples were also analyzed by anodic stripping voltametry. The concentrations of SPM, Cu, Zn, and Cd at each station are presented in Table 4. Inside the harbour, Cd and Zn concentrations were too high for the preconcentration/extraction procedures chosen for this study. As a result, very low extraction efficiencies led to low results for the analysis of samples 803320 and 803321 by atomic absorption. Anodic stripping voltametric analysis gave the much higher results for these two samples that are quoted in Table 4. Otherwise, the ASB and AAS methods yielded comparable results.

RESULTS

The concentrations of Cu, Zn, and Cd in water are shown in Fig. 19-24. These concentrations are best appreciated in the context of the ranges of each metal in typical uncontaminated coastal seawater. These are as follows:

Cu	0.3-0.5 µg/L	(Yeats et al. 1978);
Zn	0.8-2.0 µg/L	(Bewers 1979);
Cd	0.05-0.10 µg/L	(Bewers and Yeats 1979).

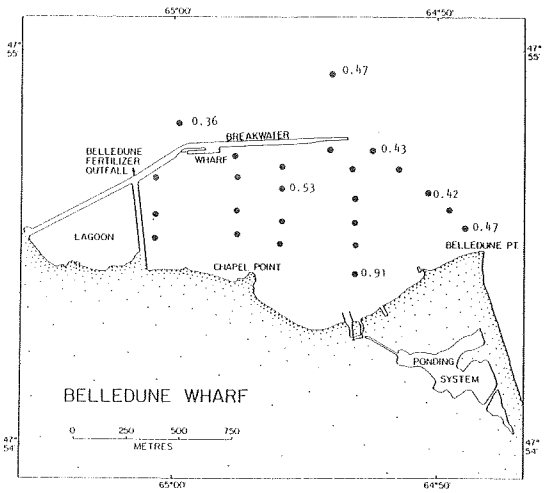

Fig. 19. Copper (Cu) in water (µg/L) in Belledune Harbour.

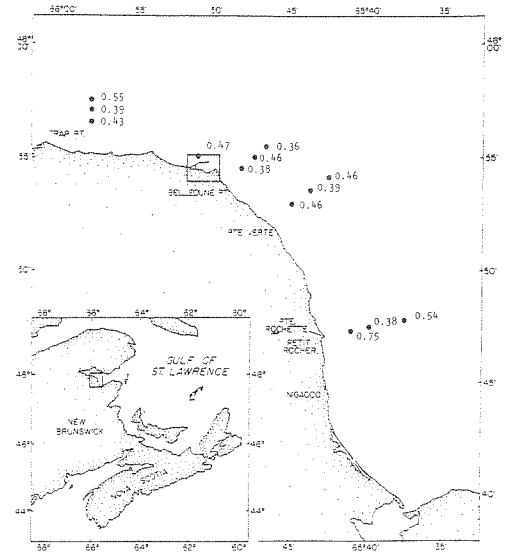

Fig. 20. Copper (Cu) in water (µg/L) in coastal zone transects.

Table 4. Concentration of copper (Cu), zinc (Zn), and cadmium (Cd) in seawater. Data in parentheses are based upon anodic stripping voltametry. All other analyses were conducted by atomic absorption spectrometry following chelation/solvent extraction procedures.

Station no.	Salinity o/oo	Suspended particulate matter mg/L	Cu conc. µg/L (SD)	Zn conc. µg/L (SD)	Cd conc. µg/L (SD)
8033 01	21.61	1.34	0.472(0.007)	6.29(1.08)	2.26 (0.19)
03	23.48	1.49	0.481(0.032)	2.79(0.32)	0.506(0.021)
05	23.69	1.95	0.433(0.062)	1.68(0.15)	0.173(0.006)
06	23.70	1.14	0.471(0.066)	5.40(0.53)	2.50 (0.25)
07	27.34	1.42	0.540(0.024)	2.41(0.22)	0.125(0.016)
08	24.24	1.66	0.384(0.031)	1.91(0.34)	0.196(0.016)
09	24.33	-	0.751(0.053)	1.93(0.18)	0.198(0.018)
10	27.61	1.38	0.460(0.011)	2.37(0.15)	0.176(0.020)
11	27.08	1.67	0.387(0.010)	2.10(0.09)	0.161(0.006)
12	25.89	1.44	0.460(0.010)	2.65(0.26)	0.190(0.006)
13	27.66	1.32	0.356(0.018)	2.62(0.07)	0.185(0.027)
14	27.68	1.93	0.464(0.024)	1.92(0.05)	0.108(0.004)
15	27.02	2.45	0.384(0.023)	2.26(0.18)	0.182(0.008)
16	29.98	1.51	0.363(0.017)	3.19(0.12)	0.458(0.032)
17	27.93	1.87	0.553(0.018)		0.168(0.012)
18	28.20	1.24	0.393(0.013)	2.68(0.02)	0.235(0.018)
19	28.05	2.00	0.426(0.005)	2.58(0.10)	0.230(0.010)
20	24.29	7.84	0.907(0.019)	260	125(23)
21	25.10	-	0.531(0.005)	34.3(2.7)	25.5(2.1)

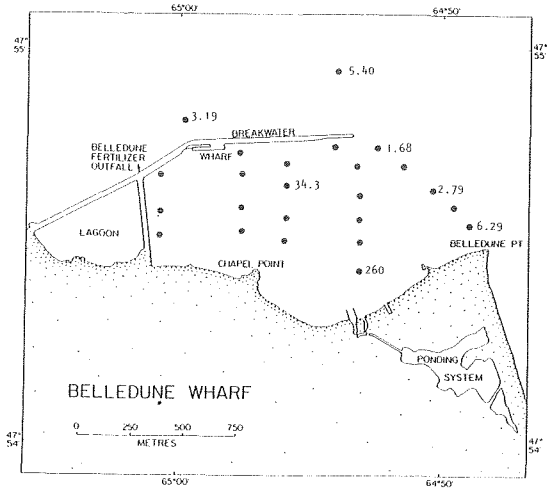

Fig. 21. Zinc (Zn) in water (µg/L) in Belledune Harbour.

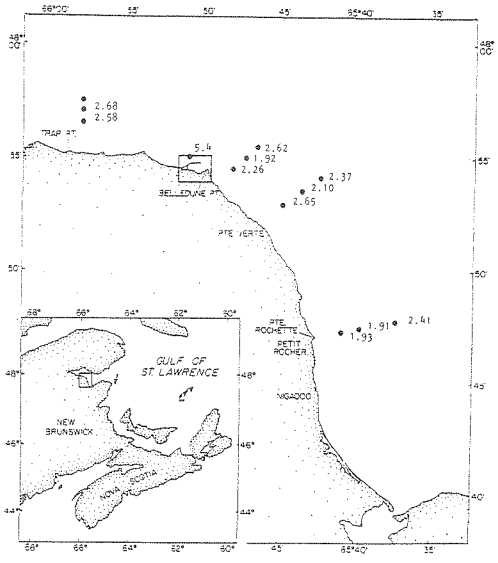

Fig. 22. Zinc (Zn) in seawater (µg/L) in coastal zone transects.

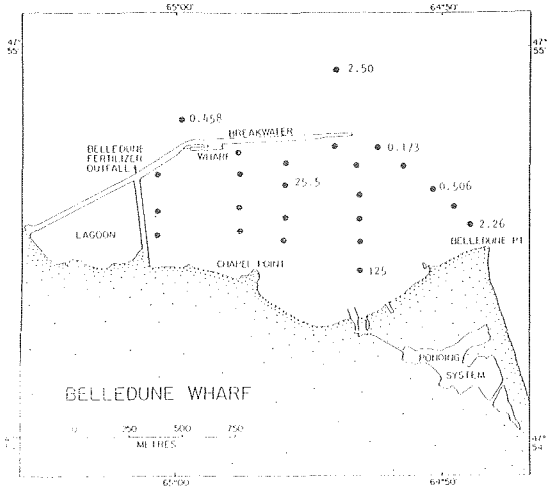

Fig. 23. Cadmium (Cd) in seawater (µg/L) in Belledune Harbour.

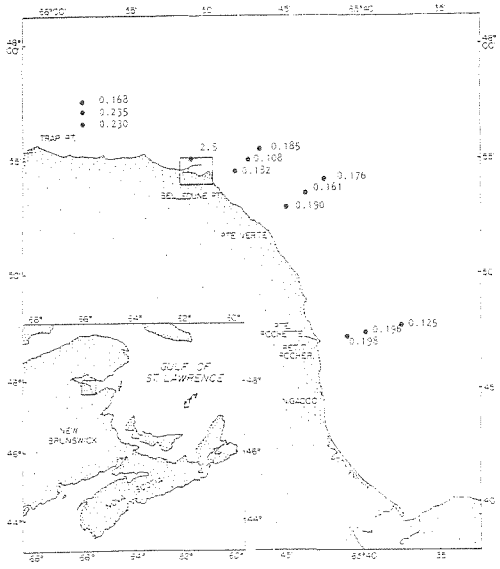

Fig. 24. Cadmium (Cd) in seawater (µg/L) in coastal zone transects.

As can be seen in Fig. 19-20, copper concentrations in the vicinity of Belledune, even inside the harbour, are comparable with typical coastal water concentrations. The copper concentration only exceeds the coastal seawater levels on the station adjacent to the outfall of the smelting plant where the concentration approaches 1 µg/L.

In contrast, both Zn (Fig. 21-22) and Cd (Fig. 23-24) show considerably increased levels in the vicinity of Belledune Harbour. The Zn concentration of the sample collected adjacent to the outfall (260 µg/L) was ~150 times the normal levels in coastal waters but the concentrations rapidly decrease to 1.7-6.3 µg/L in the section across the harbour mouth. Outside the harbour high concentrations were found only at stations 6 and 16 along the outside of the breakwater. These elevated concentrations may have arisen from releases from the outfall of the fertilizer plant or transport of water from Belledune Harbour through or around the end of the breakwater. All Zn concentrations on the four nearshore transects in the Bay of Chaleur were close to expected coastal water values. Thus, it appears that the major Zn contamination of the water is confined to Belledune Harbour with somewhat elevated levels also evident in the area adjacent to the harbour breakwater.

The Cd distribution is very similar to that of Zn. Levels in the general area are somewhat higher than typical coastal water samples but this may be the consequence of mineralization of the local land area. Higher values are confined to the vicinity of Belledune Harbour where concentrations can be 100 times ambient. As in the case of Zn, the samples obtained just outside the northern breakwater are elevated in Cd, but not to the extent found inside Belledune Harbour.

CONCLUSIONS

Preliminary conclusions that may be drawn from the data contained in this paper are that: 1) Belledune Harbour is heavily contaminated with Cd and Zn in both aqueous and sedimentary phases, and 2) contamination levels decrease rapidly away from the harbour such that none of the coastal zone transects further than 3 km from Belledune Harbour can be regarded as yielding abnormal metal distributions. Within the harbour, Cd-rich particles are found to have a bimodal distribution which suggests that both fine and coarse particles are important agents of Cd transport. Most of these Cd-rich particles remain trapped at the western end of the harbour, close to the breakwater, and little material seems to be transported more than 1.5 km seaward. It is possible that any exported metals have been transported predominantly into the deeper regions of Chaleur Bay to the northeast. Nevertheless, it is our conviction that, under normal atmospheric conditions, residual coastal circulation would be the most prominent mechanism of particle and aqueous transport. It is likely that storm events could produce considerable redistribution of the contaminated sediments and this aspect of transport would be worthy of further investigation.

ACKNOWLEDGMENTS

We wish to thank others involved in the OAS activities reported here. The field party included John Butters, John Dalziel, Dennis Pottie and Peter McGinn. Without their perserverance under difficult weather conditions, the samples could not have been collected. The samples were subsequently analyzed by Ray Rantala, Byron Amirault and John Dalziel. Their efforts and the valuable advice and assistance of Dr. Phil Yeats are greatly appreciated.

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THE LEACHING OF CADMIUM FROM THREE BELLEDUNE SMELTER MATERIALS

by

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INTRODUCTION

Emissions of cadmium (Cd) associated with Brunswick Smelting's operation at Belledune were described by Sergeant and Westlake (this report). The levels of Cd found in water, sediment and biota from this area may be related to the leaching of Cd from various materials used in, or produced by, the smelting process. Of particular concern with respect to leaching are: the slag because of its quantity (at present, 1.4 million metric tons (MT) in the slag pile); the baghouse dust which is removed from the process when the Cd concentration reaches about 10% (545 MT last year), because of its high concentration of Cd and the Cottrell dust received from Noranda Horne mine, because of its volume and 1% Cd content (4542 MT for each of 1978 and 1979). These materials were subjected to leaching tests to determine their relative leaching potential in a rapid preliminary study covering a short time period and using arbitrary, non-standard techniques. Leaching criteria in relation to smelting and refining technologies are discussed, for example, by Leonard et al. (1977).

METHODOLOGY

Polyethylene bottles (1 L, soaked overnight with 50% nitric acid and then washed three times with deionized distilled water) were filled with: a) distilled water at pH 1; b) normal seawater; c) seawater at pH 1 (pH was adjusted by addition of nitric acid, BDH Aristar grade). The initial volume in each bottle was 900 mL. Approximately 0.9 g (wet weight) of each sample was added.

The bottles were thoroughly shaken periodically and the contents were allowed to settle for 16 h prior to sampling. No haziness was observed at the time of sampling. A 20-mL water sample was withdrawn by pipette without filtering from each bottle at 0, 17, 41, 94, 166, 304, 376, 475 and 523 h. The unacidified seawater samples were acidified with two drops of Aristar nitric acid at the time of sampling. Cd concentrations in the unfiltered water samples were determined by graphite furnace atomic absorption spectroscopy. Accuracy and precision of Cd analyses were checked against NBS standard reference material 1643a (trace elements in water).

The results (µg Cd leached/g material) of the leaching tests are presented in Fig. 1-3. Plotted lines were fitted to the data by least square regressions. Analytical (interference) problems encountered with the baghouse dust in distilled water for the 17-h sample led to sampling and analysis of water samples from this bottle being terminated after that time.

Samples of slag, baghouse dust and Cottrell dust were analyzed by the New Brunswick Research and Productivity Council (RPC) (Fredericton, N.B.). Baghouse dust was also analyzed by the National Research Council (NRC) laboratories (Ottawa, Ont.) (Table 1, 2).

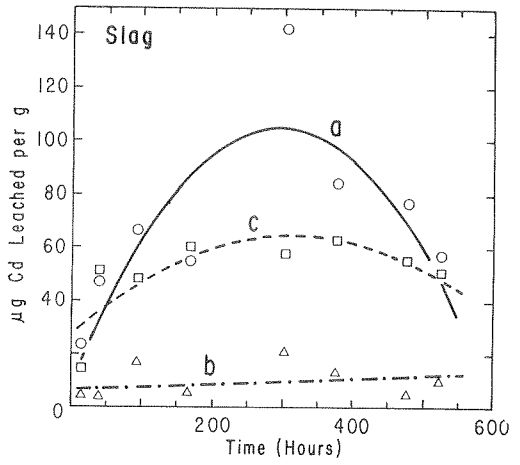

Fig. 1. Graph of µg Cd leached/g slag in different media.

○ Slag in acidified distilled water

△ Slag in normal seawater

□ Slag in acidified seawater

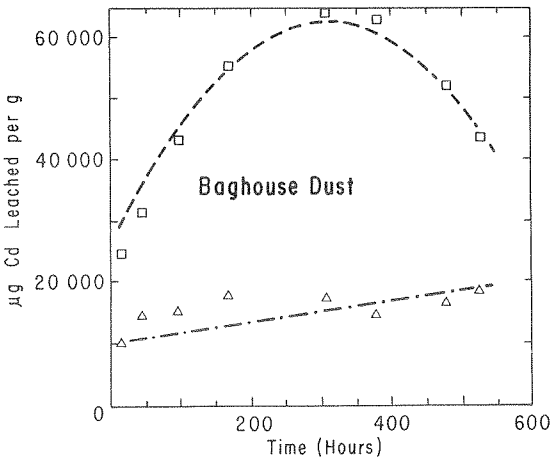

Fig. 2. Graph of µg Cd leached/g baghouse dust in different media.

△ Baghouse dust in normal seawater

□ Baghouse dust in acidified seawater

Table 1. Analysis of baghouse dust (NRC, Ottawa).

Element	Wet weight %	Element	Wet weight %
Fe (iron)	20	Sn (tin)	0.1
Pb (lead)	10	In (indium)	0.1
Zn (zinc)	10	Bi (bismuth)	0.1
Ca (calcium)	10	Sb (antimony)	0.06
As (arsenic)	5	Ag (silver)	0.03
Si (silicon)	3	Ti (titanium)	0.03
Al (aluminum)	0.3	Mn (manganese)	0.03
Cu (copper)	0.3	B (boron)	0.01
Mg (magnesium)	0.1	Cd[a] (cadmium)	7.5

[a]Flame atomic absorption.

Table 2. Analyses of slag, baghouse dust and Cottrell dust (RPC, Fredericton).

Element	Wet weight in slag %	Wet weight in baghouse dust %	Wet weight in Cottrell dust %
Fe (iron)	30.6	7.6	19.2
Zn (zinc)	12.8	5.3	4.6
Cu (copper)	0.30	0.61	0.33
Pb (lead)	4.06	32.6	30.3
Cd (cadmium)	0.01	9.7	0.01[a]
Bi (bismuth)	0.01	0.11	0.04
Sb (antimony)	0.03	0.06	0.08
Ca (calcium)	10.4	2.32	0.06
As (arsenic)	0.12	1.6	0.32
Ag (silver)	34 ppm	219 ppm	467 ppm

[a]Problems with precipitation of material during dissolution process.

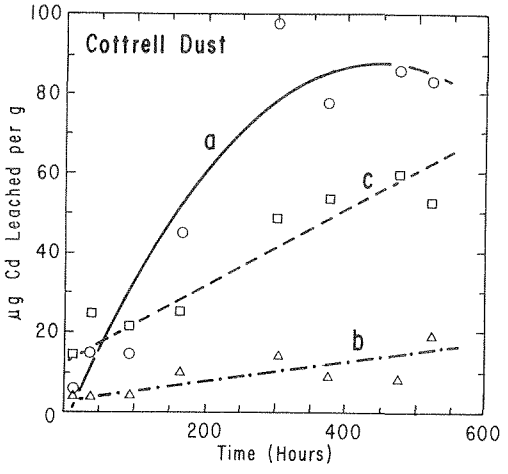

Fig. 3. Graph of µg Cd leached/g Cottrell dust in different media.

○ Cottrell dust in acidified distilled water

△ Cottrell dust in normal seawater

□ Cottrell dust in acidified seawater

RESULTS AND DISCUSSION

LEACHING STUDY

Slag, baghouse dust and Cottrell dust are potentially subject to leaching of Cd by contact with water. Of particular concern is the baghouse dust which contains high levels of Cd (approximately 10%). Rozovsky (1974) reported that the Cd in the dust from the electrostatic precipitator on the sintering machine was in the oxide form, and thus presumably readily leachable.

Acidified distilled and seawater were chosen as leaching solvents to give an estimate of the maximum leaching potential of each material, compared to the leaching potential in seawater.

Physical characteristics

The slag sample was a slightly wet, black, coarse, sand-like, glassy material; the baghouse dust from the stockpile was wet, greyish-green and very fine with some agglomerations; the Cottrell dust was a dry, very fine, dark-grey powder and

again had some agglomerations. These characteristics explain why fine material from the baghouse dust and Cottrell dust was observed floating on the surface in the acidified distilled water bottles. Fewer floating particles were observed in the seawater tests with these materials. In both acidified and normal seawater the baghouse dust became a silvery color.

Although settling rates were not determined, the tendency to settle was slag > baghouse dust > Cottrell dust. At the end of the experiment, fines of Cottrell dust were still visible on the surface of all three media, whereas a small amount of baghouse dust fines was observed on the surface of the seawater only.

Slag

A gradual leaching of Cd by acidified distilled water over the first 150 h was observed (Fig. 1a). An equilibrium was established after that time. There was some fluctuation in the data and the last three sample points may indicate some adsorption of Cd, or could be a reflection of sampling (drawing of fine particles into pipette) or analytical variations. Leaching of Cd from slag in normal seawater was very slow (Fig. 1b). There is considerable fluctuation in the data points, and at the end of the test the Cd level was only 15% of the maximum observed for the acidified distilled water case. The leaching in acidified seawater (Fig. 1c) shows a rapid leaching of Cd with equilibrium being established after about 50 h. The amounts of Cd leached by acidified seawater were about 50% of that leached by acidified distilled water.

Slag is the major byproduct of the smelting operation and even a limited amount of leaching by seawater could contribute large amounts of Cd to the environment. Assuming the slag contains 0.01% Cd, then 100, 20 and 63% of the Cd was leached from the slag by acidified distilled water, seawater and acidified seawater.

Baghouse dust

Rapid leaching by acidified seawater followed by equilibrium was observed (Fig. 2). The dust in normal seawater leached rapidly at first, then gradually through the remainder of the experiment. In both media the quantities of Cd leached were 10^3 times the levels observed for slag. Acidified distilled water leached about the same amount of Cd as acidified seawater at 17 h when this test was terminated. The problem encountered was that of a split peak on the recorder and failure of the pen to return to baseline indicating a probable interference phenomenon. Since the dust was determined to contain 8.6% wet weight Cd (average of Cd values in Tables 1 and 2) on a wet weight basis, then normal seawater had leached about 21% of the total Cd from the dust and the acidified seawater about 67%.

The quantity of dust produced and its handling and storage are therefore of concern due to its high Cd content. If this dust is conveyed by wind or as a suspension in water into the harbour or Bay of Chaleur, it would present a major source of Cd contamination of the environment in excess of the normal dustfall (Sergeant and Westlake, Fig. 1, this report). This dust, if stockpiled in the open, can also be leached by precipitation.

Cottrell dust

Leaching in acidified distilled water proceeded slowly with equilibrium being reached after about 450 h (Fig. 3a). The amount leached was about the same as from the slag. For normal seawater leaching, a constant leaching rate was observed (Fig. 3b). In comparison with the acidified distilled water, less than one quarter of the amount of Cd was leached in normal seawater. The scatter of the points in the acidified seawater leaching test (Fig. 3c) made it impossible to say whether equilibrium was reached. It appears that leaching continued over the entire test period with the quantity leached being slightly less than that in the acidified distilled water. Assuming that this dust is 1% Cd wet weight, we find that 0.9, 0.1 and 0.6% of the total Cd in this material was leached by acidified distilled water, normal seawater and acidified seawater, respectively.

The Cottrell dust should not present a problem under normal handling conditions. However, its potential to leach should not be overlooked. Cottrell dust has demonstrable leachability and should be handled accordingly.

ANALYSIS OF BELLEDUNE SLAG, BAGHOUSE DUST AND COTTRELL DUST

The National Research Council analyzed the baghouse dust by using a dc-arc emission spectrographic method. The results of their analyses are given in Table 1. The New Brunswick Research and Productivity Council analyzed the slag, baghouse dust and Cottrell dust by X-ray fluorescence and X-ray diffraction to establish what elements were present and then quantified the elements by atomic absorption spectrophotometry. Their analyses are given in Table 2. The NRC results in Table 1 are semiquantitative and should be within a factor of 3 of the actual results. Comparing Table 1 to Table 2 for the baghouse dust, we see this type of agreement.

To see if our samples (Table 2) were representative of typical materials, we obtained some Brunswick Smelting data on the typical composition of such materials at the Belledune smelter. Except for the Cd analysis on the Cottrell dust, there was good agreement in composition.

CONCLUSIONS

The leaching potential decreases in the order baghouse dust $\ggg$ slag $\Rightarrow$ Cottrell dust. The baghouse dust is the most hazardous material from leaching and environmental points of view. The slag, despite its low Cd content and relatively limited leachability, may also be a source of Cd because of its quantity.

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UPTAKE AND EXCRETION OF CADMIUM BY MARINE ORGANISMS FROM SEA WATER WITH CADMIUM AT LOW CONCENTRATION: A REVIEW

by

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INTRODUCTION

The objectives of this review are:

(A) to assess cadmium (Cd) accumulation by marine organisms when they are exposed to sea water with Cd at low concentration (ppb range),

(B) to assess the potential for Cd excretion by marine organisms.

A. ACCUMULATION OF CADMIUM

Summary

From the data for sea weeds, crustaceans, polychaete worms and molluscs, there is evidence that Cd accumulates in the tissues of these organisms from water containing Cd at very low concentrations, from trace amounts to less than 0.5 ppb Cd in water. For animals exposed to 0.5 ppb Cd or less, uptake rates are low, ranging from 0.05 to 15.3 ppb Cd/h (dry wt).

Sea weed

Fucus vesiculosus taken from water with 0.3 ppb Cd had tissue concentration of 15 ppm Cd dry wt, and from water with 0.5 ppb Cd had tissue concentration of 30 ppm Cd (Butterworth et al. 1974) (Table 1), yielding concentration factors (CF's) of 50,000 and 60,000, respectively.

F. vesiculosus taken from water with mean annual Cd concentration of 0.27 ppb had tissue concentration of 3.8 ppm Cd dry wt, and from water with 0.59 ppb Cd, the tissue concentration was 11.3 ppm Cd (Morris and Bale 1975) (Table 2), yielding CF's of up to 25,500 and up to 51,400, respectively, close to the values reported above.

Crustaceans

Lobsters (Homarus americanus) exposed to sea water with 0, 3 or 6 ppb (added) Cd for 30 d showed no change in Cd concentration in hepatopancreas, 20.3-23.5 ppm wet wt, or in muscle tissue (<0.12 ppm wet wt, detection limit). An elevated Cd concentration occurred in the gill tissue with exposure to 6 ppb Cd but not to 3 ppb Cd (Thurberg et al. 1977) (Table 3).

Lobsters exposed to 10 ppb Cd for 10 d showed no change in Cd concentration in viscera (presumably, hepatopancreas and gonad tissues) or muscle tissue but Cd concentrations in gills and shell were elevated (Eisler et al. 1972) (Table 4). The differences in concentration between control and test animals yield CF's of 33 and 38 for shell and gills, respectively.

Brown shrimp (Crangon crangon) exposed to 1.5 ppb Cd in water for 20 d showed no uptake of Cd, the tissue concentration remaining constant at 0.55 ppm Cd dry wt. Exposure to 2.5 ppb Cd resulted in a higher concentration, 1.1 ppm Cd, in the tissues (Dethlefsen 1977/78) (Table 5). The difference (1.1-0.55) yields a CF of 220.

Crangon septemspinosa showed no accumulation of Cd when exposed for 30 d to a trace amount (<0.01 ppb) of Cd (Ray et al. 1980b).

Polychaetes

Nereis virens exposed for 30 d to a trace amount (<0.01 ppb) of Cd showed an increase in Cd concentration from 0.37 to 0.58 ppm Cd dry wt (Ray et al. 1980b).

Nereis diversicolor from two estuaries with different background levels of Cd in the sediment

Table 1. From Butterworth et al. (1974) - Fucus and molluscs from nature.

Cd in water ppb	Cd in tissues of various species (ppm dry wt)							
	Fucus	CF	Littorina	CF	Patella	CF	Thais	CF
5.8	220	38'[a]	-	-	550	95'	-	-
3.8	200	53'	210	55'	220	58'	-	-
2.0	50	25'	140	70'	200	100'	425	212'
1.3	25	19'	30	23'	110	85'	330	254'
1.0	20	20'	25	25'	50	50'	270	270'
1.2	44	37'	40	33'	70	58'	120	100'
0.5	30	60'	16	32'	30	60'	65	130'
0.3	15	50'	15	50'	30	100'	62	207'
0.1 (open sea)	-	-	-	-	-	-	-	-

[a] 38' = 38,000, etc.

Table 2. From Morris and Bale (1975) - Fucus from nature.

Annual average Cd in water ppb	Range of Cd in water	Conc. Cd in Fucus ppm dry wt	Average CF
1.34	0.91-2.02	19.5	14.5'[a]
1.07	0.65-1.50	15.8	14.8'
0.75	0.45-1.28	19.2	25.6'
0.59	0.29-1.20	9.3	15.8'
0.59	0.22-1.77	11.3	19.2'
0.27	0.15-0.38	3.8	14.2'

[a] 14.5' = 14,500, etc.

Table 3. From Thurberg et al. (1977) - lobsters exposed to 0, 3 and 6 ppb Cd for 30 d.

Cd in water ppb	Cd in various tissues (ppm wet wt)			CF
	Hepatopancreas	Gills	Muscle	Gills
0	20.3	1.5	<0.12	-
3	22.5	1.8	<0.12	100
6	23.5	3.4	<0.12	465

Table 4. From Eisler et al. (1972) - lobsters exposed to 10 ppb Cd for 10 d.

Tissue	Control Cd ppm wet wt	Test Cd ppm wet wt	CF
Whole lobster	0.51	0.72	21
Muscle	0.20	0.25	5
Shell	0.59	0.88	33
Gill	0.49	0.87	38
Viscera	1.21	1.21	0

Table 5. From Dethlefsen (1977/78) - Brown shrimp (Crangon crangon).

Cd in water ppb static tests	Cd in shrimp ppm dry wt (30 d)	CF (30 d)	Cd in shrimp ppm dry wt (40 d)	CF (40 d)
5	3.8	760	-	-
10	4	500	-	-
20	7.3	365	-	-
50	8.3	165	14.7	295
100	16.1	160	22.0	220
Flow through tests				
1.5	constant level at 0.55 ppm during 30 d			
2.5	1.1 ppm at 30 d - CF 220			

were exposed to 1 ppb Cd in water for 38 d (Bryan and Hummerstone 1973) (Table 6). Worms from the Avon area, with the lower background level, accumulated Cd to 208 ppm (CF about 208,000) and those from Restronguet Creek accumulated 140 ppm Cd dry wt (CF about 140,000). Cadmium was absorbed at a slower rate by the worms from the area with the higher background level of Cd.

Molluscs

Mytilus edulis in water with <0.5 ppb Cd showed an increase in the Cd level in tissues from 0.3 (control) to 10 ppm dry wt (CF of 20,000). Those exposed to 4.6 ppb Cd showed an increase to 70 ppm (CF of 15,200) (Westernhagen et al. 1978).

Oysters (Crassostrea virginica) accumulated Cd from water with 0.1 (CF about 1,000,000) and 0.2 ppb Cd (CF about 500,000) (Shuster and Pringle 1969) (Table 7).

C. virginica from water with 0.1 to 0.2 ppb Cd had tissue level of 2.7 ppm Cd wet wt (11.8 ppm Cd dry wt). Exposure to 5.0 ppb Cd for 40 wk resulted in elevated Cd concentrations in the tissues, 13.6 ppm wet (105 ppm Cd dry, CF of 18,600) (Zaroogian and Cheer 1976) (Table 8).

Littorina and Patella accumulated Cd from water with 1 ppb Cd, Thais from water with 0.5 ppb Cd. For all three species it is possible that there was accumulation from 0.3 ppb Cd (Butterworth et al. 1974) (Table 1). The CF's from exposure to 0.3 ppb Cd are about 50,000, 100,000 and 207,000 for Littorina, Patella and Thais, respectively.

Macoma controls had 0.24 ppm Cd dry wt; those exposed to a trace (<0.01 ppb) amount of Cd for 30 d had 0.6 ppm Cd (Ray et al. 1980b). Macoma exposed for 15 d to sea water with 1 ppb Cd showed an increase in CF from <190, initially, to about 370 at end of exposure (McLeese et al., unpubl.).

Calculated and extrapolated uptake rates for Cd

Some of the reviewed data were for organisms taken in nature, presumably at long-term equilibrium, and uptake rates could not be calculated. For the remaining data, organisms had been exposed to water with 6 ppb Cd or less and uptake rates were calculated (Table 9) according to the following formula:

$$\text{Rate of uptake} = \frac{Cd_A - Cd_{AC}}{\text{Exposure time in h}}$$

Table 6. From Bryan and Hummerstone (1973) - Nereis diversicolor from two areas exposed to Cd in water for up to 816 h (35 d).

Cd in water ppb	Worms from Avon ppm Cd dry wt	CF	Worms from Rest. Creek ppm Cd dry wt	CF
1.0	208	208'[a]	140	140'
2.5	471	188'	225	90'
10	1860	186'	629	63'
25	1970	79'	1630	65'
100	4010	40'	1300	13'

[a]208' = 208,000, etc.

Table 7. From Shuster and Pringle (1969) - oysters exposed to Cd for 16 and 20 wk.

Cd in water ppb	Cd in oysters ppm wet wt	CF
Control	1.17 and 2.64	- -
0.1 (16 wk)	122.5 and 105.7	1,225'[a] and 1,060'
0.2 (13 wk)	94.5 and 125.8	473' and 629'

[a]1,225' = 1,225,000, etc.

Table 8. From Zaroogian and Cheer (1976) - oysters exposed to 0.1-0.2 (controls) and 0.5 Cd for 40 wk.

Cd in water ppb	Cadmium in oysters ppm wet wt	Cadmium in oysters ppm dry wt	CF wet	CF dry
0.1-0.2	2.72	11.83	-	-
5.0	13.57	104.95	2.2'[a]	18.6'

[a] 2.2' = 2,200, etc.

Table 9. Calculated uptake rates of cadmium (Cd) for marine invertebrates exposed to Cd in water at concentrations of 6 ppb or less.

Animal	Reference	Exposure time (d)	Exposure conc. (ppb)	Calculated uptake rate (ppb/h)
Polychaetes				
Nereis virens	Ray et al. (1980b)	30	trace	0.3 (dry)
Crustaceans				
Crangon septemspinosa	Ray et al. (1980b)	30	trace	No uptake detected
Crangon crangon	Dethlefsen (1977/78)	30	1.5	No uptake detected
		30	2.5	0.76 (dry)
Homarus americanus	Thurberg et al. (1977)	30	3	0.4 (gill, wet)
		30	6	2.6 (gill, wet)
Molluscs				
Macoma balthica	Ray et al. (1980b)	30	trace	0.5 (dry)
Mytilus edulis	Westernhagen (1978)	163-300	0.5	1.3- 2.5 (dry)
			4.5	9.7-17.8 (dry)
Crassostrea virginica	Zaroogian and Cheer (1976)	287	5.0	13.5 (dry) 1.6 (wet)

where Cd_A = concentration in animals after exposure

Cd_{AC} = concentration in control animals

Data for animals exposed to water with higher concentrations of Cd (10 ppb or more) were not discussed previously. From these additional data, uptake rates were calculated as above and the rates were plotted against exposure concentration on log log paper. The line representing the relationship between uptake rate and exposure concentration was extrapolated to provide estimates of uptake rates for exposures to 0.5-2.0 ppb Cd (Table 10).

Uptake rates, where given in the literature, remained constant throughout the exposure periods; consequently, for calculating the rates (above), constant rates were assumed. Klöckner (1979) reported an exception. Polychaete worms were exposed to 10-1000 ppb Cd for 64 d and the concentration of Cd in those exposed to 1000 ppb Cd (1708 ppm Cd dry wt) was approaching an asymptote at 64 d. If so, the rate of uptake would not be constant, at least during the latter part of the exposure.

Generally the uptake rates, whether calculated or extrapolated, are low when animals are exposed to 0.5 ppb Cd or less. Comparison of the rates obtained by the two methods is possible for some of the data (Table 11). The data show reasonably close agreement except for oysters, where the calculated rate is 6 times greater than the extrapolated rate.

For 0.5 ppb Cd exposure, the polychaete *Ophrygotroche* has an uptake rate about 18 times greater than that for *Nereis* (Table 10). The extrapolated rates for oysters and mussels are equal and 16 times greater than the rates for quahogs, and 40 times greater than the rate for surf clams. The data for shrimp indicate no Cd uptake from water with trace to 1.5 ppb Cd, so the extrapolated rate from water with 0.5 ppb Cd may not apply.

The concentrations of Cd accumulated in 1 yr from water with 0.5 ppb Cd have been calculated for *Nereis*, shrimp and mussels (Table 12), using calculated and extrapolated uptake rates. This provides a rough idea of the magnitude of the effects to be expected from long exposure to low concentrations of Cd where uptake rates are low.

B. EXCRETION OF CADMIUM

Summary

From the data for crustaceans, polychaete worms and molluscs, there is little evidence to indicate that excretion of Cd occurs. Where there is an indication, the process of excretion is slow. For fish, there is evidence of Cd excretion if Cd is

Table 10. Extrapolated uptake rates of cadmium (Cd) at 0.5 to 2.0 ppb Cd for animals exposed to Cd at high concentrations.

Animal	Reference	Exposure time (d)	Exposure conc. (ppb)	Extrapolated uptake rates at 0.5 ppb, 1.0 ppb and 2.0 ppb Cd (rate in ppb/h)			
Polychaetes							
Nereis virens	Ray et al. (1980a)						
1-2 g		14	30-8200	0.5	1.0	1.8	(dry)
5-7 g		14	30-8600	0.3	0.6	1.0	(dry)
Ophryotrocha diadema	Klöckner (1976)	64	10-1000	9.0	13.0	19.0	(dry)
Crustaceans[a]							
Crangon crangon	Dethlefsen (1977/78)	20	5-20				
		30-40	10-100	0.7	1.1	1.8	(dry)
Pandalus montagui	Ray et al. (1980c)	14	37-40				
Molluscs							
Mytilus edulis	Phillips (1976)	35	10-40	0.2	0.4	0.7	(wet)
Quahog	Creig (1979)	43	10-20	0.012	0.022	0.040	(wet?)
Surf clam		43	10-20	0.005	0.012	0.030	(wet?)
Oyster		43	10-20	0.2	0.4	0.9	(wet?)

[a]Data for crustaceans (shrimp) show a reasonable fit by a single line.

Table 11. Comparison of rates of uptake of cadmium (Cd) obtained by calculation and by extrapolation.

Animal	Calculation exposure ppb Cd	Calculation uptake rate ppb/h	Extrapolation exposure ppb Cd	Extrapolation uptake rate ppb/h	Ratio C/E
Nereis	trace	0.3	0.5	0.3-0.5	1
Shrimp	2.5	0.76	2.0	1.8	0.4
Mussels	0.5	1.3-2.5	0.5	0.2 (wet) 2.6 (dry)	0.5-0.96
Oyster	0.5	1.2 (wet) 15.3 (dry)	0.5	0.2 (wet) 2.6 (dry)	6 5.9

administered in the diet, but not if administered in the water.

Crustaceans

In Crangon crangon, there was no loss in Cd during 12 d depuration from animals previously exposed to 5 and 10 ppb Cd in water (whole body concentrations of 2.25 and 2.60 ppm Cd dry wt respectively). For those exposed to 20 ppb Cd, there was a loss in Cd (from 5.5-4.3 ppm Cd) during the first 3 d of depuration (Dethelefsen 1977/78).

Blue crabs (Callinectes sapidus) exposed to 10 ppm Cd for 8 d showed no loss of Cd from carapace, gills or hepatopancreas during 96 h post-exposure (Hutcheson 1974).

Green crabs (Carcinus maenas) exposed to 20 ppb Cd in water for 37 d showed losses in Cd from shell, gills, hepatopancreas and whole body during 11 d depuration. About one half the Cd was lost in 11 d, the losses from exoskeleton and gills being most important (Wright 1977) (Table 13).

For Pandalus montagui exposed to 37 ppb Cd followed by 57 d in uncontaminated water, the Cd content of tail muscle, carcass, eggs and whole animal may have decreased slightly but there was a continual increase in Cd levels in the hepatopancreas (Ray et al. 1980c).

Polychaetes

Several groups of small and large Nereis virens with concentrations of Cd from 4-21 ppm dry wt showed little or no excretion of Cd during periods up to 75 d (Ray et al. 1980a). Ueda et al. (1976) reported similar results for polychaetes during 19 d depuration.

Table 12. Estimates of cadmium (Cd) accumulated in 1 yr from exposure to cadmium in seawater at 0.5 ppb.

Animal	Rate of uptake ppb/h	Concentration of Cd accumulated in 1 yr ppm dry wt
Nereis	0.3-0.5 (Tables 9,10)	2.6-4.4
Shrimp	0.7 (Table 10)	6.1
Mussel	1.3-2.5 (Table 9)	11-22
	2.6 (Table 10)	23

Table 13. Data on excretion of cadmium (Cd).

From Wright (1977) - Shore crab exposed to 20 ppb Cd for 37 d - excretion during 11 d.

Days for excretion	Cd in tissues (μ mole g^{-1}			
	Shell	Gills	Hepatopancreas	Whole body
0	.455	.266	.261	.170
7	.388	.090	.168	.147
11	.206	.132	.219	.087
	(Losses from shell and gills important)			
Average excretion rate (as μmole/kg/h)	-1	-0.75	-0.16	-0.35

Molluscs

Oysters (Crassostrea virginica) were exposed to 15 ppb Cd in water for 40 wk, followed by a 16-wk depuration period. Cadmium concentration in the oysters did not change during this excretion period (Zaroogian 1979).

Burrowing bivalves (Scrobicularia plana) transferred between two estuaries had not attained equality of Cd concentration between the transferred and nature animals even after 1 yr (Bryan and Hummerstone 1978).

Marjori and Petronio (1973) provided data for uptake and excretion of Cd by mussels (Mytilus galloprovincialis). The mussels were exposed to 50 ppb Cd in water for 4 or 8 d. For excretion, the mussels were maintained in clean water for 5 or 27 d after exposure. The results are summarized in Table 14. The excretion rates (2.6-11 ppb/h) are 9-33% of the uptake rates (27.7-33.7 ppb/h).

Macoma exposed to 1 ppb Cd in water for 15 d showed a slight decrease in Cd concentration during the initial depuration phase with no further indication of depuration. The CF at the end of 21-d depuration was about 325 compared with the previous maximum of 370 (McLeese et al., unpubl.).

Fish

Rainbow trout were exposed to Cd in water and to Cd in the food supply. The concentration of Cd in the fish declined during 10 wk post-exposures (dilution through growth). However, the total Cd load (μg Cd/fish) remained unchanged during 10 wk post-exposure for the fish exposed to Cd in water. On the other hand, for fish exposed to Cd in food, the Cd load decreased markedly during 4 wk post-exposure when fed non-contaminated food. The authors concluded that the route of uptake of Cd, from water or from food, may influence the retention or lack of excretion of Cd (Kumada et al. 1980).

Table 14. Uptake and excretion data for cadmium (Cd) and mussels (from Majori and Petronio 1973).

Exposure time (d)	Uptake rate ppb/h	Final conc. ppm (wet)	Excretion time (d)	Excretion rate ppb/h	Final conc. ppm (wet)	Total loss ppm (wet)
Control	-	0.2	-			
8	21.6	4.144	-			
8	18.2	3.504	-			
4	33.7	3.240	5	-11	1.920	(-1.320)
4	32.5	3.224	5	- 4.2	2.720	(-0.505)
4	27.7	2.660	27	- 2.6	0.959	(-1.701)

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CADMIUM IN AMERICAN LOBSTER (HOMARUS AMERICANUS) FROM THE AREA OF BELLEDUNE HARBOUR

by

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INTRODUCTION

Following an initial assessment of information documenting ecological studies carried out by the Noranda Research Centre on behalf of Brunswick Mining and Smelting Corporation Limited on Belledune Harbour, N.B. (Environmental Protection Service, confidential communication), the Department of Fisheries and Oceans immediately initiated investigations to determine the magnitude and geographical extent of cadmium (Cd) contamination in lobsters from the Belledune Harbour area.

The information received from Noranda's studies indicated that between 1977 and 1979 Cd levels in muscle tissue (claws and tail) from lobsters captured within Belledune Harbour during the summer had risen substantially. Noranda supplied information from a 1980 sample taken in early April which indicated that Cd levels in muscle tissue were lower than in 1979 but still were significantly elevated over pre-1977 levels. It was recognized that the observed changes could reflect a shift induced by sampling period changes rather than an absolute decrease. Noranda agreed to supply the Department with the hepatopancreas from the 1980 sampling to allow a more comprehensive 1979/1980 comparison in levels to be made. Following analyses of this material, it was judged that no significant decrease in levels of Cd had occurred. The Department of Fisheries and Oceans therefore proceeded to carry out a detailed survey of Cd levels in lobsters from the area.

MATERIALS AND METHODS

Lobster capture sites are shown in Fig. 1 and 2. Animals were captured by conventional trapping methods and transported live to the laboratory (Shediac, N.B., or Halifax, N.S.), individually bagged in polyethylene. Following determination of sex, total weight and carapace length, the hepatopancreas was removed and weighed. The tail muscle was also removed from the shell. These tissues and both claws were bagged separately in polyethylene and deep-frozen. Prior to analytical sampling, the hepatopancreas was homogenized by hand-kneading.

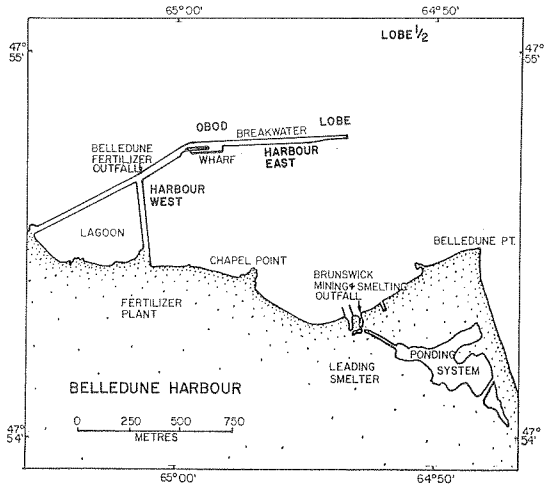

Fig. 1. Lobster sample sites within and around Belledune Harbour.

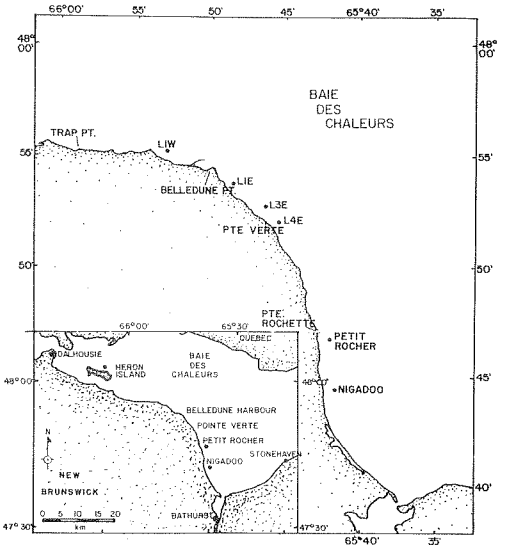

Fig. 2. Lobster sample sites surrounding Belledune Harbour.

Tail muscle samples were obtained by dissecting an appropriately sized cube from the dorsal mid-section of the right lobe. Claw samples were obtained from the dorsal mid-section of the large muscle of the claw. Care was taken to avoid thawing of muscles during dissection to avoid drip. Hepatopancreas samples (1.0-2.0 g) were digested in 50 mL Folin-Wu tubes with 5 mL conc. HNO_3 (BDH boiling chips) while muscle samples (1.0-1.5 g) were similarly digested with 1.5 mL conc. H_2SO_4 and 2.5 mL HNO_3. Initially the tubes were heated gently until frothing had ceased and then more vigorously for approximately 30 min. After cooling, each digest was made up to 25 mL in the calibrated Folin-Wu tube. Digests of hepatopancreas were assayed for Cd by flame atomic absorption spectrophotometry, taking care to ensure that the aspirator tube did not contact the coagulated fat at the surface of the digest. Flame atomic absorption conditions were: wavelength 228.3 nm, slit width 1 mm with an air/acetylene flame utilizing a deuterium arc backround corrector. External standard curves were checked by the method of standard additions. In muscle analysis, where the level of Cd was below the limits of detection of flame atomic absorption, a graphite furnace (Perkin-Elmer HGA2100) system was utilized (drying 100°C/30 sec, ashing 350°C/40 sec, and atomization 1700°C/6 sec). In both cases a Perkin-Elmer model 403 atomic absorption spectrophotometer was used. Recoveries of added Cd by these methods averaged 99.4 ± 3.05%. The laboratory has participated in the intercalibration exercises coordinated by the International Council for the Exploration of the Sea during the past number of years.

Arithmetic and geometric mean Cd levels were calculated for each sample. Relative standard deviations were calculated from the log-transformed data.

RESULTS AND DISCUSSION

In the 1979 study of lobsters captured during the summer from within Belledune Harbour and analyzed by Noranda Research the arithmetic (standard deviation) and geometric mean (relative standard deviation) levels of Cd in hepatopancreas were 88.34 (84.3) and 56.87 (28%) µg/g wet weight (28 animals). Arithmetic and geometric mean levels of Cd in hepatopancreas from lobsters captured by Noranda Research in mid-April 1980, as analyzed in our laboratory, were 151.2 (141.7) and 91.2 (23.4%) µg/g wet weight (58 animals), respectively. Resampling of the harbour was carried out by the Department of Fisheries and Oceans at the end of April 1980, and arithmetic and geometric means of 150.7 (104.5) and 105.9 (19.4%) (55 animals), respectively, were obtained. This suggests that levels of Cd in lobster hepatopancreas from Belledune Harbour had increased between the summer of 1979 and the spring of 1980. No significant difference in total weight mean exists between the two 1980 samples (arithmetic mean weights of 453 ± 235.2 g for the mid-April sample and 456 ± 239 g for the late April sample) but the 1979 mean weight of 588 ± 253 g was somewhat larger than the 1980 means. The linear relationship between total weight and Cd level in the hepatopancreas, while not always significant (see below), was always positive so the difference in Cd levels between 1979 and 1980 is probably greater still than that calculated without taking the effect of weight of the animals into account.

The late April lobster sample was taken from two areas: one along the harbour side of the conveyor causeway, designated hereafter as Harbour West; the second along the harbour side of the breakwater outwards from the loading dock, designated hereafter as Harbour East (Fig. 1, Table 1). Table 2 shows the distribution of Cd levels in hepatopancreas from lobsters captured at Harbour West and Harbour East. The distributions are skewed, showing that a geometric mean calculated from log-transformed data is a better estimate than the corresponding arithmetic means. The geometric mean for Harbour West is 175.8 µg Cd/g wet weight with a relative standard deviation of 11.7%. The geometric mean of the Harbour East samples is 62.3 µg Cd/g wet weight with a relative standard deviation of 21%, a mean significantly different ($t = 5.17$, $P < .01$) from the Harbour West mean. The data for Harbour West and Harbour East suggest that there are two distinct populations characterized by a minor degree of interpopulation migration.

Two samples were taken from the seaward side of the breakwater at Belledune Harbour. These samples had geometric means of 24.1 µg Cd/g wet weight (24.0%) for the site opposite the dock (OBOD) and 24.1 µg Cd/g wet weight (23.7%) for the site on the seaward side of the distal end of the breakwater (LOBE). This suggests that lobsters dwelling on the seaward side of the breakwater are a single population as far as Cd is concerned and that few highly contaminated animals from within the harbour migrate to the seaward side of the breakwater (both sites had a combined total of 44 animals, one of which had a hepatopancreas concentration of Cd of 209 µg/g; the next highest level observed was 68.3 µg/g). One sample of only six animals was obtained from approximately 1 km seaward from the mouth of Belledune Harbour. One of these animals had a Cd level in its hepatopancreas of 399 µg Cd/g while the six had a geometric mean of 23.1 (49.7%). The wide variation was caused by the presence of this one animal.

Levels of Cd in hepatopancreas from lobsters captured at greater distances from Belledune Harbour are shown in Table 3. Cd levels decrease in both east and west directions from Belledune harbour. The decrease is greater over the same distance towards the west than towards the east. The major current flow along this coast is from west to east (Bewers and Loring, this report), suggesting that a focus of Cd contamination exists within Belledune Harbour and that lobsters with large body burdens of Cd are somehow prevented from moving westward.

This can be best illustrated by use of frequencies for single sites from within and external to Belledune Harbour (Table 2). The data for

Table 1. Cadmium (Cd) levels (µg/g wet weight) in lobster hepatopancreas from the area of Belledune Harbour.

Sample site	Geometric mean	Relative standard deviation (%)	Range	Arithmetic mean (SD)	N
Harbour West	175.8	11.7	47.6-372	203.7 (96.5)	29
Harbour East	62.3	21.2	13.7-263	88.8 (78.9)	28
Outside harbour, opposite dock (OBOD)	24.1	24.0	4.70-68.3	30.4 (19.9)	15
Outside harbour at end of breakwater (LOBE)	24.1	23.7	4.65-209	32.8 (36.8)	30
0.8 km off end of breakwater (LOBE½)	23.1	49.0	5.97-399	80.3(156.6)	6

Table 2. Frequency of cadmium (Cd) levels in hepatopancreas of lobsters from nine areas (Fig. 1, 2).

Cadmium concentration µg/g wet weight	Number of lobsters								
	Harbour West	Harbour East	Beach Point P.E.I.	L1W	L0BE	L1E	L3E	L4E	L5E
0-10			5	15	3		3	1	2
10-20		3	14	11	7	11	8	4	8
20-30		1	4	3	10	7	10	8	5
30-40		8	2	1	2	3	6	7	1
40-50	2	3			5	3	1	3	
50-100	2	4	1		2	6	1	2	1
100-150	6	2		1		1			
150-200	3	3							
200-250	7	3			1		1		
250-300	5	1							
300-350	3								
350-400	1								
	29	28	26	31	30	31	30	25	17

Table 3. Cadmium (Cd) levels (µg/g wet weight) in lobster hepatopancreas from areas around Belledune Harbour.

Sample site	Geometric mean	Relative standard deviation (%)	Range	Arithmetic mean (SD)	N
Heron Island, N.B.	3.85	22.0	2.19-8.26	4.03 (1.31)	30
1.6 km west[a] (L1W)	11.8	28.5	4.13-119	16.3(20.0)	29
Belledune Harbour (Chapel Point) - Zero point					
1.6 km east (L1E)	28.9	18.2	10.2-110	34.9(23.7)	31
4.8 km east (L3E)	22.8	24.5	4.55-246	32.5(43.5)	30
6.4 km east (L4E)	28.0	12.5	7.13-53.9	30.15(10.8)	26
9.6 km east (L6E)	17.3	22.2	6.1-50.9	20.5 (9.6)	19
Petit Rocher, N.B.	11.6	12.3	6.26-22.1	21.2 (3.9)	31
Nigadoo, N.B.	12.1	21.1	5.00-77.3	14.4(12.6)	33
Stonehaven, N.B.	7.2	20.9	3.90-16.2	7.8 (3.3)	35
Beach Point, P.E.I. (1973)	17.2	17.7	6.07-52.0	19.4(10.3)	26

[a]Highway distances (on highway opposite Chapel Point) were: L1E - 3.1 km; L3E - 5.6 km; L4E - 7.8 km; L6E - 10.0 km; Petit Rocher - 17.4 km, and Nigadoo - 21.0 km.

lobsters from Beach Point, P.E.I. are included for comparative purposes. This area, which has no known anthropogenic inputs of Cd, had the highest levels of Cd of any of the areas sampled within eastern Canada (Uthe and Freeman, this report).

The Harbour West site is characterized by a large number of highly contaminated lobsters, i.e. 25 of the 29 (86%) individuals had hepatopancreas levels of greater than 100 µg Cd/g and none of the animals had a level below 40 µg Cd/g. In the Harbour East sample, highly contaminated animals were still present but at a substantially decreased frequency, i.e. only 9 of 27 (33%) individuals had hepatopancreas Cd levels greater than 100 µg/g while 15 (55%) of the animals had hepatopancreas Cd levels less than 50 µg Cd/g.

The frequency of highly contaminated lobsters fell sharply within the samples obtained outside of Belledune Harbour. Considering the L1W, LOBE, L1E, L3E, L4E and L6E sites as a group, only 4 (2%) out of 164 animals had a Cd level greater than 100 µg/g while 148 (90%) had levels of less than 50 µg Cd/g. These data suggest that an acute Cd contamination situation exists within Belledune Harbour and a more chronic one in areas surrounding the harbour.

Animals within the western part of Belledune Harbour are grossly contaminated (86% >100 µg Cd/g) but the frequency decreases with increasing distance from this site. All the geometric means of the Petit Rocher, Nigadoo and Stonehaven samples were below that found in the sample from Beach Point (Table 3) and may be considered as background or normal levels. However, these levels should be compared with (1) the geometric mean of 3.85 µg Cd/g in the Heron Island sample, and (2) the geometric mean of 4.30 µg/g in a sample from Petit Rocher obtained in 1973 (Uthe and Freeman, this report). These values may be a more realistic evaluation of the levels to be expected within this area had no major anthropogenic sources of Cd been present. This suggests that a generalized area of Cd pollution in lobster exists between at least the L1W site and downstream beyond Stonehaven. It is impossible to determine how much of this contamination originates from Belledune Harbour and how much from other sources such as the city of Bathurst and various rivers in this highly mineralized area. Although the geometric mean of the Nigadoo sample is larger than the corresponding mean of the Petit Rocher sample, a "t test" of the two samples using only canner-sized animals (63-80 mm carapace length) showed that there was no significant difference between the two means.

The distribution of lobsters with high body burdens of Cd with respect to the Harbour West site is approximately what has been observed in tag-recapture studies of healthy lobsters. The majority of recaptures are normally made in the immediate vicinity of the release site, and in the area adjacent to the release site, and the few others are recaptured with decreasing frequency as distance from the release site increases. Movement is usually random in direction and the apparent eastward directivity observed here is explained by two factors: the orientation of the harbour, and a gypsum-effluent bed seaward of the breakwater. Both factors would act to restrict movement to the west. It is recognized that the 1980 mean for Petit Rocher is elevated significantly over that documented in 1973. It may be suggested, based on the above, that a major single point-source of Cd pollution exists at Harbour West, some of which has been transported downstream where it has been incorporated into the lobsters' food web (e.g. mussels) and accounts for the elevation over the 1973-1980 period. The individuals containing high levels of Cd in their hepatopancreas probably reflect movement of animals from Harbour West following incorporation of Cd into their bodies.

In a few of the samplings a large enough size range was obtained to justify investigating the relationship between the animal weight and Cd level in the hepatopancreas. The results are shown in Table 4. A relatively small but significant positive relationship between total animal weight and Cd level in the hepatopancreas was found for the Harbour West and L6E sample sites (coefficients of determination of 0.24 and 0.33 respectively) but such a relationship is not precise enough to have any meaningful predictive value for calculation of the Cd levels in hepatopancreas from total weight of animals. The very low coefficients of determination found with the Harbour East and Heron Island (New Mills) sampling probably reflect mixing of animals from within and external to the harbour in the first case and the narrow weight range present in the second instance.

Time has allowed only limited studies on Cd levels in lobster muscle tissue. The results of this study are shown in Table 5. It is obvious that the hepatopancreas is a major Cd storage site since mean levels of Cd in hepatopancreas exceed corresponding muscle means by from 96 to 2560 times. Levels of Cd in tail muscle are considerably lower (3-10x) than corresponding crusher claw muscle levels. The relationships between Cd levels in the three tissues analyzed are also shown (Table 6) and again, while some significant linear relationships were found between tissue pairs, only in the case of the L1E tail and crusher claw Cd levels is the relationship tight enough to be used in a predictive manner. The lack of such a tight agreement in the L4E and L6E samples is probably due to the very low level of Cd in tail muscle from these sites in which case the analytical variance becomes a significant contributor to the variance within the system.

There was no significant difference between the geometric means for male and female lobsters from five sites (Table 7). The wide range in Cd levels, the smaller, non-normalized weight and length ranges, would mask small sexual-related differences but such small differences would have little, if any, importance to the geographical investigations carried out in this study.

It is obvious from these results that a serious problem exists with Cd in lobsters from the area of Belledune Harbour. The economic problems posed by closures and controls necessitated by the presence of Cd in human foodstuffs from the area are obvious but in addition to this is the more subtle but potentially more serious possibility of long-term ecological effects in the region. This article has only discussed Cd levels in lobster. The article by Ray et al. (this report) discusses Cd levels in biota but we have little or no information on the effects of Cd on the well-being of the ecosystem of the Belledune Harbour area in general or of lobsters in particular. Obviously, continued monitoring and studies related to human foodstuff safety considerations will have to continue. Assessment of the ecological impact upon the area should receive equivalent support.

Table 4. Hepatopancreas cadmium (Cd) levels/total weight relationships in Belledune area lobsters.

Sample site	Carapace length (mm)	N	Geometric mean (µg Cd/g wet wt)	Regression equation (coefficient of determination)
Harbour West	>80	20	223.1	
	63-80	7	100.6	(Cd) = 90.47 + 0.21 (total wt) (0.24)[a]
	<63	2	114.5	
Harbour East	>80	8	85.8	
	63-80	15	56.3	(Cd) = 60.97 + 0.07 (total wt) (0.04)[c]
	<63	3	54.4	
L6E	>80	2	36.69	
	63-80	9	16.53	(Cd) = 4.87 + 0.056 (total wt) (0.33)[b]
	<63	2	14.95	
New Mills	>80	2	5.66	
	63-80	28	3.75	(Cd) = 2.82 + 0.0035 (total wt) (0.02)[c]
	<63	0	-	

[a]Significant at $p < 0.01$
[b]Significant at $0.01 < p < 0.05$
[c]Not significant; $p > 0.05$

Table 5. Comparison of cadmium (Cd) levels (geometric means) in hepatopancreas, tail muscle, and crusher claw muscle (relative standard deviation) in lobsters from the Belledune area.

Sample site	N	Hepatopancreas (µg Cd/g wet wt) (%)	Tail muscle (µg Cd/g wet wt) (%)	Crusher claw muscle (µg Cd/g wet wt) (%)
L1E	10	24.7 (16)	0.02 (14)	0.26 (11)
L4E	12	28.2 (7)	0.01 (13)	0.04 (10)
L6E	12	20.8 (17)	0.01 (20)	0.04 (14)

Table 6. Relationships between cadmium (Cd) content (µg Cd/g wet wt) of hepatopancreas (µg Cd/kg wet wt), tail muscle, and claw muscle for lobsters from three sample sites.

Sample site	Regression equation	Coefficient of determination
L1E	(Cd) crusher = 3.645 + 0.069 (Cd) tail	0.94[a]
	(Cd) tail = 12.38 + 0.36 (Cd) hepatopancreas	0.41[b]
L4E	(Cd) crusher = 14.62 + 1.99 (Cd) tail	0.29[c]
	(Cd) tail = 6.03 + 0.19 (Cd) hepatopancreas	0.22[c]
L6E	(Cd) crusher = 15.41 + 2.61 (Cd) tail	0.54[a]
	(Cd) tail = 4.16 + 0.31 (Cd) hepatopancreas	0.44[b]

[a]Significant at $p < 0.01$
[b]Significant at $0.05 > p > 0.01$
[c]Not significant; $p > 0.05$

Table 7. Cadmium (Cd) levels (µg/g wet weight) in male and female lobster hepatopancreas; number sampled in parentheses.

Sample site	Male Geometric mean (N)	Female Geometric mean (N)	Statistical significance
Heron Island	3.66 (14)	4.10 (16)	NS[a]
Harbour West	170.1 (21)	191.8 (8)	NS
Harbour East	58.79 (18)	69.93 (9)	NS
L1E	27.05 (16)	26.54 (15)	NS
Petit Rocher	12.23 (15)	11.08 (16)	NS

[a] $p > .10$

CADMIUM IN HEPATOPANCREAS OF AMERICAN LOBSTER (*HOMARUS AMERICANUS*) FROM EASTERN CANADA

by

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INTRODUCTION

This study was initiated in 1973 when the Inspection Service of the Canadian Department of Environment became concerned about the levels of cadmium (Cd) in commercially canned lobster paste. Lobster paste is a popular food product prepared, in large part, from lobster hepatopancreas. Therefore, a study of Cd levels in the hepatopancreas of lobsters from different areas of eastern Canada was carried out.

MATERIALS AND METHODS

Thirteen live lobsters of each sex weighing between 400 and 700 g were obtained from each site by field personnel of the Department of Environment. Each sample area was not over 15 km in diameter and each sample was purchased during a single day of fishing.

Before sampling the lobsters, the sex, carapace length, and total weight were recorded. The hepatopancreas was removed intact, placed in a polyethylene bag, and frozen for transportation to our laboratory. There, each hepatopancreas was weighed, thawed, and thoroughly mixed by kneading within the bag. The Cd concentrations were determined in duplicate acid digests (5 mL H_2SO_4/HNO_3; 4:1, v/v) of 0.2-0.5 g portions by atomic absorption spectrophotometry (Perkin-Elmer Model 403 fitted with deuterium arc background correction). Either a flame or graphite furnace was employed as well as the method of standard additions as required.

The geometric mean concentration of Cd was calculated for each site since probit analysis indicated that the log-transformed concentrations were more normally distributed than the non-transformed data. Relative standard deviation of each geometric mean was also calculated.

RESULTS AND DISCUSSION

The atomic absorption method used in this study for determining Cd was previously validated by using a common standard atomic absorption method where chelated (APDC) Cd was extracted into an organic solvent (MIBK) followed by atomic absorption spectrophotometry. When our laboratory participated in the International Council for Exploration of the Sea's intercalibration studies, our results for Cd fell in the mid-range of the values submitted by the other participants (Topping and Holden 1978).

The results are shown in Table 1. The geometric means vary from a low of 2.82 µg of Cd/g (wet wt) of hepatopancreas for lobsters from Comfort Cove to a high of 17.22 µg/g (wet wt) for those from Beach Point. The arithmetic mean of all geometric means was 8.92 ± 3.47 µg Cd/g (wet wt).

In spite of the log transformation of the data, the relative standard deviations of each geometric range were generally in the 20-35% range, demonstrating that a wide range of Cd levels exists within the population of each sample area. Some of the variance in the results can be explained by the differences in the sizes (total weights) of the animals obtained at each site. The samples taken at Caraquet, Neguac, and Petit Rocher had a significantly lower average weight than the other samples. The effect of weight on Cd is small, as judged from the following observations: The selection of the larger animals from Petit Rocher (mean weight 516 ± 40 g) and Shippegan (mean weight 503.2 ± 37.2) gave geometric means of Cd for hepatopancreas of 5.13 and 4.37 µg/g (wet wt) respectively, compared with the means of 4.30 and 3.79 µg/g (wet wt) respectively for the complete samples.

The narrow range of sample weights from each site makes it impossible to assess accurately the relationship between Cd levels in hepatopancreas and total weights. We therefore sampled as wide a total weight range (market size) of lobsters as possible (N=41) in one area (Lismore). The relationship between the total weight in grams and the hepatopancreas's Cd level in µg/g (wet wt) was (Cd HP) = 3.233 + 0.013 (total wt, g) with a coefficient of determination of 0.301 where (Cd HP) = µg Cd/g (wet wt) hepatopancreas. The log transformation of the Cd values gave $\log_{10}$ (Cd HP) = 0.763 + 0.000381 (total wt) with a coefficient of determination of 0.346.

In a similar manner the relationship between carapace lengths and Cd level in hepatopancreas from the Lismore sample was calculated to be log (Cd HP) = 0.247 + 0.084 (carapace length, cm). The coefficient of determination was 0.314. The total weight and length ranges used in the Lismore study were 425-1731 g and 7.7-13.6 cm respectively.

Some caution should still be used in interpreting differences in levels among the various areas sampled (Table 1) since mean weights of sampled lobsters differ.

It is of interest to consider temporal changes in Cd levels in hepatopancreas. Five areas were sampled 1 yr later (Table 2). In this case, animals were selected from within the same weight range. The geometric means of the Cd levels at Beach Point and Meat Cove were significantly different from those found at the same sites the previous year ($p < 0.01$ and $p < 0.05$ respectively). We believe that the difference at Meat Cove could be caused by the last sample being caught at somewhat different locations but in the same general areas. The marked difference in means for the Beach Point samples is probably a result of sampling completely different sites in the 2 yr. Support for this hypothesis is apparent in the Belledune study where significant changes in Cd levels in lobster hepatopancreas occurred over distances less than 15 km. These five resampled areas are not in the vicinity of intensive human habitation or industrial activity and it is unlikely that the changes observed in two of them were due to changes in Cd loading of the environment. The distances between the designated sites on the north side of Prince Edward Island were not large; therefore, changes in climate between the 2 yr is an unlikely cause of the difference obtained in the Beach Point means for, if this were true, the other areas would also be affected.

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Table 1. Cadmium (Cd) in hepatopancreas of American lobster (Homarus americanus) from sites in eastern Canada in 1973.

Location	No. of animals	Total weight of lobsters (g) Arithmetic mean (SD)[a]	Weight of hepatopancreas (g) Arithmetic mean (SD)	Cd in lobster hepatopancreas µg/g wet weight Geometric mean (RSD %)[b]	Range
NEWFOUNDLAND					
Arnold's Cove	50	532(96)	32.1(9.7)	8.82(31)	2.45-48.9
Comfort Cove	21	538(115)	28.1(6.3)	2.82(34)	1.72- 6.43
Lark Harbour	25	528(75)	28.5(73)	10.89(21)	3.78-33.6
Britannia	20	639(200)	33.1(100)	12.63(19)	4.82-30.3
Bay l'Argent	26	559(97)	32.9(8.0)	10.83(16)	4.59-20.2
Port aux Basques	26	476(91)	29.5(5.1)	10.70(23)	4.25-45.0
NOVA SCOTIA					
Arichat	25	536(70)	26.0(4.6)	7.87(27)	3.22-19.6
Cape John	25	468(54)	22.9(3.8)	9.34(20)	3.06-18.0
Cheticamp	23	613(119)	34.4(7.9)	9.14(36)	2.13-36.3
Dingwall	26	573(139)	33.9(8.6)	13.30(20)	5.14-27.4
Frambois	25	499(61)	23.3(5.5)	12.51(22)	4.05-52.9
Gulliver's Cove	25	466(52)	24.3(3.1)	8.84(16)	4.70-18.0
Judique	25	442(73)	23.3(5.6)	8.78(18)	4.54-25.8
La Have	26	557(83)	28.0(5.1)	8.15(18)	2.94-15.4
Larry's River	26	541(94)	27.4(4.3)	8.27(22)	3.92-25.1
Lismore	17	542(72)	25.3(3.7)	8.64(22)	3.82-24.5
Liverpool	26	547(80)	21.5(5.3)	5.19(34)	1.33-16.7
Lockeport	23	506(83)	26.9(5.3)	7.38(28)	2.80-22.4
Main-à-Dieu	26	552(98)	30.8(4.8)	12.58(22)	4.89-47.2
Meat Cove	26	547(124)	29.6(5.8)	6.32(23)	1.85-12.1
Parrsboro	26	612(104)	32.4(8.4)	9.13(31)	1.02-59.0
Pictou	26	481(59)	23.1(3.3)	14.36(27)	3.77-79.1
Pubnico	24	480(65)	24.5(4.8)	10.42(4)	5.18-21.4
Sambro	25	679(79)	30.1(5.6)	8.03(23)	2.0 -18.0
Tangier	26	495(58)	24.5(3.7)	6.15(35)	2.4 -20.0
Victoria Beach	26	495(58)	22.3(2.9)	6.47(38)	1.60-22.8
Whale Cove (Digby)	24	457(52)	23.6(4.7)	7.05(29)	3.77-20.7
PRINCE EDWARD ISLAND					
Beach Point	26	526(89)	24.7(5.2)	17.22(18)	6.07-52.0
French River	26	491(94)	24.5(5.5)	11.95(31)	2.29-48.3
North Lake	24	599(135)	31.4(7.1)	16.73(20)	7.18-53.4
Tignish	25	538(119)	27.3(7.0)	14.03(19)	5.52-37.8
NEW BRUNSWICK					
Back Bay	26	471(82)	24.6(4.4)	5.31(30)	1.76-14.5
Cape Tormentine	25	561(112)	27.0(6.3)	10.90(18)	4.13-22.4
Caraquet	17	380(52)	22.2(3.2)	3.47(33)	1.37-8.46
Grand Manan	26	530(52)	26.8(4.4)	6.93(35)	2.60-31.5
Maces Bay	25	580(99)	29.7(5.7)	9.65(31)	2.93-40.7
Miscou River	26	489(88)	25.4(6.9)	4.38(31)	1.65-10.6
Nequac	26	350(61)	21.3(5.7)	4.72(24)	2.38- 9.7
Petit Rocher	26	436(74)	25.0(5.2)	4.30(32)	1.89-10.7
Richibucto	26	460(70)	22.2(3.3)	6.26(23)	3.54-14.4
Shippegan	26	478(89)	25.6(4.6)	3.79(39)	1.35-11.3
QUEBEC					
Ste-Thérèse-de-Gaspé	26	561(124)	27.3(5.5)	10.53(33)	3.82-47.4
Arithmetic overall mean				8.92(3.47)	1.02-79.1

[a] - Standard deviation
[b] - Relative standard deviation

Table 2. Cadmium (Cd) content in hepatopancreas of lobsters from five areas in 1974 and 1975; total weight range of lobsters 452-581 g.

Area	Cd in hepatopancreas, µg/g wet weight: Geometric mean 1974	Geometric mean 1975	t	N	Significance
Tignish, P.E.I.	14.48(11)[a]	15.54(19)	0.23	28	NS[b]
French River, P.E.I.	11.88(6)	12.06(20)	0.04	24	NS[b]
Beach Point, P.E.I.	17.05(7)	8.44(19)	4.30	24	HS[c]
North Lake, P.E.I.	14.65(14)	14.34(13)	1.02	25	NS[b]
Meat Cove, N.S.	6.32(17)	10.77(8)	2.475	23	S[d]

[a]Number of animals
[b]Not significant
[c]Highly significant ($p < 0.01$)
[d]Significant ($p < 0.05$)

STUDIES ON THE FORCED DEPURATION OF CADMIUM FROM THE AMERICAN LOBSTER (*HOMARUS AMERICANUS*)

by

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INTRODUCTION

Cadmium (Cd) levels in lobster hepatopancreas are very high (1-50 µg Cd/g wet weight) even in lobsters obtained from areas with minimal human impact (Uthe and Freeman, this report). High levels of Cd could be expected to affect sales of lobster since Cd is known to accumulate in the kidney of man throughout life and believed to begin inducing irreversible kidney damage at kidney cortex levels of approximately 200 µg Cd/g (Friberg et al. 1971). The availability of a simple operation by which a frontal lobe of one side of the hepatopancreas could be removed enabled timed, forced depuration studies to be carried out by using measurements over time of an individual animal's hepatopancreas level of Cd.

MATERIAL AND METHODS

Lobsters weighing between 450 and 600 g were held in individual plastic tanks, each with running sea water (~10°C at 100-200 mL/min) which was aerated prior to individual delivery to each tank. The frontal lobe of the hepatopancreas was removed by scribing a circular groove with a dental drill (#2 round burr) about 1-1½ cm in diameter into the carapace, just enclosing the white tendon mark (behind and below the eye) at about 10 o'clock and cutting the rest of the way through the shell with a scalpel at an angle describing a cone with its apex inside the lobster. This sloping cut will allow easy reattachment of the shell piece. The shell and its underlying structure are cut far enough around to allow lifting of the incised area. The frontal lobe of the hepatopancreas is carefully grasped and the isthmus joining the two lobes severed with a sharp scalpel. The shell and underlying parts were replaced and the shell piece glued to the main body of the shell with a thin layer of N-histoacryl-Blau, a commercially available surgical cyanoacrylate glue (Dr. D. E. Aiken, St. Andrews Biological Station). Cd levels were determined in each lobe after homogenization and digestion (200 mg tissue/5 mL HNO_3) by atomic absorption spectrophotometry. Lobsters did not appear to be adversely affected by this surgery. They resumed eating about 10 d post-operatively and could successfully molt at a minimum of 40 d postoperatively.

Partially degraded sodium alginate and sodium pectate were prepared by Dr. Y. Tanaka (McGill University). The efficiency of these materials in preventing intestinal absorption of administered Cd has been described (Skoryna et al. 1972).

Diets were composed of 20% cellulose (replaced by either 5% alginate or pectate as required) and 75% clams cast in 5% gelatin (J. Castell, personal communication). All animals were fed the control diet (20% cellulose) for 1 wk, 14 d postoperatively prior to initiation of the experiment. Lobsters were fed 10 g diet (as one piece) three times a week over the course of the experiment. After 10 wk the remaining frontal lobe was removed. Feeding was resumed 2 wk postoperatively for an additional 88 d. Consumption by each animal was determined by removing, weighing and discarding remaining food just prior to the next feeding. Each of the three feeding groups, containing 28 animals (mixed sexes) at the start of the experiment, suffered some mortalities during the course of the experiment (control group - 14 mortalities; alginate group - 9 mortalities; pectate group - 6 mortalities). Analysis of sections of tissue from a single hepatopancreas demonstrated that insignificant (<10%) differences in Cd levels exist among various sections of the gland. Cd analyses were carried out as described by Uthe and Freeman (this report).

Since long-term feeding experiments are not practical from an industrial point of view, the feasibility of removing Cd by injection of a common chelator was investigated. Disodium calcium ethylenediaminetetracetate (Na_2Ca EDTA), as well as the divalent cations calcium or zinc or calcium gluconate were tried. All solutions were made up in 3% NaCl to approximate isotonicity with lobster blood.

Animals were to be injected every 2 d from 9 d postoperatively. The injections were administered into the ventral sinus, using disposable tuberculin syringes fitted with 25G needles. The following solutions were administered:

1. 10 mg calcium/mL as Na_2Ca·EDTA;
2. Solution No. 1 containing 7.5 mg Ca gluconate/mL;
3. Solution No. 1 containing 0.3 mg Na_2Zn·EDTA/mL.

All animals received 5 mg Na_2Ca EDTA/kg per dose. Gaffkemia infection was found in the lobsters after five injections (12 d). The experiment was terminated immediately and the remaining frontal lobe of the hepatopancreas removed.

RESULTS AND DISCUSSION

The results of the modified sodium alginate or pectate feeding experiments are shown in Table 1. The mean values for the individual groups at the start of the feeding period, the 70-d point and the 158-d termination are also shown. In all cases means were calculated only for those animals sampled at two relevant periods. Differences were calculated on the basis of individual data obtained for each animal and significance levels were determined by paired *t*-tests. Positive difference values indicate decreasing levels of Cd over the test period. Only animals which consumed more than 5 g food per feeding period were included in these calculations.

It is obvious from the data that dietary administration of insoluble, indigestible, Cd-binding agents is unable to bring about a large decrease in the level of Cd in the hepatopancreas. Of course, it is possible that lobsters could digest these degraded carbohydrates but, from the copious quantities of feces present in the tanks, it is doubtful. Attempts were made to collect and weigh feces but variability in feeding and losses through overflow prevented accurate determination of daily fecal mass. In the control group it appears that Cd levels might increase somewhat over the course of the experiment. If this is so, then the alginate and pectate feeding effect may be greater than the small decreases in Cd levels found but the effects are still of little significance to any practical usage.

The large relative standard deviation in hepatopancreas Cd levels (ranges from 35-80%) makes

studies of this sort extremely difficult unless techniques like the surgical removal of hepatopancreas portions are used to utilize each animal as its own control. The operation is fast, simple and apparently does not harm the animal since feeding, growth and molting are readily resumed. Gross autopsy of animals which had been held post-operatively for 1 yr or more in our aquaria showed that the frontal lobe does not regenerate. However, adhesions between the hepatopancreas and other soft tissues at the severing site were noted.

Injection of divalent ions and chelators at relatively high levels did not induce removal of Cd from the hepatopancreas (Table 2). The log of the binding constant of Cd EDTA is 16.5. This suggests that, if any appreciable Cd equilibrium exists between haemolymph and hepatopancreas cells and a reasonable level of Cd is present in the haemolymph, EDTA should effect rapid removal of Cd from the hepatopancreas. This did not happen, suggesting that either the concentration of free Cd in the blood is too low to form appreciable amounts of EDTA complex or the animal is unable to excrete the complex. It is doubtful that the latter suggestion holds since EDTA is rapidly excreted by mammalian kidney and not reabsorbed. It was also hoped that the fortification of the injection solution with calcium gluconate or zinc could increase the availability of Cd in the haemolymph by competitive action of these divalent cations for Cd binding sites. No such effect was found within the relatively short time period of this experiment.

All of these results suggest that Cd in lobster hepatopancreas is extremely tightly bound and dynamically unavailable to either the haemolymph or the gut lumen. This is not unexpected since, if one considers a background level of Cd in water to be around 0.1 µg/L or less, the concentration factor to yield 10 µgCd/g of hepatopancreas would be at least 100,000.

The very high concentration of Cd present in hepatopancreas makes it tempting to postulate a biochemical role for Cd in the lobster. This would not be novel since a biological requirement for Cd in optimum growth in rats has been described (Schwarz and Spallholz 1978) but the high level may simply reflect a phenomenon similar to man in which Cd is stored in the kidney, the level increasing with age (Friberg et al. 1971). Cd in lobster increases with increasing weight (Uthe and Freeman, this report) so it is likely that the hepatopancreas simply acts as a reservoir for Cd in lobster.

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Table 1. Effect of feeding degraded sodium alginate or sodium pectate upon cadmium (Cd) levels in lobster hepatopancreas - means and differences (µg Cd/g wet wt).

Days 0	Days 70	Days 158	N	Difference[a]
		Control group		
7.88 ± 2.91	6.93 ± 2.70	-	22	0.75 ± 2.46[b]
-	6.83 ± 3.20	9.27 ± 4.09	12	-2.52 ± 2.59
7.68 ± 3.36	-	9.27 ± 4.09	12	-1.59 ± 2.74
		Sodium alginate group		
10.61 ± 6.40	9.50 ± 4.23	-	22	1.10 ± 4.64
-	9.28 ± 4.72	7.98 ± 4.22	16	1.37 ± 3.78
10.42 ± 7.15	-	7.98 ± 4.22	16	2.45 ± 4.82[c]
		Sodium pectate group		
12.41 ± 9.70	10.59 ± 7.12	-	26	1.81 ± 4.15
-	10.78 ± 7.26	10.87 ± 7.79	20	-0.09 ± 2.07
12.40 ± 9.07	-	10.87 ± 7.79	20	1.53 ± 2.50

[a] A positive value indicates a drop in Cd levels over the time period.
[b] Mean ± standard deviation for individual animal differences.
[c] Not significant at the 5% level; all other differences are not significant.

Table 2. Effectiveness of intrasinal injections upon cadmium (Cd) levels in hepatopancreas.

Treatment		Mean ± Std. Dev.	N	Difference[a]
$CaNa_2$ EDTA	Start	12.13 ± 3.43	7	
	End	11.71 ± 5.28	7	0.41 ± 1.60[b]
$CaNa_2$ EDTA + $ZnNa_2$ EDTA	Start	11.25 ± 3.33	7	
	End	10.78 ± 2.12	7	0.49 ± 1.58
$CaNa_2$ EDTA + Ca Gluconate	Start	13.88 ± 8.15	7	
	End	13.78 ± 9.33	7	0.27 ± 2.30

[a]Mean ± standard deviation for individual animal differences.
[b]All differences are not significant.

THE UPTAKE OF CADMIUM BY RATS FROM A DIET BASED ON CANNED AMERICAN LOBSTER (*HOMARUS AMERICANUS*) HEPATOPANCREAS PREPARATION

by

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INTRODUCTION

This paper is a summary of a publication (Uthe and Chou 1980) on the uptake of cadmium (Cd) by rats from a diet based on canned lobster hepatopancreas, and from a generally used experimental diet based on casein. Both casein- and hepatopancreas-based diets contained equivalent amounts of the essential nutrients for rats. Canned lobster hepatopancreas was selected as one of the diets because hepatopancreas is the major ingredient of canned lobster paste, a commercial product retailed in Canada.

MATERIALS AND METHODS

Diets were prepared based upon either casein (10 or 20% protein) or canned lobster paste (10% protein). Diets were isocaloric (332 cal/g), contained Cd at 20.9 µg/g wet weight and for all practical purposes had the same coefficients of digestibility. Cd chloride was used to fortify both casein diets. Levels of calcium, selenium, and zinc in deficient diets were fortified to the highest diet level with calcium chloride, sodium selenite and zinc sulfate. Groups of 18 young female Sprague-Dawley rats were individually fed ad libitum for 90 d. Body weights were determined twice weekly and food ingestion was determined daily. At the end of the feeding experiment, following decapitation, the brain, liver, kidneys and spleen were removed from each animal, blotted, freed of extraneous tissue and weighed. After homogenization with water, the samples were frozen for storage until analyzed for Cd by atomic absorption spectrophotometry. Due to analytical difficulties encountered by the contractor, we reanalyzed as many of the original homogenates as possible (Uthe et al., this report).

RESULTS AND DISCUSSION

The animals in the various groups did not eat significantly different quantities of food, nor show significantly different weight gain.

Average Cd levels for kidney, liver, spleen and brain from each dietary group are shown in Table 1. Cd levels in kidney, liver and spleen of rats on the lobster hepatopancreas diet were significantly lower than those of rats on the casein-based diets spiked with Cd chloride. Cd uptake in kidneys and livers of rats on the first diet was only 55 and 68% respectively of the amounts taken up by these tissues in rats on the second diet (Table 2).

The amount of "free" (polarographically active) Cd in the canned lobster hepatopancreas was 45% of total Cd.

These results indicate that "free" rather than "total" Cd of a diet determines the extent of Cd uptake. Hepatopancreas, when assayed after removal from freshly killed animals, had no detectable amount of free Cd. If the uptake is dependent upon the presence of free Cd, then Cd in hepatopancreas from a boiled lobster may be much less available than Cd from canned lobster hepatopancreas, since boiling is a much less vigorous treatment than the canning process.

REFERENCES

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Table 1. Cadmium (Cd) (µg/g wet weight) in tissues from rats fed various diets.

	Diet								
	Casein						Lobster hepatopancreas		
	10% protein			20% protein			10% protein		
	Mean	SD	N	Mean	SD	N	Mean	SD	N
Kidney	12.32	2.41	15	11.2	3.0	16	5.06	1.41	11
Liver	5.29	0.97	16	5.80	1.36	12	2.72	1.56	10
Spleen		-		0.79	0.28	18	0.23	0.117	18
Brain	0.019	0.014	15	0.034	0.015	16	0.017	0.013	18

Table 2. Total uptake of cadmium (Cd) (µg), mean and (SD), in tissues from rats fed various diets.

	Diets		
	Casein		Lobster hepatopancreas
	10% protein	20% protein	10% protein
Total Cd ingested	20300 (2800)	27200 (2300)	25700 (2000)
Total Cd kidney	20.6 (5.1)	20.1 (5.5)	10.5 (2.0)
% ingested	0.072	0.074	0.040 (55%)[a]
Total Cd liver	35.4 (6700)	39.0 (10800)	22600 (3000)
% ingested	0.118	0.140	0.088 (68%)[a]

[a] % ingested relative to casein diets

ADENYLATE ENERGY CHARGE AND ATPASE ACTIVITY IN AMERICAN LOBSTER (HOMARUS AMERICANUS) FROM BELLEDUNE HARBOUR

by

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INTRODUCTION

It has become evident that acute toxicity tests by themselves are not always reliable for the monitoring or assessment of aquatic pollutants. There is an increasing need to augment such data with indicators of sublethal effects. The pollution with cadmium (Cd) of Belledune Harbour is an example. There is no apparent health problem to resident lobsters (Homarus americanus); however, the Cd concentration in hepatopancreas has reached undesirable levels for human consumption.

Adenylate energy charge (AEC) is a measure of the metabolic energy state of the animal and is a prime factor in controlling catabolic and anabolic processes (Atkinson 1977). The rationale for the use of AEC as a biochemical indicator of stress has been reviewed (Ivanovici 1979). Changes in the adenine nucleotide pool and AEC occur in marine organisms under stressful conditions of anoxia (Zs-Nagy and Ermini 1972; Wijsman 1976; van den Thillart et al. 1980; Schottler 1978), reduced salinity (Rainer et al. 1979) and hydrocarbon pollution (Ivanovici 1979); however, with toxic levels of toluene, narcotization is just as sensitive as significant changes in ATP or AEC (Bakke and Skjoldal 1979).

Sodium, potassium-dependent adenosine triphosphatase (Na,K-ATPase) is generally considered to be the primary mediator of ion transport across cell membranes (Neufeld et al. 1980). Ouabain selectively inhibits Na,K ATPase but not other ion-regulating ATPases (residual ATPase, mostly Mg-ATPase). The effect of salinity (Neufeld et al. 1980) and metals (Watson and Beamish 1980) on the activity of ATPases has been described for aquatic fauna. Ouabain insensitive ATPase activity of lobster is stimulated by sublethal concentrations of Cd (Thurberg et al. 1977; Tucker 1979).

In the present study, AEC and ATPase activity were investigated as potential biochemical indices of Cd pollution.

MATERIALS AND METHODS

The Cd-contaminated lobsters were collected during May 1980 from coastal waters within a 1.6-km radius of Belledune Harbour breakwater. They were held for a maximum of 1 wk in commercial holding tanks, then transferred to St. Andrews, N.B. Control lobsters were purchased from commercial sources in Stonehaven, N.B., which is situated 38 km directly across Nepisiquit Bay from Belledune Harbour. Upon transfer to St. Andrews, the lobsters were held without food for 2 d in continuously flowing, aerated sea water before tissue samples were obtained. This minimized any stress induced by the transfer process. All tissue samples for ATPase, AEC and Cd assay were excised simultaneously from each lobster.

ADENYLATE ENERGY CHARGE DETERMINATIONS

The gills from the right side, a section of the hepatopancreas and a thin transverse slice of tail muscle (approximately 2 cm from the anterior end of the muscle) were freeze clamped at -196°C, using liquid N_2 temperatures (Hess and Brand 1974). The tissues were ground to a powder in liquid N_2 and stored at -80°C.

Approximately 1 g of accurately weighed, pulverized gill or hepatopancreas tissue was homogenized, at ice bath temperatures, in 2 mL of 6% $HClO_4$ with a Tekmar Tissumizer. For tail muscle 1 g of pulverized tissue was homogenized in 3 mL of 6% $HClO_4$. The homogenates were allowed to stand at 25°C for 30 min, then centrifuged (IEC-B20A) at 30,000 x G and 4°C for 15 min. The supernatants were collected, the pellet resuspended and extracted with 2 mL of 6% $HClO_4$. After centrifugation the supernatants were pooled and 0.5 mL of 1M MOPS buffer (Sigma) added. The supernatants were neutralized (pH 6.8-7.4) with 3N KOH dropwise and the resulting precipitate removed by centrifugation. The adenylates were stable in this neutralized extract for 1 wk at -80°C.

The concentrations of ATP, ADP and AMP in the neutralized extracts were determined by the enzymatic methods of Lamprecht and Trautschold (1974) and Jaworek et al. (1974) using a Beckman Model 25K spectrophotometer with the following modifications: For ATP analysis, 300 µL of gill, 300 µL of hepatopancreas or 100 µL of muscle neutralized extracts were used for the assay. For ADP and AMP determinations, 500 µL of neutralized extracts from each of the tissues were analyzed. All assays were performed in duplicate. Tris buffer (pH 7.6, 50 mM) was substituted for triethanolamine buffer and final assay volumes were kept constant for each nucleotide by addition of Tris buffer. All enzymes and substrates were purchased from Sigma (St. Louis, MO, USA) or Boehringer-Mannheim (Montreal, Que., Canada).

The AEC was calculated from:

$$AEC = (ATP + 0.5\ ADP) \div (ATP + ADP + AMP).$$

Freeze-dried tissue samples were analyzed for Cd using an atomic absorption spectrometric technique (Ray et al., this report). On the basis of Cd content, the biochemical assay results of Belledune Harbour lobsters were divided into data from contaminated and non-contaminated lobsters.

Results from the Cd and adenine nucleotide assays were evaluated on the basis of dry tissue weights. Values based on wet tissue weights can be calculated from the following moisture contents (percent moisture ± standard deviation, N=36), 77.5 ± 0.6%, 86.0 ± 1.3% and 58.1 ± 3.8% for tail muscle, gills and hepatopancreas, respectively.

ATPASE ASSAY

The ATPase assay was modified from the procedures of Neufeld and Pritchard (1979) and Tucker (1979). For each ATPase assay, all gills from the left side of each lobster were removed and immediately washed in an ice-cold solution of 250 mM sucrose and 5 mM EDTA. The gill tissue was transferred in fresh sucrose-EDTA solution to a 2-4°C constant temperature room where the tissue was blotted on Kimwipes and weighed to the nearest milligram. The tissue was transferred to 5-10 mL fresh sucrose-EDTA solution in a polycarbonate centrifuge tube and frozen in liquid nitrogen. Following freezing, the samples were stored at -80°C until ATPase measurements were carried out.

The tissue was quickly thawed, blotted and placed in a fresh volume of sucrose-EDTA in the ratio of 40 mg gill tissue per milliliter sucrose-EDTA. The gill tissue was homogenized by using a Tekmar Tissumizer. After vigorous stirring, 0.2 mL

of the gill homogenate was drawn into an Oxford pipette and added to 0.6 mL of the assay medium and preincubated for 5 min at 37°C. The reaction was started by adding 0.2 mL $MgCl_2$ - ATP solution to give a total reaction volume of 1.0 mL. Final concentrations of the assay medium were: 50 mM NaCl, 10 mM KCl, 4 mM $MgCl_2$, 6 mM disodium ATP, and 90 mM Tris buffer (pH 7.6). To determine Na,K ATPase and residual ATPase activity, the reaction was run with and without a final concentration of 0.2 mM ouabain for 10 min at 37°C and stopped by adding 4 mL of cold 1% ammonium molybdate in 1.15N H_2SO_4 into which 40 mg/mL ferrous sulfate had been dissolved just before use. Samples were centrifuged for 3-5 min at 5000 RPM (Sorvall SS3 and SS-34 rotor). The blue color of the reaction mixture was allowed to develop for 1 h before the absorbancy of each sample was measured at 700 nm. All assays were done in triplicate. The amount of inorganic phosphorus (Pi) liberated in the reaction ATP → ADP + Pi was expressed as the number of μmoles Pi/mg protein/h. The protein content of the homogenate was determined by using a modification of the Lowry method (Hartree 1972), and bovine serum albumin as a standard.

RESULTS AND DISCUSSION

Cd ANALYSIS

There was a wide variation in Cd levels between individuals, especially in those from Belledune Harbour area (large range values, Tables 1-3). Two distinct populations were found in the Belledune lobsters (N=24): those having significantly elevated Cd levels (N=17, $p < .001$) compared to the Stonehaven lobsters (N=12), and those showing no significant difference (N=7). There was no correlation between size of Belledune lobsters and their Cd content.

All data were classified, according to the Cd content of lobsters, into the following: Belledune Harbour lobsters having elevated Cd levels (BC), Belledune Harbour lobsters with control levels of Cd (BU) and Stonehaven (SH) controls. The BU lobsters probably recently migrated into the Cd-contaminated area, and were caught before significant Cd uptake occurred.

The Cd concentration of all lobsters varied among the hepatopancreas, gill and muscle (483.2 ± 153.1, 69.79 ± 27.22, 2.18 ± 1.28 ppm dry wt, respectively, Tables 1-3). The hepatopancreas Cd concentration was 5-8 times greater than the Cd concentration of gill, and gill Cd concentration was 30 times greater than the Cd concentration of muscle. Induction of metallothioneins may be involved in the preferential deposition of Cd in the hepatopancreas. The occurrence of metallothioneins has been found in several marine invertebrates (Olafson et al. 1979; Talbot and Magee 1978).

ANALYSIS OF ADENINE NUCLEOTIDES

The size of the total adenylate pool per gram of tissue in the lobsters increased in the order hepatopancreas < gills < muscle (Tables 1-3). The major cause of this disparity in distribution of the adenylates is the ATP component. The concentrations of adenine nucleotides in the tail muscle are comparable to those found in the abdominal muscle of Homarus vulgaris (Beis and Newsholme 1975). The distribution of adenine nucleotides was as expected, with tissues having high energy requirements maintaining higher levels of adenylates. Interestingly, the hepatopancreas has the highest metabolic activity but maintains the lowest concentration of adenine nucleotides. The AEC follows the same pattern, being highest in the muscle, followed by gill, then hepatopancreas, but in no instance was the AEC less than 0.8, the lowest ratio indicative of a healthy, unstressed animal (Ivanovici 1979).

The muscle and hepatopancreas of Belledune lobsters (BC and BU) showed no significant difference from SH controls in the levels of ATP, ADP, AMP, total adenine nucleotides or AEC (Tables 1, 2, 4). Gills of BC lobsters had significantly depressed levels from SH controls with ATP ($p < .001$, 76% of control), AMP ($p < .05$, 87% of control), total adenylates ($p < .001$, 79% of control) and a small but significant difference in AEC ($p < .001$) (Table 3). The BU lobsters also had significantly lower levels from SH controls (Table 4) with ATP ($p < .01$, 78% of controls), total adenylates ($p < .05$, 85% of controls) and AEC ($p < .001$). The apparent lack of difference of AMP levels of BU and SH lobsters could be related to sample size (N=7 for BU vs N=17 for BC). There were no differences in the adenine nucleotide levels and AEC for all three tissues between BU and BC lobsters.

The effect of anoxia or salinity stress on the adenine nucleotides and AEC of aquatic animals has been variable. Exposure of Mytilus edulis to air (Wijsman 1976) and exposure of three estuarine molluscs to reduced salinity (Rainer et al. 1979) caused a decrease in AEC but the total adenlyate pool remained constant. Both the AEC and total adenylates decreased during hypoxia in the muscles of goldfish, Carassius auratus (van den Thillart et al. 1980). With Tubifex, anoxia caused a decrease in AEC and ATP levels (Schottler 1978). The AEC may also remain stable when ATP decreases, by reduction of the total adenylate pool (Chapman and Atkinson 1973). The results from our lobster studies resemble the latter case. Although the AEC of gills from BU and BC lobsters is significantly lower than SH controls, the difference is small. Additionally, the AEC is not too different from the stabilized ideal value of 0.85 (Chapman et al. 1971) and well within the range (0.8-0.9, Ivanovici 1979) expected for viable cells.

The causative agent for lower AEC and adenine nucleotides of BU and BC lobsters compared to SH controls is unknown. Cd is probably not responsible since the values for BU lobsters were identical to BC lobsters. No correlation between Cd levels and any of the biochemical parameters was observed (Tables 1-3). High Cd levels were observed in the BC, but apparently have not reached toxic levels as others have remained healthy since their capture. The differences observed between Belledune Harbour area and Stonehaven lobsters may be due to geographically related reasons or to the presence of sublethal concentrations of other pollutants.

ATPASE ACTIVITY

No significant differences were found in the Na,K ATPase or residual ATPase (ouabain-insensitive) activity among BU, BC or SH lobster gills (Table 3). Previous studies with Homarus americanus demonstrated a 25% increase in residual ATPase activity after 30 d exposure to 6 ppb Cd (Tucker 1979; Thurberg et al. 1977). In these studies, the 30-d levels of Cd in the gills were 3-4 times lower than in BC lobsters. One possibility for this discrepancy is that this stimulation of residual ATPase

Table 1. Cadmium (Cd) levels and biochemical analysis results for muscle tissues of Cd-contaminated lobsters from Belledune Harbour.

Parameter analyzed		N	Mean ± 95% confidence interval	Range	Correlation[a] coefficient
Cd ppm dry wt	Control[b]	12	0.10 ±0.06	0.01- 0.35	
	Belledune[c]	17	2.18 ±1.28**[d]	0.41-11.15	
ATP µmole/g	Control	12	20.77 ±1.10	17.11-23.59	-0.291
	Belledune	17	20.49 ±2.20	6.61-26.17	-0.187
ADP µmole/g	Control	12	3.32 ±0.16	2.71- 3.88	-0.599*
	Belledune	17	3.45 ±0.36	1.95- 4.10	-0.192
AMP µmole/g	Control	12	0.52 ±0.04	0.41- 0.61	-0.594*
	Belledune	17	0.53 ±0.07	0.38- 0.94	-0.102
Total adenylates µmole/g	Control	12	24.98 ±0.96	22.95-28.08	0.174
	Belledune	17	24.49 ±2.45	8.96-30.50	-0.190
AEC	Control	12	0.911±0.003	0.902-0.920	0.437
	Belledune	17	0.905±0.009	0.845-0.921	-0.038

[a]Cd concentration vs parameter analyzed, * $p < .05$.
[b]Control animals: 6 males, 6 females; mean wt = 473 ± 71 g.
[c]Belledune Harbour Cd-contaminated animals: 14 males, 3 females; mean wt = 601 ± 77 g.
[d]Level of significance in t-test - ** $p < .01$.

Table 2. Cadmium (Cd) levels and biochemical analysis results for hepatopancreas tissues of Cd-contaminated lobsters from Belledune Harbour.

Parameter analyzed		N	Mean ± 95% confidence interval	Range	Correlation[a] coefficient
Cd ppm dry wt	Control[b]	12	30.88 ± 9.68	9.48 - 70.11	
	Belledune[c]	17	483.2 ±153.1***[d]	106.30 -1103.00	
ATP µmole/g	Control	12	2.62 ± 0.19	2.23 - 3.25	0.180
	Belledune	17	2.49 ± 0.16	1.85 - 3.03	-0.087
ADP µmole/g	Control	12	0.69 ± 0.11	0.33 - 1.03	0.366
	Belledune	17	0.67 ± 0.07	0.40 - 0.91	0.366
AMP µmole/g	Control	12	0.25 ± 0.03	0.18 - 0.35	0.338
	Belledune	17	0.24 ± 0.03	0.11 - 0.41	0.236
Total adenylates µmole/g	Control	12	3.56 ± 0.29	2.89 - 4.63	0.292
	Belledune	17	3.37 ± 0.19	2.36 - 4.13	0.125
AEC	Control	12	0.836± 0.014	0.808- 0.869	-0.055
	Belledune	17	0.831± 0.013	0.781- 0.876	-0.326

[a]Cd concentration vs parameter analyzed.
[b]Control animals: 6 males, 6 females; mean wt = 473 ± 71 g.
[c]Belledune Harbour Cd-contaminated animals: 14 males, 3 females; mean wt = 601 ± 77 g.
[d]Level of significance in t-test - *** $p < .001$.

Table 3. Cadmium (Cd) levels and biochemical analysis results for gill tissues of Cd-contaminated lobsters from Belledune Harbour.

Parameter analyzed		N	Mean ± 95% confidence interval	Range	Correlation[a] coefficient
Cd ppm dry wt	Control[b]	12	3.68 ± 0.76	1.91 - 7.19	
	Belledune[c]	17	69.79 ±27.22***[d]	24.40 -267.30	
ATP µmole/g	Control	12	5.27 ± 3.69	4.24 - 6.78	0.268
	Belledune	17	3.99 ± 0.27***	3.10 - 5.22	0.051
ADP µmole/g	Control	12	0.63 ± 0.07	0.43 - 0.85	-0.033
	Belledune	17	0.65 ± 0.07	0.32 - 0.85	0.007
AMP µmole/g	Control	12	0.55 ± 0.44	0.43 - 0.68	-0.019
	Belledune	17	0.48 ± 0.05*	0.32 - 0.64	0.082
Total adenylates µmole/g	Control	12	6.45 ± 0.44	5.10 - 8.24	0.217
	Belledune	17	5.12 ± 0.34***	4.02 - 6.62	0.054
AEC	Control	12	0.866± 0.006	0.843- 0.882	0.333
	Belledune	17	0.844± 0.008***	0.820- 0.872	-0.030
Na,K ATPase µmole Pi/mg protein/h	Control	12	4.93 ± 0.73	3.45 - 7.29	-0.330
	Belledune	16	5.07 ± 1.24	0.83 - 10.93	-0.050
Residual ATPase µmole Pi/mg protein/h	Control	12	14.88 ± 2.61	9.65 - 23.28	0.221
	Belledune	16	14.70 ± 2.69	2.65 - 20.88	0.084

[a] Cd concentration vs parameter analyzed.
[b] Control animals: 6 males, 6 females; mean wt = 473 ± 71 g.
[c] Belledune Harbour Cd-contaminated animals: 14 males, 3 females; wt = 601 ± 77 g.
[d] Level of significance in *t*-test - * $p < .05$, *** $p < .001$.

Table 4. Cadmium (Cd) levels, adenine nucleotides and adenylate energy charge of Cd-uncontaminated lobsters (N=7) from Belledune Harbour.

Tissue	Parameter analyzed	Mean ± 95% confidence interval	Range	Correlation coefficient compared to [Cd]	t-test results[a]
Gills	Cd ppm dry wt	5.58 ± 1.74	2.42 - 9.31		NS SH p <.001 BC
	ATP µmole/g	4.13 ± 0.60	2.65 - 4.95	0.289	p <.001 SH NS BC
	ADP µmole/g	0.69 ± 0.13	0.51 - 0.92	0.243	NS SH NS BC
	AMP µmole/g	0.58 ± 0.09	0.32 - 0.72	-0.325	NS SH NS BC
Total adenine nucleotides	µmole/g	5.39 ± 0.74	3.48 - 6.33	0.218	p <.05 SH NS BC
	AEC	0.830± 0.014	0.792-0.845	0.755	p <.001 SH NS BC
Tail muscle	Cd ppm dry wt	0.18 ± 0.2	0.04 - 0.45		NS SH p <.01 BC
Hepatopancreas	Cd ppm dry wt	27.17 ±14.46	7.53 -65.46		NS SH p <.001 BC

[a]SH = Stonehaven control lobsters, N=12.
BC = Belledune Harbour Cd-contaminated lobsters, N=17.
Belledune Harbour uncontaminated lobsters: 4 males, 3 females; mean wt = 503 ± 100 g.

activity is transitory and, under the more chronic exposure to Cd of BC lobsters, the stimulatory effect of Cd on residual ATPase activity is lost through some adaptive mechanism. There are large individual variations in Na,K ATPase and residual ATPase (see range Table 3) and a larger sample size may have demonstrated some differences.

CONCLUSION

Lobsters residing in the Cd-polluted waters of Belledune Harbour have remained healthy. The high levels of Cd accumulated by Belledune lobsters have not resulted in deleterious changes in ion transport in the gills, or the metabolic energy state of the animal because:

1. The AEC was within the range expected for healthy individuals;
2. Lack of correlation of Cd levels to changes in AEC and adenine nucleotide pool;
3. No significant differences of ATPase activity between Cd-contaminated and non-contaminated lobsters;
4. Lack of differences in adenine nucleotides and AEC between Belledune Harbour lobsters with high Cd levels and those with Cd levels similar to Stonehaven controls.

These criteria may still be useful to indicate sublethal effects in lobsters under stressful conditions of Cd; however, the Belledune Harbour area lobsters are unaffected by present levels of Cd.

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A PRELIMINARY STUDY OF THE HISTOLOGY OF THE HEPATOPANCREAS, GILL AND GREEN GLAND TISSUES OF AMERICAN LOBSTERS (HOMARUS AMERICANUS) COLLECTED FROM THE BELLEDUNE AREA OF THE BAY OF CHALEUR

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INTRODUCTION

High levels of cadmium (Cd) had been found in the hepatopancreas tissues of lobsters caught in the Belledune Harbour area (Uthe et al., this report). In addition to the concern that the levels were potentially dangerous to human health, it was also considered possible that the Cd could be harmful to the lobster itself. For this reason, sampling of lobster tissues for histopathological examination was carried out in conjunction with the sampling for Cd analysis.

The hepatopancreas was selected for histological study both because of its sometimes high Cd levels (Uthe and Freeman, this report) and because of its active role in the storage and metabolism of divalent metals - iron, copper and calcium - during the molt cycle (Davis and Burnett 1964; Travis 1955a). Cd affects cell membranes and for this reason the gill tissues with their thin epithelial cell covering and the epithelial cells of the tubules of the green glands were considered as potentially susceptible tissues which would be target organs of Cd toxicity.

Also, in a study on factors affecting the toxicity of Cd to the fiddler crab, Uca pugilator, O'Hara (1973) reported finding the highest concentration of Cd in the green gland, followed by gill, hepatopancreas and muscle.

MATERIALS AND METHODS

During the period April 28-May 5, 1980, 372 lobsters were sampled for chemical analysis. During this time we took tissue samples from 118 lobsters for histological study. In all cases, carapace length, weight of hepatopancreas, total body weight, sex, date and location of capture of each animal were recorded and the lobster sample numbers used for chemical and histological examination corresponded. Whenever possible, the molt cycle stage was determined.

The sampling record is summarized in Table 1. The lobsters which were sampled, the tissues sampled and the fixatives used are listed together with a key to the area where the lobster was captured and, if available, its stage in the molt cycle. An "a" in the table indicates lobsters which were selected by Dr. J. Uthe for a blind study in which the histology and the pathology (if any) of the lobsters were compared without a prior knowledge of the Cd levels found in the tissues of these animals. After fixation, all the tissues were embedded in paraffin, sectioned at 5μ and stained with haematoxylin and eosin. The slides were examined and photomicrographs taken, using a Zeiss Photomicroscope III.

RESULTS AND DISCUSSION

HEPATOPANCREAS

In considering the results of the examination of the hepatopancreas tissues, it is desirable to first consider the available literature on the histology of this organ. Three relatively recent studies are pertinent: Travis (1955a, 1957) and Davis and Burnett (1964) have described the histology of the hepatopancreas of the spiny lobster (Panulirus argus Latreille) and the crayfish (Astacus fluviatilis), respectively. Travis was concerned with cellular changes during the course of the molt cycle while Davis and Burnett used autoradiography to demonstrate the growth and cell differentiation, or ontogeny, as these authors prefer, of the cells of the hepatopancreas during the intermolt period. Of these studies, the work by Davis and Burnett is the most definitive and, although the study was done on the crayfish, our results indicate that the work may be extrapolated to apply to the American lobster.

In essence, the hepatopancreas consists of a vast number of tubules which open into a duct system. The tubules are blind sacs and, at the apex, embryonic cells are found which divide and move proximally down the tubule to become in order an absorptive cell, a secretory cell, a fibrillar cell and finally an atrophying cell at the entrance into the hepatopancreas ducts. The absorptive cell has a basal or a central nucleus, small granules and, during the premolt or early postmolt stages, has many small vacuoles at the periphery filled with calcospherites or calcium phosphate spherules. In the secretory stage the nucleus is basal and large vacuoles appear in the cytoplasm and are sometimes seen discharging their contents into the lumen of the tubule. Secretory cells become fibrillar cells and these are seen only in small numbers among the other cells. They contain iron and show striations running from the base to the lumen, and have a basally located nucleus with a large nucleolus. Finally, at the proximal end of the tubule, atrophying cells are found which are small, have pycnotic nuclei and, in some instances, the nuclei have undergone karyorrhexis.

In the examination of the Belledune lobster hepatopancreas sections, all these types of cells were observed. Sections may be cut through one tubule at the level of the absorptive cells, another tubule at the level of the secretory cells, or a longitudinal or oblique section may demonstrate the presence of all these cell types in one tubule. Most of the lobsters were staged by the method of Drach (1939) and were found to be in the intermolt or early premolt stage. Travis has observed that the cellular composition of the tubule varies during the molt cycle. She also notes that the animal's size and water temperature may affect the rate of change of cellular composition of the tubules during the molt cycle, while Ogura (1959) notes that the properties of the epithelial cells of the gland vary to a certain extent according to the individual animal studied. It is thus in the light of this information about cell types and individual variation that the sections of the hepatopancreas must be considered. Our conclusion is that no histopathological changes are to be seen in the sections which were examined and hence the elevated Cd levels in the hepatopancreas have been without detectable effect on this tissue. The cell types and variations observed were all within the normal limits of variation. However, it should also be noted that other workers have reported that crustaceans are more susceptible to the effects of toxic substances during molting or ecdysis (Saroglia and Scarano 1979). Lobsters examined in this study were all in the intermolt or early premolt stage. It is possible that, during the molt, pathological changes might be detected in the hepatopancreas of those lobsters with high Cd levels in this tissue. Representative photomicrographs of the hepatopancreas are shown in Fig. 1-4.

GREEN GLAND

In the Crustacea, the paired green or antennal glands produce a secretion or "urine" and are involved in ionic and osmotic regulation. Travis (1955b) suggests that in the spiny lobster this organ is instrumental in conserving calcium for hardening the skeleton following the molt, and this is accomplished by reducing the amount of calcium excreted in the urine. In the lobster the pair of glands are found at the base of the antennae just in front of the stomach sac and consist of an end sac, an excretory tubule or labyrinth, and a bladder which opens to the outside by a short duct. The tubules in the labyrinth are lined by single cell layers of cuboidal epithelial cells with central nuclei. Figure 5 shows a portion of the labyrinth. The cells are in the secretory stage and large vacuoles appear to be discharging into the lumen. A brush border is present on the proximal or lumen surface of the cells. The nuclei are large with scattered chromatin. Blood cells are present between the layers of tubular epithelium. The gland is normal in appearance and corresponds in description to the secretory stage of the green gland of crayfish Astacus astacus described by von Buddenbrock (1954). Once again there is no evidence of histopathological change in the green gland of lobsters containing elevated levels of Cd in their hepatopancreas tissue. This is also shown by Fig. 6, a photomicrograph of the tubules of lobster #95, with a Cd level of 304 ppm in its hepatopancreas. Cellular detail is normal.

GILLS

The lobster has 20 pairs of gills, found in narrow branchial chambers on each side of the body and covered by the overlapping carapace. The anatomy of the gills has been described briefly by Herrick (1911) while the histology and ultrastructure of two closely related species, Panulirus argus, a spiny lobster, and Procambarus clarkii, a crayfish, have been examined by Strangways-Dixon and Smith (1970) and by Burggren et al. (1974), respectively. In the lobster the gills are "feathery" structures with many fine lateral filaments extending out from a main axis. The filaments are covered with a thin chitinous layer below which is a layer of epithelial cells. Blood flows through an afferent vessel to the tip of the filament and returns to the base of the filament through a parallel efferent vessel. The vessels are separated from each other by a connective tissue septum similar to the pillar cell arrangement in fish gills. Holes in the septum near the outer edge of the filament allow some blood to cut across from the afferent vessel to the parallel efferent vessel without flowing out to the tip of the gill filament.

In this study the sections of gill examined showed the integrity of the epithelial cell layers was preserved and no differences were detectable between the gills of lobsters containing either high or low levels of Cd in their hepatopancreas (Fig. 7). Certainly it might be expected that the epithelial cells would be the first to show evidence of the toxic effects of Cd. These cells are in direct contact with the seawater and would be constantly exposed to any toxic substance in the environment.

SUMMARY AND CONCLUSIONS

The hepatopancreas, gill and green gland tissues of 118 lobsters from various areas at or near Belledune Harbour in the Bay of Chaleur have been examined histologically. No histopathological changes were observed regardless of the Cd level found in the hepatopancreas of the animals being examined. While this is an encouraging result, any enthusiasm should be modified by the following considerations: First, the lobsters examined were all in their intermolt or early premolt stage. As noted earlier, crustaceans are more susceptible to toxic substances during the ecdysis or molt period. Secondly, the hepatopancreas was examined because of the high level of Cd found in this tissue, and the gills and green glands for reasons given above. However, Cd also is known to be toxic to gonad tissues, and a more complete study should include consideration of the reproductive tissues of the lobster. Thirdly, the tissues examined were studied by light microscopy, using haemotoxylin and eosin stained material. Closer examination, staining for specific cellular constituents, or ultrastructural studies might reveal deleterious changes induced by the high Cd levels. Thus this report indicates no effect of Cd upon the lobster tissues examined but should be considered as a preliminary study. Gonadal and other tissues were also fixed and can be studied further.

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Table 1. Lobsters used in the histopathological survey.

Number	Lobster area	Stage	Tissue: Hepatopancreas H	Hepatopancreas FC	Gill H	Gill FC	Green gland H	Green gland FC	Cd in hepatopancreas (μg/g wet weight)
2	OBOD			x	x		x		
3	"			x	x		x		
4	"			x	x		x		
5	"			x	x		x		
6	"			x	x		x		
16	L1E			x	x		x		
18	"			x	x		x		
19	"			x	x		x		
21	"			x	x		x		
22	"			x	x		x		
23	"			x	x		x		
24	"			x	x		x		
25	"			x	x		x		
26	"			x	x		x		
27	"			x	x		x		
28	"			x	x		x		
29	"			x	x		x		
30	"			x	x		x		
31	HW			x	x		x		
32	"			x	x		x		
33	"			x	x		x		
34	"			x	x		x		
35	"			x	x		x		
36	"			x	x		x		
75	HE			x	x		x		
81	"			x	x		x		
83	"			x	x		x		
85	"			x	x		x		
87	"			x	x		x		
88	"			x	x		x		
89	"			x	x		x		
90	"			x	x		x		
91	"			x	x		x		
92	"			x	x		x		
93	HW		x	x			x		
94	"		x	x			x		
95[a]	"	C-Do	x	x	x	x	x	x	304
96	"		x	x		x	x	x	
97	"			x		x	x	x	
98[a]	"	C	x	x	x	x	x	x	243
99	"		x	x		x	x	x	
100[a]	"	Do	x	x	x	x	x	x	304
101[a]	"	C	x	x	x	x	x	x	329
102[a]	"	C-Do	x	x	x	x	x	x	54
103[a]	"	C	x	x	x	x	x	x	104
104[a]	"	Do	x	x	x	x	x	x	336
105	"		x	x		x	x	x	
106[a]	L1W	C	x	x	x	x	x	x	7.50
107	"		x	x		x	x	x	
108[a]	"	C	x	x	x	x	x	x	9.93
109	"		x	x		x	x	x	
110	"		x	x		x	x	x	
111	"		x	x		x	x	x	
112	"		x	x		x	x	x	
113[a]	"	C	x	x	x	x	x	x	5.66
114[a]	"		x(?)	x	x	x	x	x	119
115	L3E	C		x					
116	"			x					
117	"	C-Do		x					
118	"	C		x					
119	"			x					

Table 1. (cont'd)

Number	Lobster area	Stage	Tissue: Hepatopancreas H	Hepatopancreas FC	Gill H	Gill FC	Green gland H	Green gland FC	Cd in hepatopancreas (µg/g wet weight)
120	"			x					
121	"			x					
122	"			x					
123	"	C-Do		x					
124[a]	HW	C		x					333
125[a]	"	Co		x					222
126	"			x					
127	"			x					
128	"			x					
129	"			x					
130	"			x					
131	"			x					
132[a]	"	C-Do		x					47.6
133	"			x					
134	L1W			x					
135[a]	"			x					7.88
136	"			x					
137	"			x					
138	"			x					
139	"			x					
140	"			x					
141	"			x					
142	"			x					
143	L1E			x					
144	"			x					
145[a]	"	C-Do		x					110
146	"			x					
147	"			x					
148[a]	"			x					10.2
149	"			x					
150	"			x					
151	"			x					
152[a]	"			x					25.8
153[a]	"			x					43.9
156[a]	"			x					154
223	L6E		x	x	x		x		
224	"		x	x	o		x		
226[a]	"	C-Do	x	x	x	x	x		50.9
227	"		x	x	x		x		
228[a]	"	C	x	x	x	x	x		6.12
229	"		x	x	o		x		
230[a]	"	C	x	x	x	x	x		16.3
231[a]	"	C	x	x	x	x	x		21.9
232[a]	"	C-Do	x	x	x	x	x		
233	"		x	x	x		x		26.4
242	L1W		x	x	x		x		
243	"		x	x	x		x		

Table 1. (cont'd)

Number	Lobster area	Stage	Tissue: Hepatopancreas H	Hepatopancreas FC	Gill H	Gill FC	Green gland H	Green gland FC	Cd in hepatopancreas (µg/g wet weight)
250[a]	NIGADO	Do-C	x	x	x	x	x	x	12.2
251[a]	"	C-Do	x	x	x	x	x	x	5.66
252[a]	"	C	x	x	x	x	x	x	14.6
253[a]	"	C	x	x	x	x	x	x	8.57
254[a]	"	C	x	x	x	x	x	x	8.46
255[a]	"	C-Do	x	x	x	x	x	x	18.3
256[a]	"	C-Do	x	x	x	x	x	x	17.1
257[a]	"	C-Do	x	x	x	x	x	x	10.3
258[a]	"	C-Do	x	x	x	x	x	x	10.2
259[a]	"	C-Do	x	x	x	x	x	x	12.3

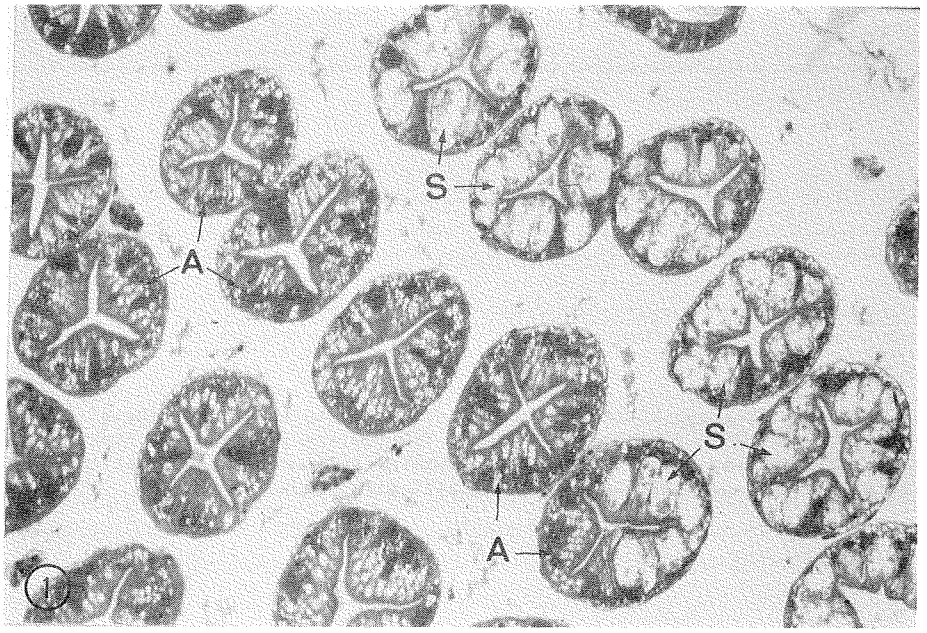
x - Tissues fixed and processed
H - Helly's fixative
CF - formal calcium fixative

Areas:
- L1W - 1 mi west of Belledune Harbour
- L3E - 3 mi east of Belledune Harbour
- HW - harbour west, inside harbour
- L1E - 1 mi east of Belledune Harbour
- OBOD - outside harbour, opposite dock
- HE - harbour east, inside harbour
- L6E - 6 mi east of Belledune Harbour
- NIGADO - Nigadoo, N.B.

[a]Samples which were compared in a blind study to attempt to discover if there was a correlation between Cd levels in the hepatopancreas and any histopathological findings in these animals.

Fig. 1. A photomicrograph of a transverse section of several tubules of the hepatopancreas of lobster #103, fixed in Helly's fluid, sectioned at 5μ and stained with haematoxylin and eosin. The tubules are shown sectioned at different levels. Some consist mostly of absorptive cells (A), tall columnar cells with a basal nucleus and a striated border facing the lumen. Others are mostly secretory cells (S), also with basal nuclei, but large secretory vacuoles. Some tubules were sectioned in the transition zone and both secretory and absorptive cells are present. The Cd level in this lobster was 104 ppm. There is no evidence of tissue damage; magnification 160x.

Fig. 2. A photomicrograph of the hepatopancreas of lobster #232, in the early premolt stage. A tubule is seen sectioned transversely through the absorptive cell region and many small vacuoles are evident containing calcospherites (Arrows). The calcospherites are also evident in an adjacent oblique section of a tubule. Loose connective tissue is present between the tubules (C). The Cd level in the hepatopancreas of this lobster was 26.4 ppm (Helly's fixative, H and E stain; magnification 250x).


S
A
S
A
1

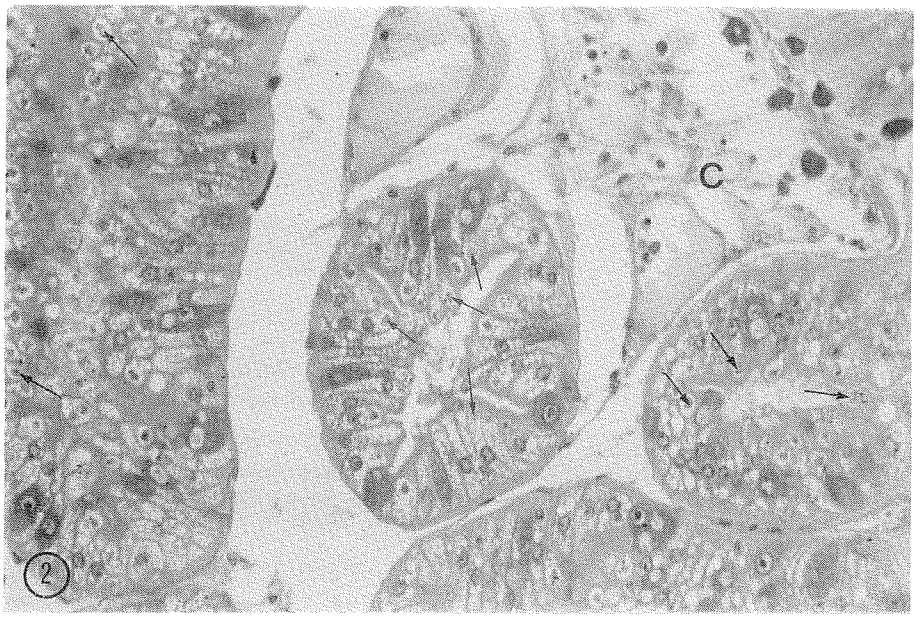
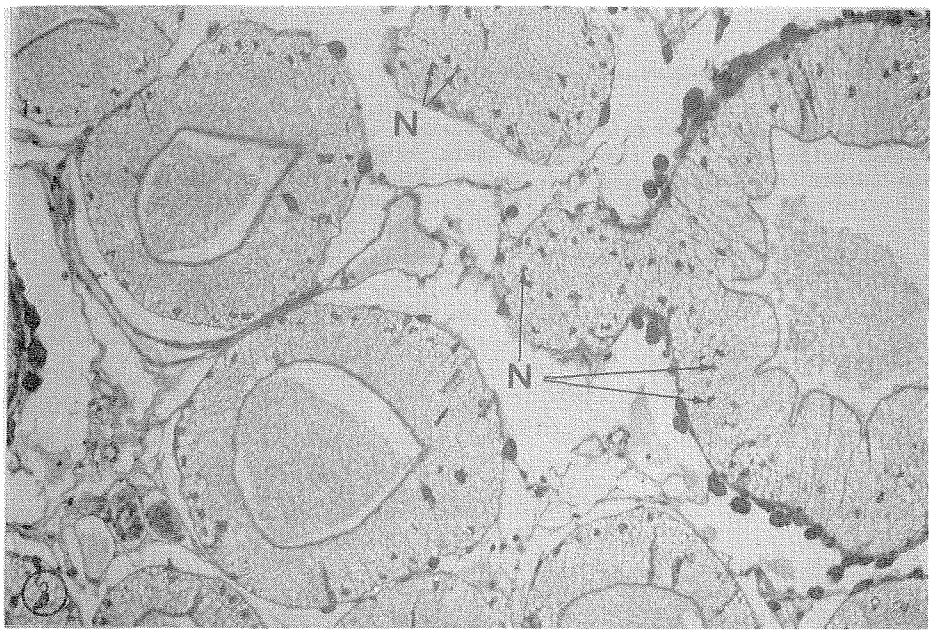

C
2

Fig. 3. A photomicrograph of transverse sections of tubules of the hepatopancreas of lobster #251 (Cd level 5.6 ppm). Tubules are sectioned at proximal end near hepatopancreas duct. Cells are atrophying and nuclei are pycnotic (N). Lumen is filled with granular material; magnification 160x.

Fig. 4. A photomicrograph of the hepatopancreas tubules of lobster #230, Cd level 16.3 ppm. Tubules are cut obliquely and transition from embryonic cells (E) to absorptive cells (A) to secretory cells (S) is seen. Lobster is in intermolt C stage and no calcospherites are visible (Helly's fixative, H and E stain; magnification 160x).


N
N
2

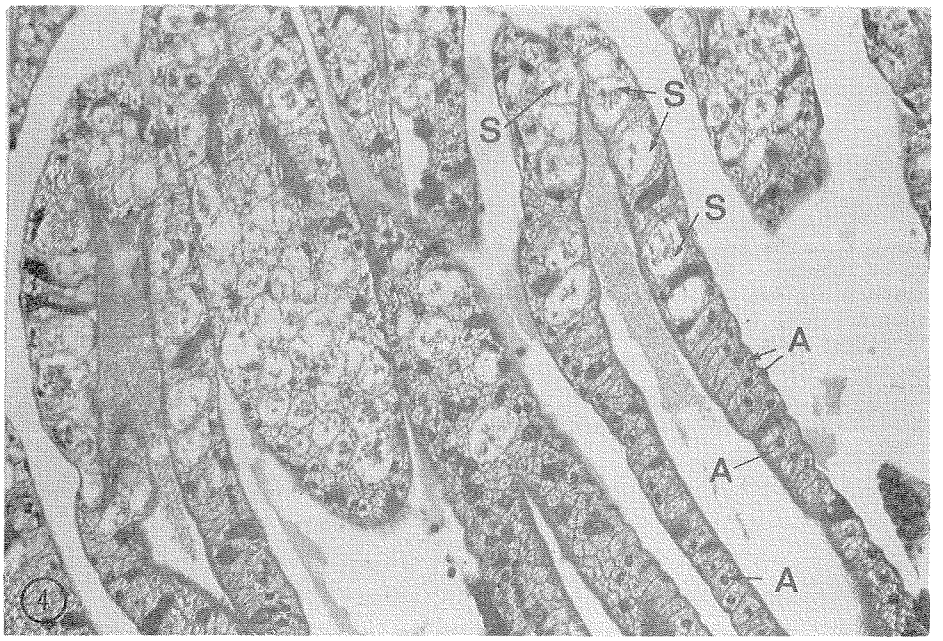
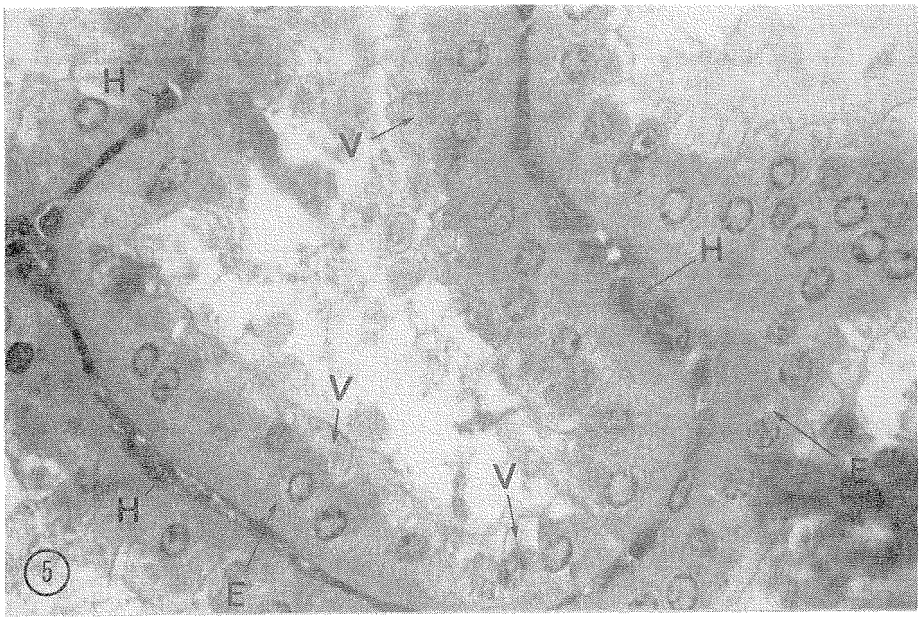

S
S
S
A
A
A
4

Fig. 5. Photomicrograph of green gland labyrinth of lobster #230. The epithelial cells (E) of the tubules are short columnar to cuboidal, with large central nuclei (N). Vacuoles (V) are present at apex of cells, discharging their contents into the lumen of the tubule. Chromatin granules in nuclei are dispersed. Blood cells (H) are present between epithelial cell layers. Cd level of hepatopancreas of this lobster was 16.3 ppm (Helly's fixative, H and E stain; magnification 640x).

Fig. 6. Photomicrograph of green gland labyrinth of lobster #95, Cd level in hepatopancreas 304 ppm. Blood cells (H) are seen between epithelial cell (E) layers of tubules. Normal central nuclei are seen in epithelial cells; some cells are secreting vacuolar contents into lumen. There is no evident membrane or other cellular damage apparent which might have been produced by Cd; magnification 160x.


H
V
H
V
H
V
E
E
5

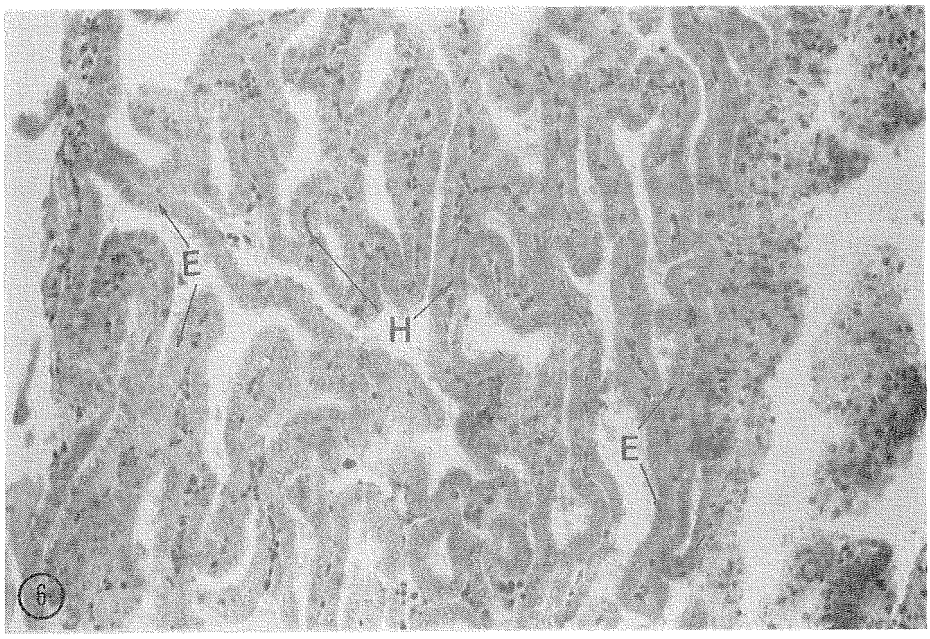
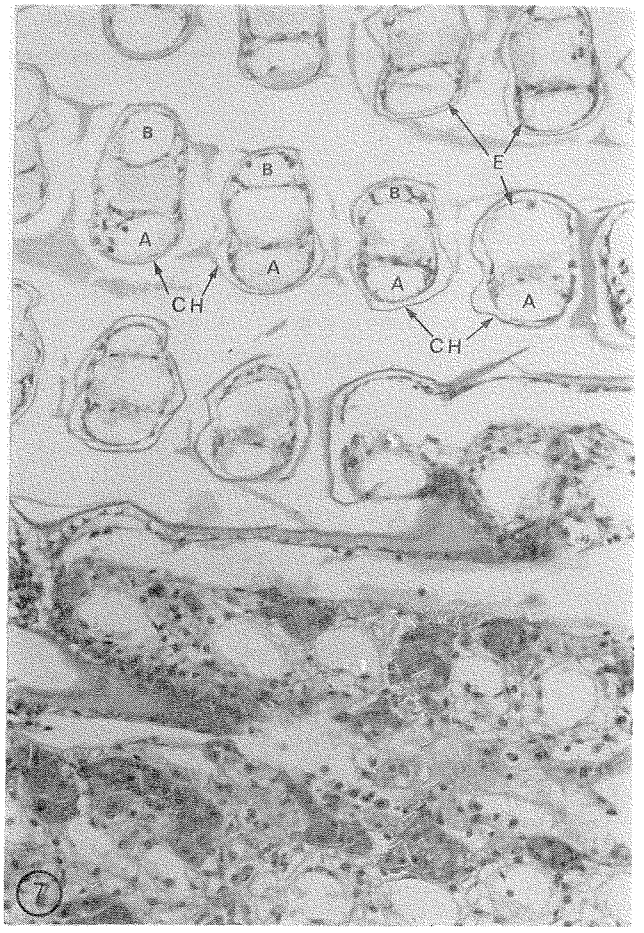

E
H
E
6

Fig. 7. Transverse section of lobster gill. The gill filament is covered with a thin layer of chitin (CH). Below this layer lie two blood vessels, afferent vessel (A) and efferent vessel (B) enclosed by an epithelial layer of cells (E) and separated by a connective tissue septum (CT). This lobster is #250 and contained 12.2 ppm Cd in its hepatopancreas; magnification 160x.


B
B
B
E
A
A
A
A
CH
CH
7