Mercury in the Sediments of the Gulf of St. Lawrence

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In sediments collected from the Saguenay fjord, the St. Lawrence estuary, and open Gulf of St. Lawrence, total mercury varies with sediment texture and location from 10 to 12 300 ppb (average 386 ppb). The highest concentrations occur in the Saguenay fjord (average 2980 ppb) and the lowest in the open Gulf of St. Lawrence (average 150 ppb). The concentration of mercury increases with decreasing grain size, the highest concentrations occur in the fine-grained sediments of the submarine troughs and shelf valleys and the lowest in the sandy shelf sediments. Analyses of the sediments from the Saguenay fjord, where mercury values range from 12 300 ppb at its head to >500 ppb in the lower reaches, indicate that most of the mercury (70 to 90% of the total) is held by the organic matter in the sediments. The distribution of mercury in the fjord is apparently controlled by the downstream dispersal from local industrial sources of mercury-rich organic matter, most likely of terrestrial origin because of its high C/N ratio. In the St. Lawrence estuary where mercury values range from 30 to 950 ppb, and in the open Gulf where correlations between variables are lower and scattered anomalies exist, analyses indicate that mercury accumulates along with the fine-grained inorganic and organic matter in response to the present depositional processes. The distribution of mercury appears to be controlled by the sedimentation pattern. Terrestrial organic matter and industrial waste originating in the Saguenay drainage area have the strongest influence on its distribution.

Dans des sédiments collectionnés dans le fjord du Saguenay, l'estuaire du Saint-Laurent et le Golfe Saint-Laurent, le contenu total en mercure varie de 10 à 12 300 ppb (386 ppb de moyenne), en fonction de la texture des sédiments et de la localisation. Les concentrations les plus élevées sont rencontrées dans le fjord du Saguenay (2980 ppb de moyenne) et les plus faibles dans le Golfe Saint-Laurent (150 ppb de moyenne). Les concentrations en mercure augmentent avec la diminution de la taille des particules: les concentrations les plus élevées se trouvent dans les sédiments à grains fins des dépressions sous marines et des vallées et les concentrations les plus faibles dans les sédiments sablonneux des plateaux. Les analyses des sédiments du fjord du Saguenay, où les teneurs en mercure varient de 12 300 ppb au sommet à > 500 ppb dans ses étendues les plus basses, indiquent que la majorité du mercure (70 à 90% du total) est retenue par la matière organique des sédiments. La distribution du mercure dans le fjord est apparemment controlée par les dispersions apportées, par le courant, des industries locales produisant de la matière organique riche en mercure; le rapport C/N élevé indique que cette matière organique est fort probablement d'origine terrestre. Dans l'estuaire du Saint-Laurent où les teneurs en mercure varient de 30 à 950 ppb, et dans le Golfe où les corrélations entre les variables sont plus faibles et où des anomalies sont enregistrées, les analyses indiquent que le mercure s'accumule avec les fines particules de matière organique et inorganique en réponse au procédé de dépôt actuel. La distribution du mercure nous paraît être controlée par le modèle de sédimentation. La matière organique terrestre et les débris industriels provenant de la zone de drainage du Saguenay exercent les influences les plus fortes sur sa distribution. [Traduit par le journal]

Introduction

High levels of mercury have been found in commercial fish from the Great Lakes, St. Lawrence River and estuary, and Saguenay fjord (Bligh 1970, 1972; Tam and Armstrong 1972). Although some of the mercury is from natural sources, most of it is believed to come initially from industrial sources. The main industrial sources of mercury appear to be the chloralkali plants and pulp mills situated in these areas (Fimreite 1970; Trites 1971). High levels of mercury, apparently derived from industrial sources, are also found in the sediments of the Great Lakes (Thomas 1972, 1973, 1974). Very little is known, however, about mercury levels in the St. Lawrence estuary, Saguenay fjord, and the open Gulf of St. Lawrence. This paper examines the abundance and regional distribution of mercury in these areas and the factors that govern the dispersal of this element. The study is part of a series of geochemical investigations which are being

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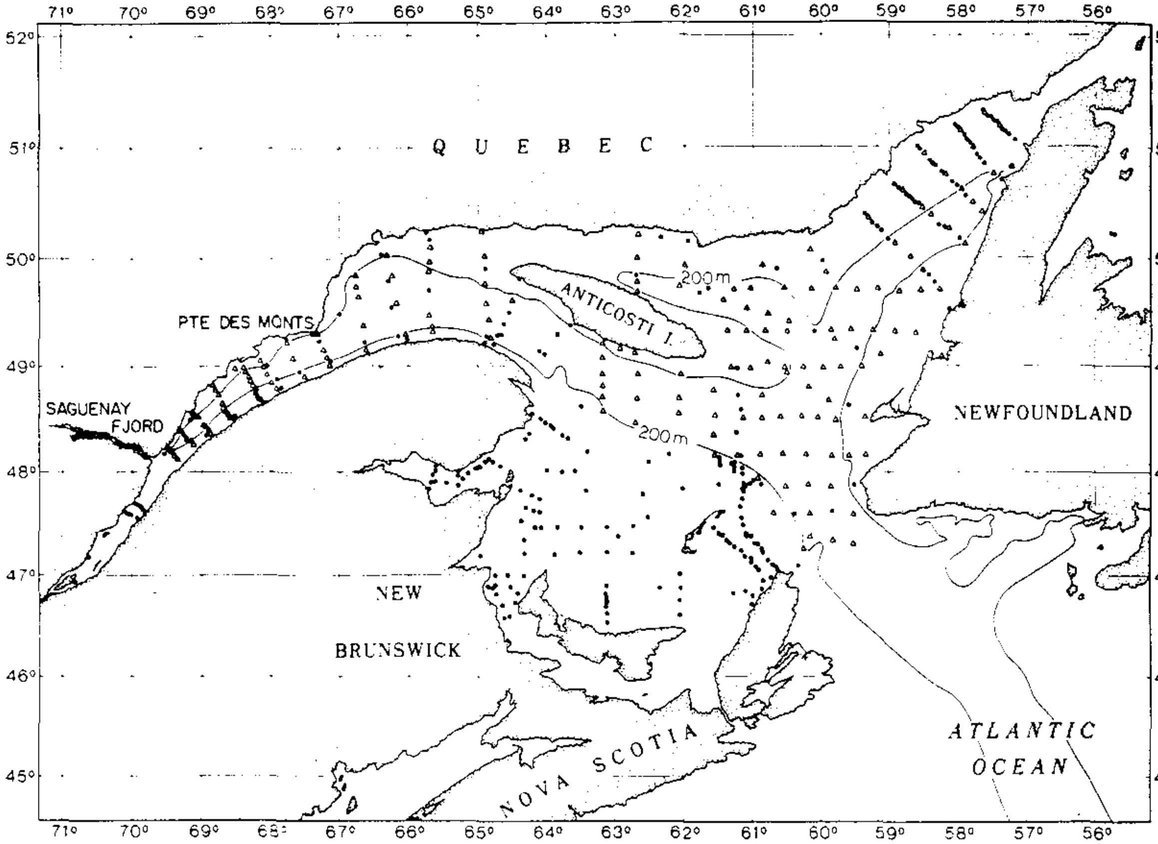

FIG. 1. Location of samples analyzed for mercury in the Gulf of St. Lawrence. Triangles refer to samples for which complete statistical data are available.

carried on by the Marine Ecology Laboratory at the Bedford Institute of Oceanography.

Field and Laboratory Methods

Sediment samples (455) were obtained from different parts of the region with a Van Veen ($0.1 m^2$) grab (Fig. 1). These were collected in 1961 (Cruise S-56 Bedford Institute), 1962 (S-62), 1964 (S-79), 1965 (27-65), (B-65); 1967 (20-67, 24-67), 1971 (71-027), 1972 (72-017), 1973 (73-012), and 1974 (74-006). Core samples were collected from the Saguenay fjord in 1973 (73-012) and 1974 (74-006) using a Ewing type piston corer with a plastic liner (1.5 in. (3.8 cm) diameter). The grab samples were placed in plastic bags or vials, stored in wax containers, and refrigerated or frozen on board until returned to the laboratory. Suspended matter samples were collected and their concentrations determined by Sundby (1974).

In the laboratory, representative portions of the samples were air dried and stored in air-tight bottles until used for sedimentological and chemical analyses. Sedimentological analyses of subsamples were carried out by the standard combined sieve pipette method described by Nota and Loring (1964). After removal of sea salts, chemical analyses of subsamples for the oxides of Si, Al, Ti, Fe, Mn, Ca, Mg, and K were made by colorimetric methods similar to those described by Riley (1958). After 1966, these methods were replaced by atomic absorption techniques, described by Loring and Nota (1973), using a Perkin-Elmer Model 303 atomic absorption spectrometer. Acetic acid soluble iron and manganese were also determined by atomic absorption (Loring and Nota 1968).

Initial mercury analyses were begun in 1970 using the samples stored in air-tight glass bottles. Samples collected in 1972–74 were kept frozen and analyzed as soon as possible after each cruise. Mercury was determined in duplicate for each sample by digestion in 1:2 concentrated $HNO_3:H_2SO_4$ using the cold vapour atomic absorption method similar to that described by Hatch and Otto (1968). Replicate analyses

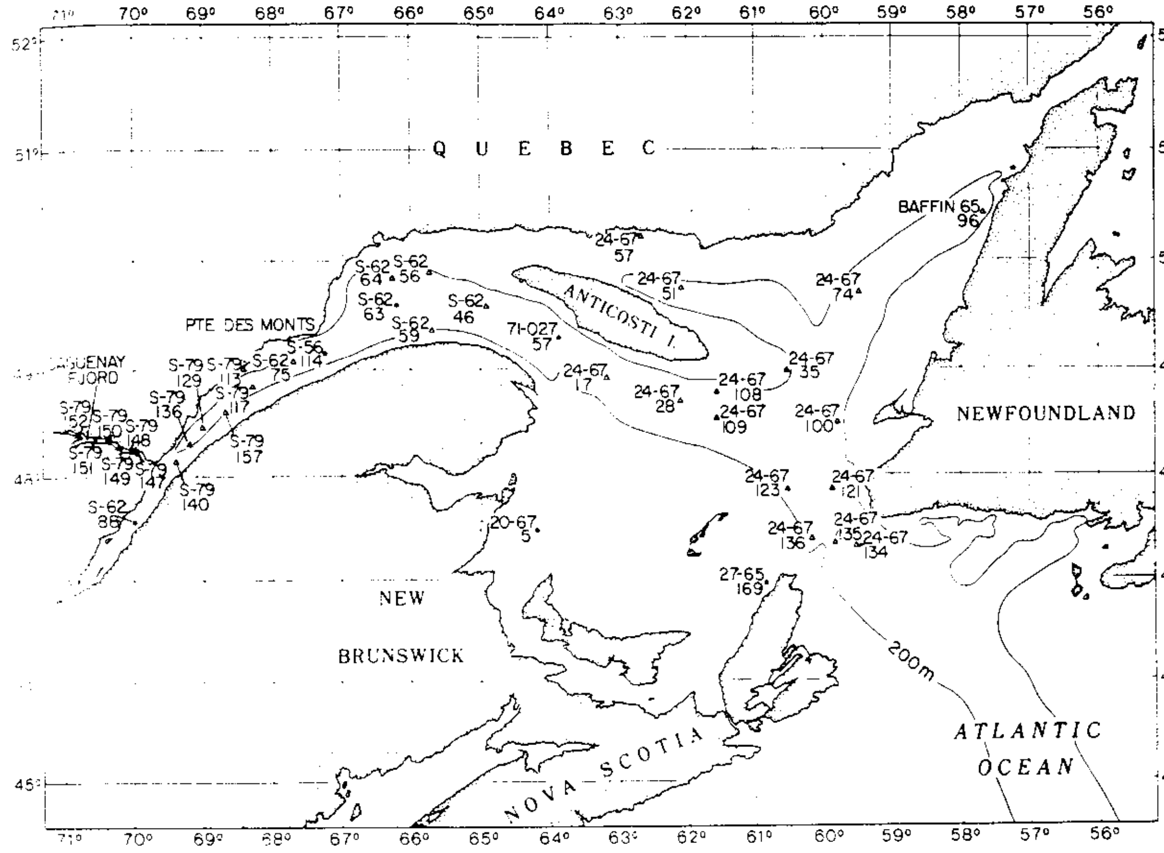

FIG. 2. Location of samples referred to in the text.

[illegible] a sample with an average of 180 ppb [illegible] standard deviation of ±23 ppb (coefficient of variation of 12.8%). At a level of 410 ppb the standard deviation of replicate analyses (6) was ±30 ppb, coefficient of variation of 7.3%. At a level of 9.5 ppm, the standard deviation of replicate analysis (12) was ±0.95 (coefficient of variation of 10%). Accuracy of the analysis was established by the use of the rock standards G-1 and W-1 for which we obtained values within ±10% of the value reported for these standards.

Given the volatility of mercury, the validity of analyzing for mercury in samples stored up to nine years is, of course, open to question. This was recognized at the beginning of the investigation in 1970. It was believed, however, that the results would at least indicate the relative abundance of mercury in the sediments and reveal the salient features of any regional distribution pattern that might exist. Besides, the loss of mercury during storage would give low values, *i.e.*, conservative results. After the initial results were plotted, it was evident that a regional distribution pattern existed and that mercury anomalies were present in the Saguenay fjord and the St. Lawrence estuary. On the basis of this, new sampling programs were designed to verify the previous results and to extend the coverage. Thus, the initial distribution pattern of mercury in the Saguenay fjord was based on five samples obtained in 1964 and analyzed in 1971. Since then 27 additional samples were collected and analyzed immediately after each cruise. These have verified the original distribution and more clearly defined its boundaries (Fig. 8). Consequently, the old values together with the new values could be used with confidence to outline the regional distribution of mercury in the sediments (Fig. 7, 8 and 9).

Additional determination on selected samples (Fig. 2) were made to investigate the partition of mercury between the individual size grades (500–53 μm, 53–2 μm and <2 μm). These size fractions were separated and afterwards dispersed in distilled water by wet sieving material >53 μm and by settling in an Attenburg sedimentation cylinder. The partition of mercury between the inorganic and organic phases of the sediments was also studied.

A selected sample from the Saguenay River, whose average total Hg content was 9.6 ppm, was first treated with an acetic-acid solution (25% v/v). The acetic acid treatment is used to remove elements held in carbonates, easily soluble amorphous compounds of Mn and Fe, and/or loosely held in ion exchange positions. This treatment leaves the lattice structure of the silicate minerals intact and does not attack the resistant iron and manganese oxides such as hematitite, ilmenite, pyrite, and hausmannite (McKeaque and Day 1966; Heintze and Mann 1951; Hirst and Nicholls 1958). No mercury was detected in the acetic acid extract, and no significant loss of mercury was detected in the acid insoluble residue within the precision of the method. This implies that little or no mercury is loosely held to the surfaces of the sediment particles or associated with acid soluble Fe and Mn grain coating, or carbonate in these sediments because this treatment usually removes significant amounts of metallic elements. Next, the same sample was treated with a cold 10% solution of H_2O_2 and 90% of the total mercury in the sample dissolved. This treatment is usually used to remove metallic elements from within the organic fraction of sediments (Angino and Billings 1967). Some of the reduced forms of iron in the sediment may be partly oxidized by the peroxide and so release some mercury adsorbed on particles of iron monosulphide (FeS) in the sediments but apparently none of the insoluble HgS in the sediment. After the H_2O_2 treatment, the sediment residue was analyzed to determine the absolute concentration of mercury in the detrital (H_2O_2 insoluble) fraction. The detrital contribution to the total Hg level was then calculated, and the nondetrital contribution was obtained by difference. From this value, the concentration of Hg in the H_2O_2 soluble phase was calculated (absolute concentration Hg × % nondetrital fraction equals the nondetrital contribution).

Readily oxidizable organic matter in the sediments was determined using the wet oxidation method (cold H_2SO_4 and chromic acid) described by Walkey (1947). The leaching and the use of Ag_2SO_4 in the digestion mixture nullified the effect of chloride. The results (organic carbon) were converted to organic carbon matter by using a conversion factor of 1.72.

The data have been analyzed by statistical techniques similar to those described by Spencer and Degens (1968) and Cameron (1968, 1969). Correlation matrices and *R* mode factor analysis have been used to clarify the relationships between the distribution of mercury and the textural and chemical characteristics of the sediments. Variables are grouped on the basis of their degree of intercorrelation into factors that are associations of highly correlated variables. The factors, which are statistically the most dominant features of the data variations, are derived from the correlation matrices. In this study the principal factor matrices were rotated to produce varimax orthogonal solutions.

Geological and Geochemical Framework

The Gulf of St. Lawrence is an inland sea covering about 250 000 km^2 that is connected to the Atlantic Ocean by the Strait of Belle Isle and Cabot Strait and to the urban and industrial heartland of Canada by the St. Lawrence River and estuary (Fig. 3). It is bounded to the north by the highland edge of the Canadian Shield. In the west, the St. Lawrence lowlands, which are underlain by sedimentary rocks, border the St. Lawrence River and estuary and extend seaward with remnants appearing as islands and narrow coastal lowlands. The south side of the lower estuary and the southern and eastern margins of the Gulf are enclosed by the highlands, uplands and lowlands of the Appalachian region of Canada and are developed on mixtures of crystalline and sedimentary rocks.

A number of important physical and chemical factors contribute to the present sedimentary environment in the Gulf of St. Lawrence (Loring and Nota 1973). Dissolved and suspended inorganic matter in fresh water is supplied to the Gulf from numerous rivers and from rain and snow. Two-thirds (~367 000 ft^3/s (10 276 m^3/s)) of this water is supplied by the St. Lawrence River which extends inland for almost 2000 miles (3219 km) and has a drainage area of almost half a million square miles (~125 000 000 km^2) (Trites, 1971). The Gulf is a marine body of water with salinity ranging up to 35‰ and contains small amounts of suspended matter. The

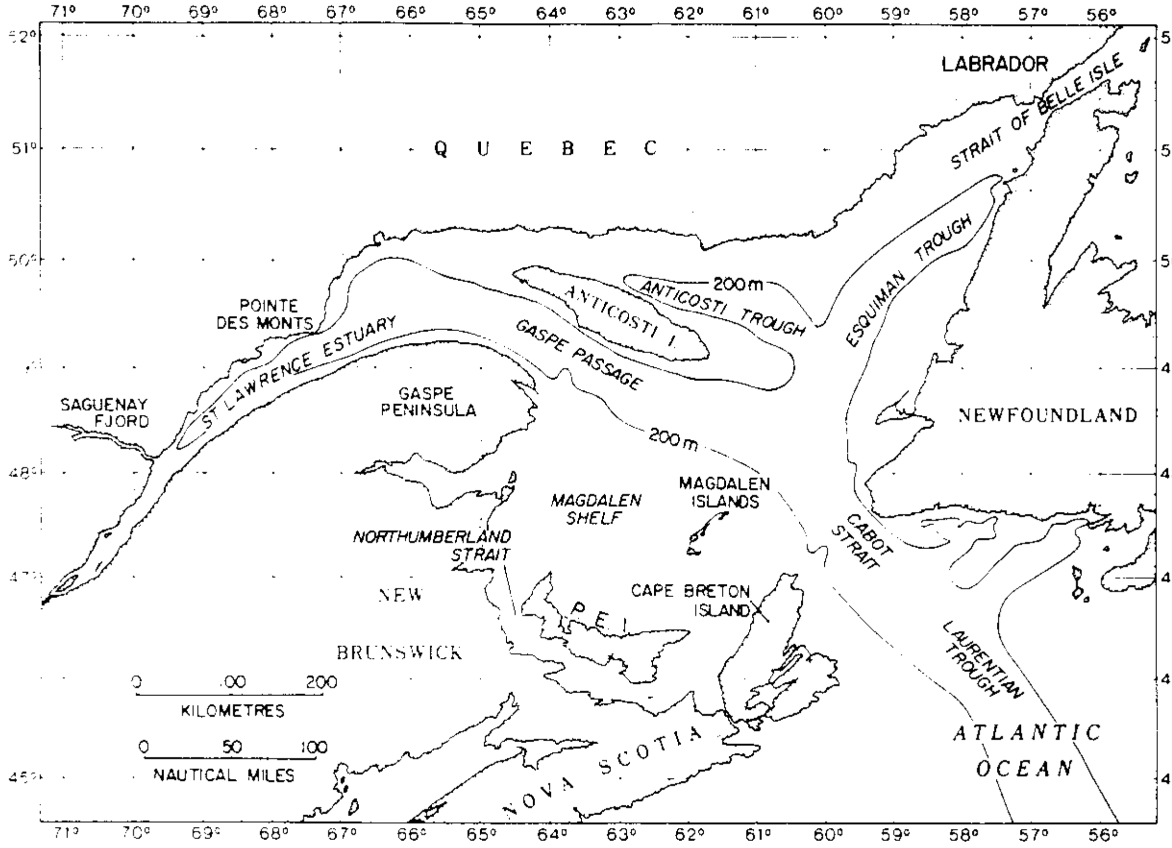

FIG. 3. Place names and general bathymetry of the Gulf of St. Lawrence.

eanographic conditions are characterized by at three water masses of differing temperatures rface, cold intermediate, and warm bottom layers) and salinities as well as a complex estuarine type of circulation (tidal and nontidal) patterns (Trites 1971). In addition, the Gulf has an ice cover in winter.

The floor of the Gulf is composed of long trough-shaped valleys and shelves of varying widths and relief (Fig. 3). The Laurentian channel or trough is 200 to 500 m deep, about 40 km long, and about 37 to 93 km wide. It extends from a position at the mouth of the Saguenay River in the St. Lawrence estuary through the Gulf to the edge of the continental shelf. The trough has three main tributary valleys: the Saguenay, the Anticosti, and the Esquiman. The fjord-like valley occupied by the Saguenay River joins the head of the main trough discordantly. It is a long (~92 km) narrow (1 to 6 km) valley with straight high rock walls from over 300 m above to 274 m below sea level. Elongated deep basins as deep as 274 m, separated by shallow (20–150 m) sills, occur along its entire length (Fig. 4). Southwest of Anticosti Island, the main trough is joined by the Anticosti trough or channel 200 to 300 m deep, which enters from the northwest, and the Esquiman trough, 200 to 300 m deep, which enters from the northeast. The submarine platform shelves adjacent to the trough, with water depths less than 200 m, vary in width and relief. The largest of these is the Magdalen Shelf, which occupies the southern Gulf.

Fine-grained sediments referred to as muds or pelites (material <0.05 mm) and calcareous muds or calcipelites (if containing $>5\%$ $CaCO_3$) cover the deep central parts of the major troughs as well as some of the shelf valleys and vary in thickness from 3 to 40 m (Fig. 5). These sediments were deposited from suspension in response to the present depositional conditions. They are very soft and have a water content of about 60% by weight; a few millimetres below the sediment water interface, they are dark greenish-grey, whereas at the interface

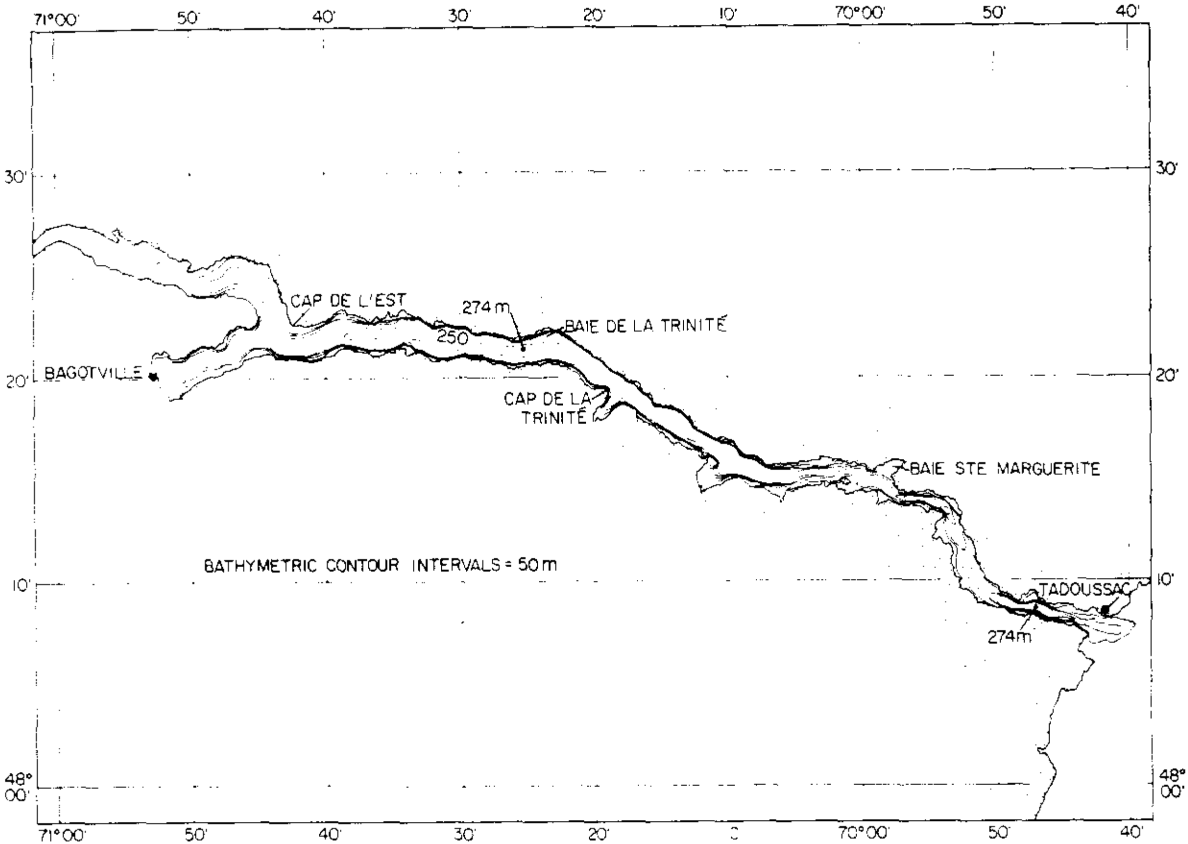

FIG. 4. Bathymetry of the Saguenay fjord contoured from data derived from C.H.S. Chart 1225.

they are dark yellowish-brown. Oxidizing conditions prevail at this interface and below it reducing conditions prevail. The clay content is usually between 45 to 65% by weight and the silt content ranges from 30 to 50% by weight. In the calcareous muds, the carbonate content varies from 5 to 20%. Material coarser than 50 μm generally amounts to a few percent and increases as these sediments merge into coarse sediments (pelitic sands, etc.). On the upper slopes of the troughs, various types of sand and gravels and their calcareous equivalents occupy most of shelf and bank areas. These sediments are reworked, resorted and redistributed glacial deposits. In the Saguenay fjord (Fig. 6), black anoxic sandy (5–10%) muds occur in the upper arms of the fjord. These merge seaward into dark greenish-grey muds that occupy the deep inner basin. Downstream the sediments become coarser grained with very sandy (>30%) muds occurring in the basins and muddy sands covering the broad sills between them. Sand and gravel cover the floor in the basin at the mouth of the fjord.

These sediments may be regarded as a mixture of inorganic and organic material that has been deposited as solid particles or has been incorporated into them from solution. The sand and silt size material (2 to 0.002 mm) is composed of rock fragments, feldspar, quartz, accessory heavy minerals, whereas the clay fraction is composed of quartz, feldspar, amphiboles (rock flour) and the clay minerals, illite and chlorite. The geochemistry of the major elements indicates that Si, Al, Ti, K, Na, Mg, Ca, and to a lesser extent Fe and Mn, have entered the depositional basin structurally combined in detrital minerals. (Loring and Nota 1973.) The textural and regional variations in major element concentrations are determined by the nature, abundance, grain size and provenance of the host minerals of these elements. Most of the major elements, therefore, accumulate at the same rate as detrital sedimentary material although studies show that significant quantities of Fe (3 to 16% of the total) and Mn (5 to 60% of the total) have also entered as oxide grain coating on suspended matter (Loring and Nota 1968). The organic fraction varies from 0.12 to 5.79% by weight. The highest concentrations

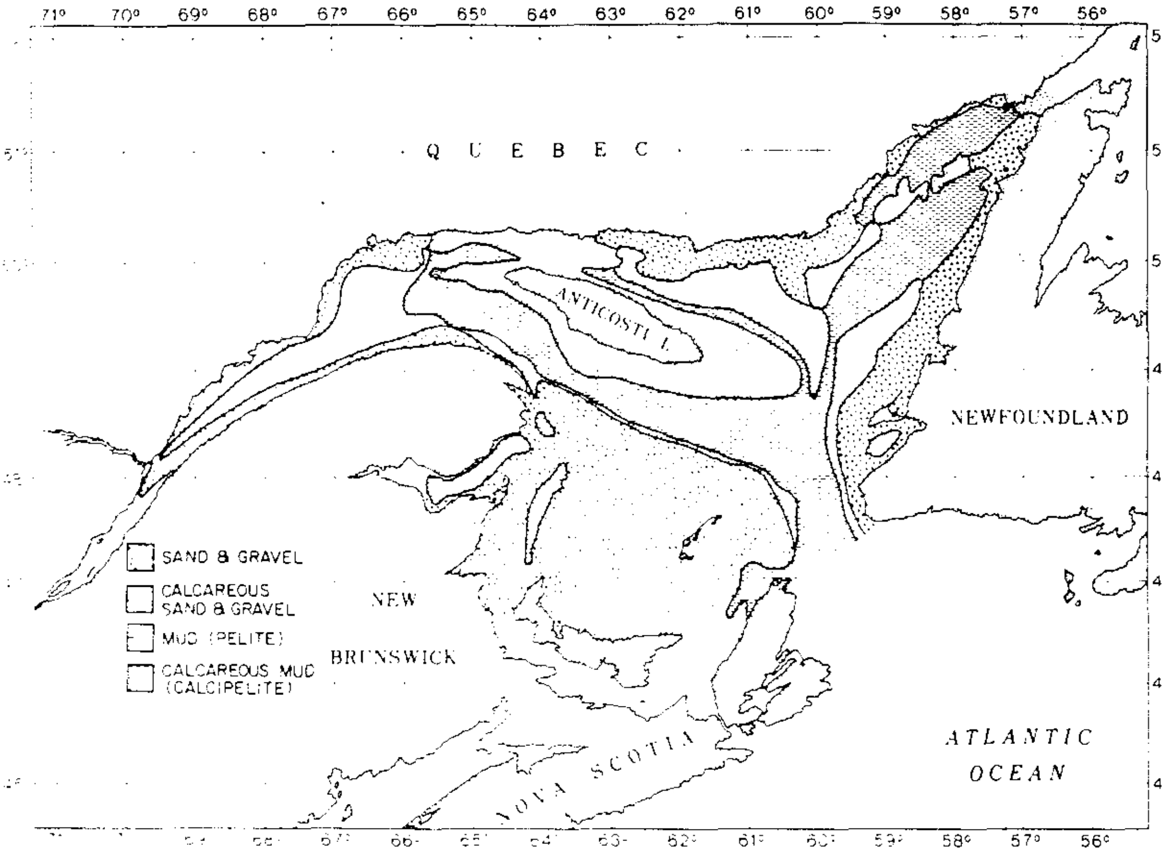

FIG. 5. Simplified sediment map of the Gulf of St. Lawrence (modified from C.H.S. Chart 811G).

occur along with the fine-grained sediments in deep parts of the submarine troughs and valleys and the lowest values occur in the dy shelf sediments. Analysis of individual size fractions from different parts of the Gulf shows that the organic matter content increases with decreasing grain size and the highest concentrations occur in the clay size fraction. An important exception, however, is found in the Saguenay fjord sediments where organic matter occurs in the sand size fraction as well as in the finer sediments. This is believed to be due to wood fibers in the samples (Marlowe 1970; Loring and Nota 1973). Organic matter accumulates at the same rate as the inorganic matter, except near pulp mills; the greatest accumulations of both take place in the troughs in response to the present depositional conditions.

Results and Discussion

Abundance and Distribution of Mercury

Mercury contents vary from 10 to 12 300 ppb and average 386 ppb in the Gulf of St. Lawrence sediments (Table 1). This average value is higher by a factor of 2 to 13 than the average values reported for different types of igneous, metamorphic and sedimentary rocks by other workers. Since these rock types are similar to those found in the land areas adjacent to the Gulf, some enrichment of Hg in the Gulf sediments is indicated relative to their source rocks. Compared to sediments from the Great Lakes the average Hg content of the St. Lawrence sediments is higher by 164 ppb than the average Hg content of Lake Huron sediments but lower by 265 ppb than the average value reported for Lake Ontario. These differences are undoubtedly due to the high amounts of industrially derived Hg in the Lake Ontario sediments (Thomas 1972) compared to more natural levels of Hg in the Lake Huron sediments (Thomas 1973). The average Hg content of the Gulf sediments is 286 ppb higher than in oceanic sediments. These average values are misleading because the distribution pattern of Hg and the variation in Hg content in texturally equivalent sediments clearly shows that mercury enrichment does occur in certain parts of the Gulf of St. Lawrence.

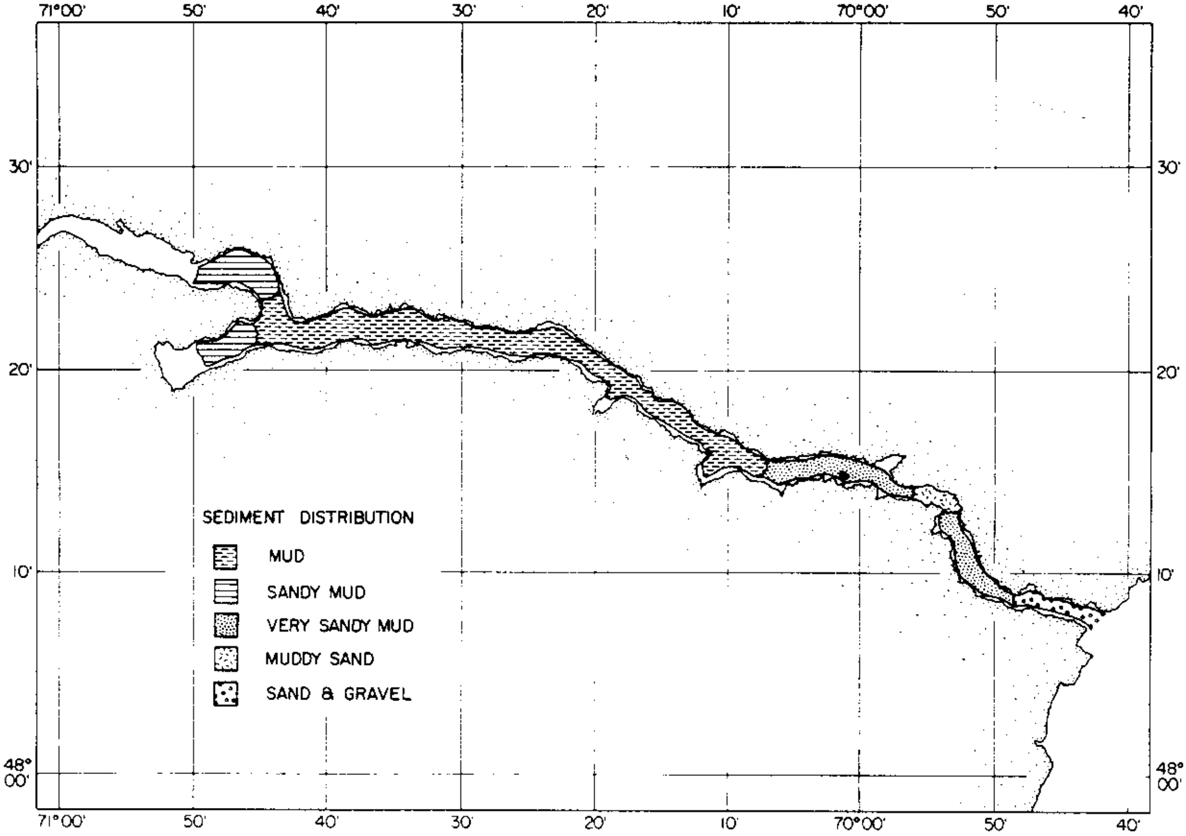

FIG. 6. Surface sediment map of the Saguenay fjord.

TABLE 1. Hg contents (ppb) of the St. Lawrence sediments and those of other rocks and sediments

	No. of samples	Average	Range
St. Lawrence sediments	455	386	10–12 300
Acid intrusive rocks[1]		62	7–200
Intrusive basic rocks[1]		28	5–84
Ultra basic rocks[1]		168	5–250
Schists[1]		100	10–1000
Gneisses[1]		50	25–100
Sandstones[1]		55	10–300
Shales[1]		67	5–300
Limestones[1]		40	< 10–220
Lake Huron sediments[2]	163	222	54–805
Sandy inshore sediments	89	171	54–655
Fine-grained basin sediments	74	283	63–805
Lake Ontario sediments[3]	248	651	32–2100
Fine-grained basin sediments	122	997	220–2100
Nearshore sandy sediments	126	335	32–1820
Oceanic sediments[1]		100	10–2000

[1] Jonasson and Boyle (1971).
[2] Thomas (1973).
[3] Thomas (1972).

TABLE 2. Mercury contents (ppb) of the different sediment types in the Saguenay Fjord, St. Lawrence Estuary, and open Gulf of St. Lawrence

	No. of samples	Average	Range
Saguenay fjord	32[1]	2980	160–12 300
Sands	7[2]	369	160–920
Muds	13[3]	3659	600–9500
St. Lawrence estuary	75[1]	370	30–950
Sands	20[2]	242	120–490
Muds	29[3]	449	150–860
Open Gulf of St. Lawrence	348[1]	150	10–750
Sands	64[2]	128	30–330
Muds	72[3]	221	50–600

[1] Total number of samples analyzed for Hg.

[2] All samples containing > 30% sand for which all statistical data are available.

[3] All samples containing > 30% mud for which all statistical data are available. These do not include samples containing > 30% sand size material (very sandy muds).

Table 2 shows that mercury concentrations vary both with the location and texture of the sediments. The highest concentrations occur in the Saguenay fjord and the estuary and decrease on the whole in the open Gulf. High concentrations of Hg are found in the muds occupying the submarine troughs and shelf valleys, and the low concentrations are usually associated with the sandy shelf sediments.

On the average, the sands from the open Gulf are lower in Hg than those in Lake Huron and Lake Ontario whereas the sands from the

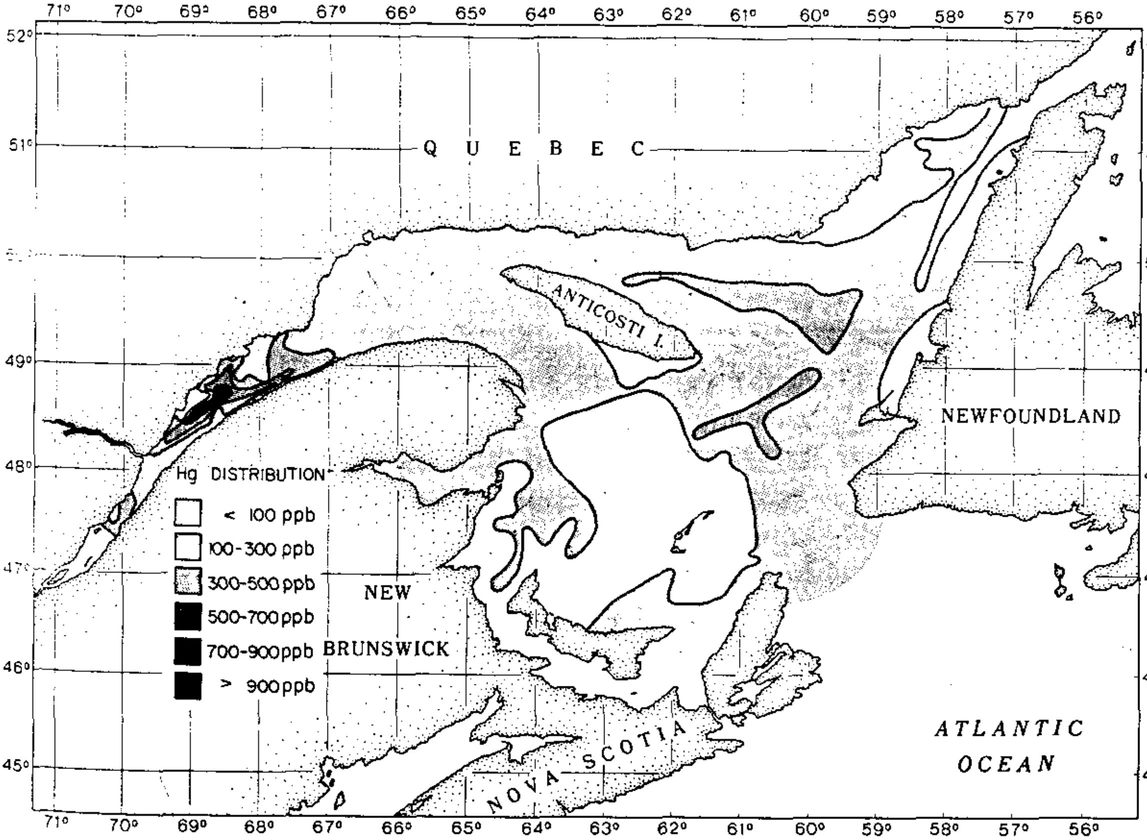

FIG. 7. Distribution of total mercury in the surface sediments in the Gulf of St. Lawrence (1000 ppb = 1 ppm).

St. Lawrence estuary and the Saguenay fjord contain amounts of Hg comparable to those reported for Lake Huron and Lake Ontario respectively (Table 1). The Saguenay muds, however, are much higher in mercury than those from Lake Ontario, which contain high amounts of industrially derived Hg. The St. Lawrence muds are lower in Hg than those from Lake Ontario, but higher in Hg than the Lake Huron muds. In contrast muds from the open Gulf contain amounts of mercury comparable to Lake Huron muds but contain much less mercury than Lake Ontario muds. Mercury, therefore, appears to be enriched in the sediments of the Saguenay fjord and to a lesser extent in those from St. Lawrence estuary, but not in the open Gulf sediments in particular or in the average sediment as a whole.

Distinctive geographic variations occur within different parts of the area (Fig. 7). In the Saguenay fjord, total mercury concentrations decrease from 12 300 ppb at its head to less than 500 ppb in its lower reaches. This suggests a major input of mercury from a local source near the head of the fjord (Fig. 8). In the estuary, mercury concentrations are lower (Fig. 9). They range from 30 to 950 ppb and increase with decreasing grain size of the sediments so that the highest concentrations occur in fine-grained muds that occupy the center of the estuary. This pattern does not suggest a local source but rather a close relationship with the deposition of fine-grained sediments in the estuary. In the open Gulf, where most of the sediments contain less than 300 ppb, there is not a clear distribution pattern for mercury and there are only a few anomalies (300 to 500 ppb), one of which occupies the Anticosti trough and another of which is found in a small part of the Laurentian trough southeast of Anticosti Island (Fig. 7).

Relationship of Mercury to Texture

The relationship between Hg content and sediment texture and between Hg content and geographic location of the sediments is confirmed by the analyses of individual size fractions of texturally different sediments from various

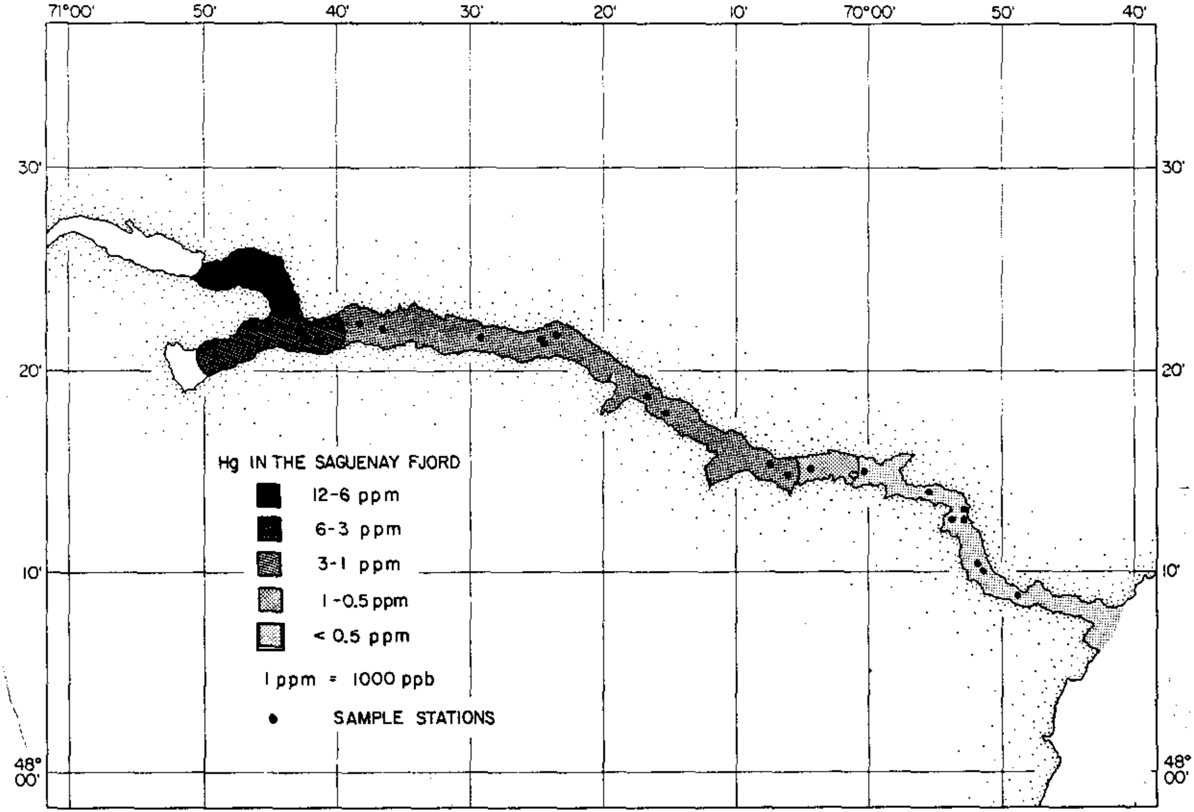

FIG. 8. Distribution of total mercury (ppm) in the Saguenay fjord.

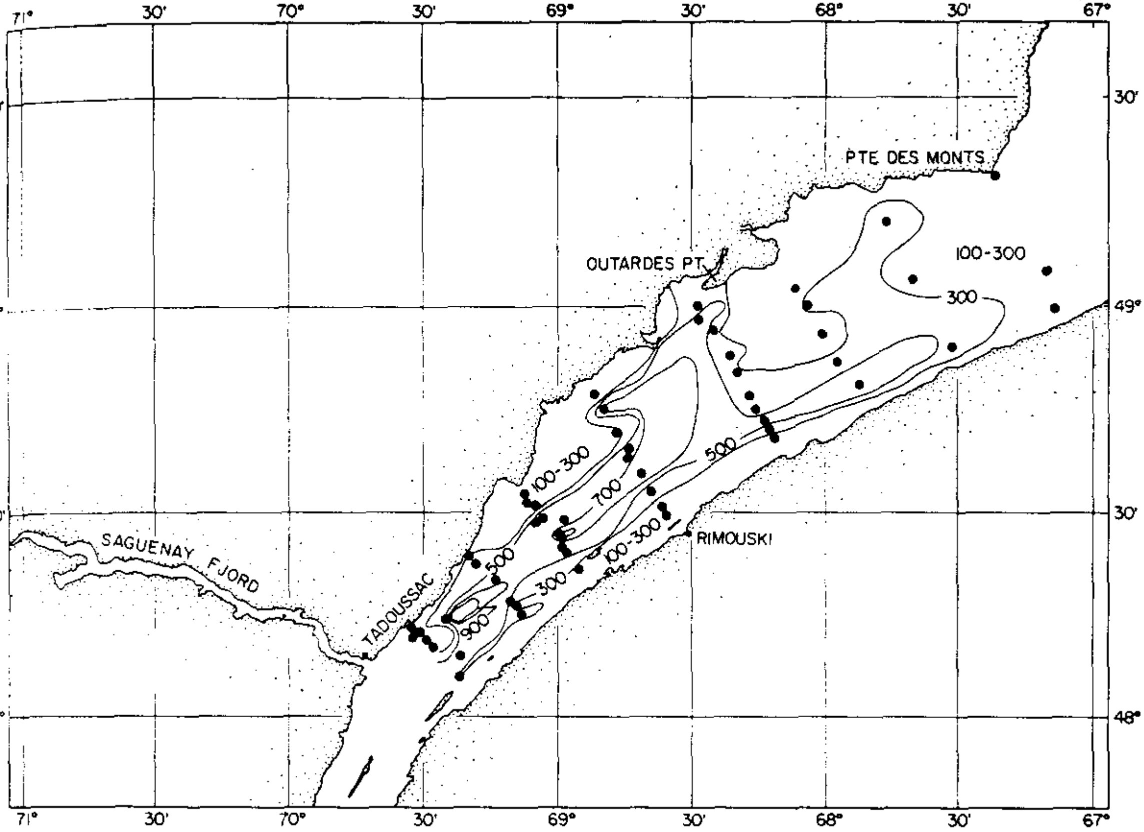

FIG. 9. Detailed distribution of total mercury (ppb) in the surface sediments of the St. Lawrence estuary.

locations. Within each sample the absolute Hg content of each fraction increases with decreasing grain size and the highest concentrations occur in the clay size fraction. The total Hg content depends on the Hg content of each of the individual size fractions and on the contribution that each fraction makes to the total sediment (Table 3). The sands are usually low in mercury because of the relatively low concentration of Hg in the sand fraction (500–53 μm) and the high proportion (30–95%) of this fraction in sediments of this type; similarly, muds are usually higher in mercury because of their high content of fine-grained material ($<53\ \mu$m in diameter), which contain higher amounts of Hg. There is also a wide variation in the Hg content within each size fraction of different samples. Since grain size is obviously not responsible for these variations, compositional changes in the material within the same size fractions must account for these wide variations in the Hg contents of texturally equivalent sediments from different parts of the Gulf.

Chemical Partition

Table 4 shows the results of the simple extraction procedure with hydrogen peroxide (H_2O_2). The results from texturally equivalent sediments from different parts of the Gulf show that: (1) between 45 and 90% of the total Hg in the sediment was removed by the H_2O_2 treatment; (2) the absolute amount of Hg in this fraction varies from 300 to 70 420 ppb; (3) the contribution that this fraction makes to the total mercury content of any one sample depends on the amount of the H_2O_2 soluble fraction in the sediment as well as on the absolute amount of Hg in that fraction; and (4) the absolute amount of Hg in the soluble fraction varies regionally with the highest concentrations occurring in samples from the head of the Saguenay fjord (Stations 152 and 151).

These results suggest that a large part of the total Hg is associated with the organic matter in the sediment. There is also a regional variation in the amount of mercury held in the soluble fraction that might be attributed to differences in the nature of the organic material in sediments from different parts of the Gulf.

Geochemistry of Mercury

The geochemistry of mercury has been documented and reviewed by Jernelöv (1968, 1969), Löfroth (1969), Jonasson (1970), Fleischer (1970), Jonasson and Boyle (1971), and Thomas (1972, 1973, 1974). Briefly, mercury derived from natural and man-made sources migrates in a variety of dissolved and particulate forms. Dissolved forms are usually removed rapidly from solution either by adsorption onto, or complex formation with, finely divided suspended inorganic and organic particles. In the bottom sediments, Hg is usually retained by these constituents especially in reducing environments where the presence of H_2S from decaying organic matter allows some Hg to dissociate and be precipitated as a rather insoluble sulphide (HgS) or be adsorbed onto iron sulphide surfaces. In such an environment inorganic mercury may be methylated into the highly toxic methyl mercury (CH_3Hg^+) by bacterial action and escapes from the bottom sediments. Consequently most workers have found that Hg is held in the organic fraction, in sorbed positions, in iron oxide grain coatings, or by sulphides in sediments.

TABLE 5. Correlation matrix, Saguenay River sediments ($n = 20$)

	Hg (total)	Sand	Mud
Depth	—	—	—
Sand	−0.55	1.00	−1.00
Mud	0.55	−1.00	1.00
C/N ratio	0.73	—	—
Organic matter[1]	0.84	−0.88	0.88
CO_2	—	−0.57	0.57
Si	—	0.91	−0.91
Al	—	—	—
Ti	—	—	—
Fe	—	−0.73	0.73
Ca	—	—	—
Mg	—	−0.96	0.96
Mn	—	—	—
Fe (nondetrital)	—	−0.68	0.68

For $r > \pm 0.54$, $P < 0.01$.
[1] Organic carbon matter.

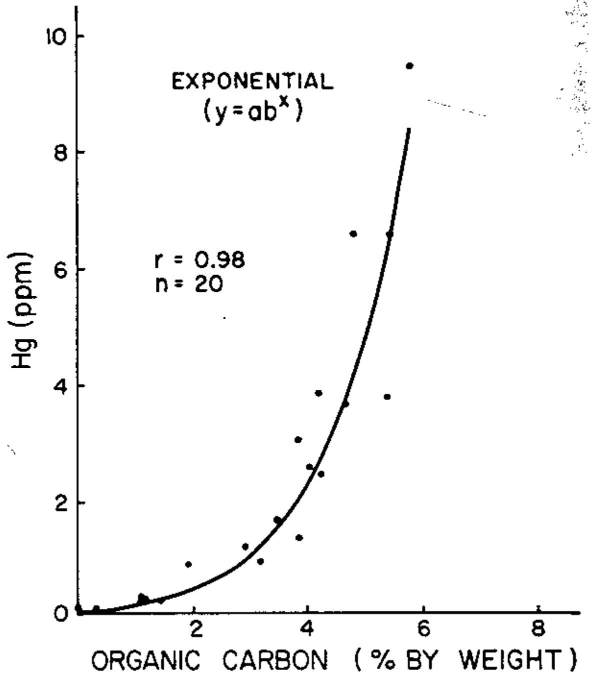

FIG. 10. Relationship between total Hg and organic carbon matter in the Saguenay fjord sediments.

The statistical relationships of mercury to the other chemical and physical properties of the sediments are shown in Tables 5–10. These data are used to determine the source and location of mercury in the Saguenay fjord, the estuary and the open Gulf.

Saguenay Fjord

Mercury concentrations are very high in parts of the fjord (Fig. 8). The correlation coefficient matrix is given in Table 5. It is based on the analyses of 20 sediment samples taken along the length of the fjord (Fig. 8). From the table it can be seen that the mercury strongly correlates at better than the 99% confidence level with organic carbon, C/N ratio, and less strongly with mud. Mercury is also negatively correlated (−0.55) with sand.

The strong correlation of total Hg to organic carbon matter shows that the concentration of Hg is very dependent on the amount of organic matter (Fig. 10). It suggests that most of the mercury is associated with the organic matter. This is confirmed by the fact that most of the mercury (70 to 90%) is extracted by the H_2O_2 treatment (Table 4).

Total mercury also varies with the C/N ratio. Figure 11 shows that total Hg increases with increasing C/N ratios and the highest Hg con-

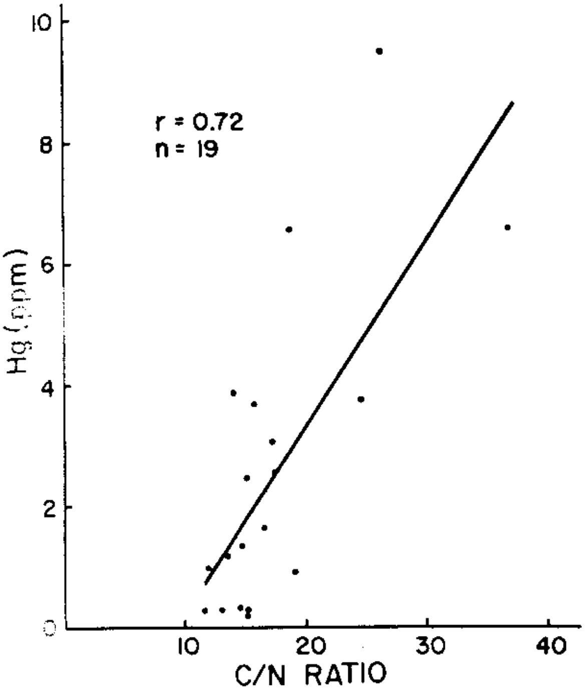

FIG. 11. Relationship between total mercury and C/N ratio (by weight) in the Saguenay fjord sediments.

centrations occur in the sediments having the highest C/N ratio. Regionally, the highest Hg values and C/N ratios are found in the organic-rich sediments at the head of the fjord. This suggests that the amount of Hg depends not only on the quantity but also on the type of organic matter. A similar strong correlation between Hg and organic carbon as well as nitrogen has been noted by Armstrong *et al.* (1972) in Hg-contaminated lake sediments from northern Ontario. It is well known that the C/N ratio of sediments varies with the type of organic matter in them (Arrhenius 1950). Pocklington (1973) has shown that the high C/N ratios in the sediments that contain large amounts of organic matter at the head of the fjord are due to an increase in the amount of organic carbon and not to a decrease in nitrogen. Hence, the high C/N ratios are attributed to high amounts of land-derived organic plant matter whereas the sediments that contain both land- and marine-derived organic matter have lower C/N ratios and the ratio decreases as the quantity of marine plant and animal remains increase. From this it is concluded that Hg-rich organic matter is derived mainly from a terrestrial source and is most likely represented by the wood fibers observed in the sediments. This was confirmed by Pocklington and MacGregor (1973); they showed that the organic-rich sediments of high C/N ratios also contain high concentrations of lignin, one of the characteristic components of terrestrial plant material. Figure 12 shows that total Hg varies directly ($r > 0.95$, $P < 0.001$) with the amount of lignin, expressed as spruce wood, in the samples with the highest concentration occurring in the organic-rich sediment from the head of the fjord.

In the fjord, therefore, most of the mercury is either adsorbed by the organic component of the sediment or forms a complex with it. Some of this mercury may be associated with sulphur-bearing organic compounds. It also may have dissociated in the sediments to form an independent sulphide or be adsorbed on iron sulphides due to the reducing environment created below the sediment–water interface by the decaying organic matter. Some evidence for this is given by the analyses for sulphide sulphur in six samples taken along the length of the fjord at about 20 km intervals. The results show that sulphur varies directly with amount of total Hg ($r = 0.95$), organic carbon ($r = 0.90$), and the C/N ratio ($r = 0.95$).

Some mercury is also undoubtedly methylated by bacterial action in the sediment under anaerobic conditions and escapes into the

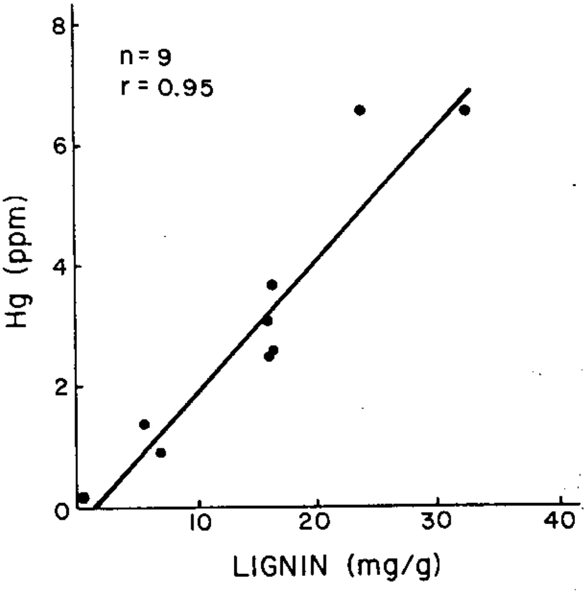

FIG. 12. Relationship between total mercury and lignin (expressed as spruce wood equivalent by Pocklington 1973) in the Saguenay fjord sediments.

TABLE 6. Varimax matrix,[1] Saguenay River sediments ($n = 20$)

	Factor	
	1	2
Hg	0.90	—
Depth	—	0.71
Sand	—	−0.80
Mud	—	0.80
C/N ratio	0.90	—
Organic matter	0.81	—
CO_2	—	—
Si	—	−0.81
Al	—	—
Ti	—	—
Fe	—	0.67
Ca	—	−0.70
Mg	—	0.82
Mn	—	—
Fe (nondetrital)	—	0.73

For values > ±0.54.

[1] Full varimax matrix (6 factors) accounts for 90.7% of total problem variance.

water column. This may account in part for the high levels of mercury in fish from the fjord (Tam and Armstrong 1972). Mercury may also be lost to the water column by the formation of soluble organometallic complexes produced by the decay of terrestrial organic matter (De Groot *et al.* 1971; Lindberg and Harriss 1974).

In the varimax matrix given in Table 6, the variables are grouped on the basis of their degree of intercorrelation as established from the correlation matrix. The matrix shows that Factor 1 accounts for most of the variance of total Hg, C/N ratio, and organic carbon in the sediments. This factor, therefore, represents the dispersal of Hg-rich organic material of terrestrial origin. In contrast, Factor 2 accounts for most of the variance of sample depth, sand, mud, and the inorganic chemical constituents, Si, total Fe, nondetrital Fe, Ca, and Mg. Factor 2 with its high negative loadings on sand and positive loadings on mud as well as their inorganic chemical constituents is a sedimentation factor. It most likely represents the physical process responsible for the sediment distribution in the fjord. The absence of any significant loading on mercury, C/N ratio and organic matter in this factor also implies that Hg does not accumulate at the same rates as the natural detrital sedimentary material but rather at a rate determined by the supply of Hg and terrestrial organic matter. The dispersal of terrestrial organic material, therefore, becomes the overriding control on the distribution of mercury in the fjord. Consequently, the downstream dispersal of mercury and terrestrial organic matter indicates that there has been, at least until recently, a major input of mercury along with land-derived organic plant matter near the head of the fjord from a local point source. This input may be due to direct injection of mercury from industrial outfalls such as a chloralkali plant or as a byproduct of paper mills in the vicinity. The enrichment of Hg in the sediment appears to be a relatively recent phenomenon. Mercury analyses of samples taken at 1 cm intervals from a 70 cm core obtained in the north arm at the upper end of the fjord in 1974 show that the high Hg values (1–11 ppm) are confined to the top 38 cm below which values they drop to about 0.25 ppm. This indicates that 75–98% of the total mercury at the sediment surface is industrially derived in this location. Seven other cores taken at intervals of 3 to 12 km show that the mercury-enriched layer decreases downstream from 38 cm to 1 cm over 60 km. Mercury-rich material is probably still accumulating as suspended matter samples throughout the water column have been found to contain from 2 to 50 ppm of mercury.

St. Lawrence Estuary

In the estuary, the factors that control the distribution of mercury are less evident than in the Saguenay fjord. The correlation matrix, which is based on 49 samples, shows that the Hg correlates strongly ($r > 0.5$) with mud, organic carbon, nondetrital iron and less strongly ($r = 0.35$ to 0.5), with CO_2, total iron, and Mg (Table 7). It also shows that Hg is negatively correlated with sand, Si, and detrital Ca.

The correlation of Hg with sand, mud and their inorganic constituents reflects the increase of Hg with decreasing grade size of the sediments in the estuary as well as with the individual size fraction of individual samples (Table 3).

The correlation of Hg with organic matter suggests that most of the Hg is associated with the organic fraction of the fine-grained sediments. This is confirmed by the H_2O_2 extractions (Table 4) which show that between 56

TABLE 7. Correlation matrix,[1] St. Lawrence Estuary ($n = 49$)

	Hg
Depth	—
Sand	−0.50
Mud	0.55
Organic matter	0.55
CO_2	0.38
Si	−0.50
Al	—
Ti	—
Fe	0.37
Ca[2]	−0.44
Mg	0.36
Mn	—
Fe (nondetrital)	0.53

[1] For $r > \pm 0.35$, $P < 0.01$.
[2] Detrital Ca.

and 73% of the total mercury is held by the organic fraction. Since the concentration of organic carbon matter increases with decreasing grain size, the relationship between Hg and texture is probably secondary, *i.e.* the correlation between Hg and texture is due to organic matter being common to both. Fine-grained suspended matter undoubtedly plays an important role in providing a large surface area for the initial adsorption of mercury from solution with organic matter being a greater collector of mercury as well as a suitable medium for its retention in sediments.

Since the C/N ratios and lignin contents have not been measured for all the estuarine sediments included in the statistical analyses, it is not possible to assess fully the relationship of Hg to these variables. Carbon and nitrogen have been measured, however, in 14 samples (Pocklington 1973). In them, the C/N ratio varies from 10 to 16. Hence most of the organic matter in these sediments is of marine origin and is comparable to the type of organic matter found in the lower reaches of the Saguenay which contain less than 2 ppm Hg. In the estuary, however, the total mercury is not significantly correlated ($r = -0.04$) with the C/N ratio. This suggests that mercury is not associated with the dominant type of organic matter in the estuarine sediments. It also implies that Hg is not greatly enriched in organic matter derived from the marine plant and animal remains, either in the estuarine sediments or in the Saguenay.

Lignin concentrations measured in four samples vary from 0.6 to 3.3 mg/g in the estuary below the mouth of the Saguenay River. These are comparable to those found in the lower Saguenay (0.03 to 1.9 mg/g) but are much lower than the concentrations in the sediments from the head of the fjord (4 to 9 mg/g). Total mercury correlates strongly ($r = 0.97$, $P < 0.001$) with the lignin content of these samples. This supports previous conclusions that mercury is closely associated with the terrestrial plant component of the organic matter. It is supported by the strong correlation ($r = 0.89$, $P < 0.001$) of Hg with lignin found in both the Saguenay and estuarine samples (Fig. 13). In contrast, a sediment sample (Station 053/72) taken from the floor of the estuary above the mouth of the Saguenay contains a relatively high lignin concentration (1.2 mg/g) but only a low mercury content (180 ppb). Although more analyses are required, the data indicate that the Hg-enriched terrestrial component of the organic matter is derived mainly from the outflow of the Saguenay River and not that of the St. Lawrence River.

In the inorganic fraction, Hg appears to be correlated with nondetrital iron and total iron. This implies that some Hg is associated with the iron oxide grain coating found in the estuarine sediments (Loring and Nota 1968). Such an association has been shown in the Lake Ontario sediments by Thomas (1972). In the sediments, nondetrital Fe and total Fe and manganese are correlated positively with mud and organic carbon, and negatively with sand, and the inorganic constituents of these components. This

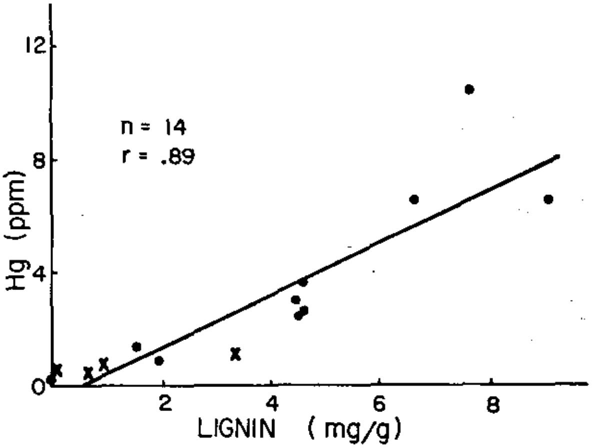

FIG. 13. Relationship between total mercury (ppm) and lignin (mg/g) in the Saguenay fjord sediments (●) and the St. Lawrence estuary sediments (x).

TABLE 8. Varimax matrix[1], St. Lawrence Estuary (n = 49)

	Factor 1
Hg	0.66
Depth	—
Sand	−0.72
Mud	0.73
Organic matter	0.79
CO_2	0.80
Si	−0.76
Al	—
Ti	—
Fe	0.37
Ca	−0.71
Mg	0.40
Mn	—
Fe (nondetrital)	0.88

For values > ± 0.35.
[1] Full varimax matrix (6 factors) accounts for 75.5% of total problem variance.

association is coincidental with the increase of total Hg, nondetrital Fe, and total Fe with decreasing grain-size fractions rather than with a real relationship between Hg and these elements. This is confirmed by the results of the weak acid extractions used to release major and trace elements from the sediments (Loring and Nota 1973). The results show that no significant amounts of Hg are released by the weak acid attack. The unextracted mercury accounts for 27 to 44% of the total in the analyzed samples, and therefore must be present as H_2O_2 insoluble components either as sulphides or is structurally combined in the rock-forming minerals found in the sediments.

The varimax matrix shows that Factor 1 accounts for most of the variance of mercury in the sediments (Table 8). This factor with its high loadings on mud and organic carbon, as well as most of the inorganic elemental constituents and negative loadings on sand, is easily identified as the sedimentation factor. It represents the physical process responsible for the present distribution of sedimentary material in the estuary. From this it is concluded that the distribution of Hg in the estuary is determined by the sedimentation pattern, and that Hg-enriched terrestrial organic matter derived from the Saguenay outflow accumulates along with the fine-grained material in the center of the estuary in response to the present depositional conditions. The seaward decrease in mercury concentrations along the axis of the estuary is therefore attributed to the seawa decrease in the concentration of land-derive organic matter noted by Pocklington (1973 This does not imply, however, that all th mercury is derived from the Saguenay Rive outflow, but rather the anomalous concentrations of Hg represent the enrichment of H from this source (above their natural levels in the estuarine sediments.

Gulf of St. Lawrence

The correlations of Hg with other variables are not as strong in the open Gulf sediments where mercury concentrations are usually low (<300 ppb) and approach natural levels. The correlation matrix (Table 9) shows low but significant (r = ±0.218, $P < 0.01$) positive correlations of Hg with depth, mud, organic carbon, Al, Ti, total Fe, K, and total Mn. Mercury is also negatively correlated with sand and carbonate (expressed as CO_2). Thus, these are similar but much lower correlations than those found for mercury in the estuary. The lower degree of correlation between the variables is attributed to the lower mercury values in the open Gulf sediments as compared to the estuarine sediments, and the large number of samples used in the statistical analysis, as well as the wide degree of variability of Hg within texturally equivalent sediments and individual size grades. A similar difference in the level of the correlation coefficients between the Lake Ontario sediments, which contain high Hg, and Lake Huron sediments, which contain low Hg, has been noted by Thomas (1973).

TABLE 9. Correlation matrix[1], Gulf Sediments (n = 136)

	Hg
Depth	0.24
Sand	−0.25
Mud	0.25
Organic matter	0.27
CO_2	−0.25
Si	—
Al	0.35
Ti	0.22
Fe	0.34
K	0.30
Ca	—
Mg	—
Mn	0.33
Fe (nondetrital)	—

[1] For r > ±0.218, $P < 0.01$.

The relationship observed in the open Gulf sediments re-affirms that mercury is mainly held by the fine-grained inorganic and organic fraction of the sediments. Some indication that organic matter is one of the most important sites for mercury is given by the H_2O_2 analyses of a few samples. The analyses show that between 44 and 88% of the total mercury is removed by this treatment and presumably represents Hg mainly associated with H_2O_2 soluble organic components. Carbon and nitrogen analysis of nine samples from the Gaspé passage and the Anticosti trough by Pocklington (1973) show that the C/N ratio is less than 11 (5.8 to 10.4). This indicates that most of the organic matter is of marine origin. Although only two lignin analyses are available from these locations, they show that no detectable lignin is present in the Gaspé passage where a mercury value of 190 ppb was recorded and at a site in the Anticosti trough where 1.63 mg/g of lignin was present along with 150 ppb mercury. There is, therefore, no obvious indication that mercury is related to terrestrial organic matter in open Gulf sediments. It appears that the very low levels of mercury associated with the organic matter represent that which might be expected to be adsorbed and are chelated with it from solution or concentrated in the marine plants and animals before their death and the deposition of their remains.

In the inorganic fraction, the remaining mercury (12 to 56% of the total) is most likely to be present as a sulphide, a site favored for small quantities of mercury in sedimentary rocks by Marowsky and Wedgepohl (1971) or structurally combined in low concentrations in the detrital silicate minerals found in the sediments (Fleischer 1970). The correlation of Hg with total Fe and Mn may also indicate some adsorption of Hg by iron and Mn oxides that coat the individual sediment grains (Loring and Nota 1968). The presence of Hg in these grain coatings, however, has not been confirmed to date by our analyses.

The correlation matrix (Table 9) indicates that sand and carbonate dilute the mercury. The negative correlation of Hg with sand in this matrix as well as the others represents the decrease of Hg with increasing amounts of sand size material. This is due to the low concentrations of mercury in quartz, the predominant mineral of sand size, and aluminosilicate minerals of this size. The diluent effects of carbonate on the Hg concentrations are more difficult to assess because they affect all size fractions and because the carbonate is detrital limestone particles derived from local sources within the Gulf (Loring and Nota 1973). Since the calcareous material, which has little Hg, is found in all size fractions, it appears that it also acts as a diluent for fine-grained material being deposited from suspension in the troughs, the greatest effect is on the sedimentary material accumulating near to the local sources of calcareous material such as Anticosti Island. This would account in part for the very low Hg values in the sediments in the Laurentian trough adjacent to Anticosti Island and the generally lower average values in the calcareous muds (177 ppb) compared to the average values in the muds (218 ppb) from the open Gulf.

TABLE 10. Varimax matrix[1], Gulf Sediments (n = 136)

	Factor	
	1	10
Hg	0.22	0.93
Depth	0.85	—
Sand	−0.83	—
Mud	0.83	—
Organic Matter	0.83	—
CO_2	—	—
Si	−0.41	—
Al	0.61	—
Ti	0.49	—
Fe	0.76	—
K	0.48	—
Ca	—	—
Mg	0.62	—
Mn	0.44	—
Fe (nondetrital)	—	—

For values > ±0.21.

[1] Full varimax matrix (10 factors) accounts for 85.3% of total problem variance.

The varimax matrix (Table 10) shows that two factors (1 and 10) account for most of the variance of Hg. Factor 1 with its high positive loading on depth, mud, organic carbon, Al, Ti, total Fe, K, Mg, and Mn, and high negative loadings on sand and Si, is obviously the sedimentation factor. Mercury is positively loaded on this factor at the same significant level that is related to other variables in the correlation matrix. This suggests that the overall distribution of mercury is loosely controlled by the sedimentation pattern and that mercury accumulates along with the fine-grained inorganic and organic matter in the deep waters of the Gulf in response to the present depositional conditions. In contrast, Factor 10, with its high positive loadings on mercury, is apparently the most important statistical factor. It is a statistically independent factor, however, because it does not contain any significant loadings on any other of the variables included in the analysis. This is not surprising in view of the low, but significant, correlations between Hg and other variables. It is interpreted as indicating that mercury at or near natural levels found in its source rocks (Table 1) is unequally distributed at low levels amongst the various sediment components rather than being predominantly concentrated in high amounts in one fraction, *i.e.* such as was found in the Saguenay fjord sediments. At low levels there is considerable 'noise' due both to low concentrations and the analytical variability in its measurement. It is believed that this would cause an apparent randomness in the mercury distribution and, when correlated with the other variables, that the factor analysis would record this as independent statistical behavior. This does not imply that there is a fundamental difference between the sedimentation process governing the Hg distribution in the Gulf and estuary, but rather that the overall part played by this process, and the components with which mercury is mainly associated within the sediments, becomes more obvious as the concentration of mercury increases above natural background levels in the estuary as well as in the Saguenay fjord.

Conclusions

Mercury is enriched in the marine sediments of the Gulf of St. Lawrence relative to their source rocks and the greatest enrichment occurs in the Saguenay fjord and the St. Lawrence estuary. Some of the mercury is from natural sources, but high concentrations can be directly attributed to industrial mercury pollution at the head of the Saguenay fjord. It appears that terrestrial organic matter, most likely wood fibres, is the major carrier of the industrially derived mercury. This material has been dispersed downstream from its source and has been deposited from suspension along with the fine-grained sediments accumulating in the fjord. Although mercury concentrations decrease rapidly away from their source, small but significant amounts of mercury-rich organic matter have apparently escaped from the Saguenay into the St. Lawrence estuary where they are accumulating along with, and are retained by, the fine-grained sediments in the center of the estuary in response to the present depositional conditions. There is no evidence to indicate that industrially-derived mercury has reached the open Gulf of St. Lawrence where mercury content appears to be at or near the natural levels found in the source rocks and Hg is accumulating at the same rates as detrital sedimentary material. This is not to say that the industrially-derived mercury will not in time reach the open Gulf, for the same process of sedimentation controlling the distribution of mercury in the St. Lawrence estuary also controls the deposition of natural mercury in the open Gulf.

It appears therefore that industrially-derived mercury along with the natural mercury from the outflow of the Saguenay together with mercury from the St. Lawrence River is transported into the marine environment where it is deposited from suspension along with other fine-grained material in response to the present depositional conditions. The amount of industrially-derived mercury in the sediment depends on the distance from source and the physical conditions of transport and deposition.

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