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**METALLIC AND ORGANIC CONTAMINANTS IN SEDIMENTS OF THE  
ST. CROIX ESTUARY AND PASSAMAQUODDY BAY**

By

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**ABSTRACT**

D. H. Loring, T.G. Milligan, D.E. Willis and K.S. Saunders. 1998. *Metallic and Organic Contaminants in Sediments of the St. Croix Estuary and Passamaquoddy Bay*. Can. Tech. Rep. Fish. Aquat. Sci. 2245: v + 44 p.

This report provides an assessment of the sedimentology and baseline levels of metallic (As, B, Ba, Be, Cd, Cr, Co, Cu, Mn, Mo, Ni, Pb, Sb, Se, Sr, Tl, U, V and Zn) and organic (PCBs and PAHs) contaminants in bottom sediments from the St. Croix estuary and the adjacent Passamaquoddy Bay on the south-western coast of New Brunswick. It also provides some insight into the anthropogenic and natural factors that control the distribution of these contaminants. Granulometric analyses indicates a high degree of floc settling and a lack of resuspension for the Passamaquoddy Bay sediments that might make this area unsuitable for salmon aquaculture. Geochemical metal analyses show that the highest concentrations of the metals occur in fine-grained sediments below the mouth of the St Croix river and the lowest in the muddy sands in the outer part of Letang inlet and the St Croix estuary. Most of the metals are at, or near, natural concentrations but 20% and 5% of the samples (n=19) exceeded the contamination threshold ( $>20$  and  $>40 \text{ mg}\cdot\text{kg}^{-1}$ ) levels for As and Ni concentrations respectively. Normalization of the metal data suggests anthropogenic inputs of Cd and perhaps Pb and Zn from the St. Croix river although the amounts have not yet exceeded contamination levels. Organic analyses indicate that measurable PCB residues occur in only one sample. Measurable PAH residues ( $67\text{-}3144 \text{ ng}\cdot\text{g}^{-1}$ ) were found in all analysed samples (n=13) and vary in concentration with the amount of material  $<63\mu\text{m}$ . Residue patterns for the contaminants surveyed are consistent with hydrocarbon residue levels from combustion and ship traffic and/or municipal sources. Although these data represent a one time sampling, the possibility of increasing contaminant concentrations in the sediment exists. The build-up of contaminants in sediment over time may have a detrimental effect on activities such as aquaculture in Passamaquoddy Bay, and, to a lesser extent, in the St. Croix estuary.

**RÉSUMÉ**

D. H. Loring, T.G. Milligan, D.E. Willis and K.S. Saunders. 1998. *Metallic and Organic Contaminants in Sediments of the St. Croix Estuary and Pasamaquoddy Bay*. Can. Tech. Rep. Fish. Aquat. Sci. 2245: v + 44 p.

Nous avons évalué la sédimentologie et les niveaux de fond de contaminants métalliques (As, B, Ba, Be, Cd, Cr, Co, Cu, Mn, Mo, Ni, Pb, Sb, Se, Sr, Tl, U, V et Zn) et organiques (PCB et HAP) dans les sédiments de l'estuaire de la St. Croix et aux alentours, dans la baie Passamaquoddy, sur la côte sud-ouest du Nouveau-Brunswick. Notre étude apporte aussi quelques indications sur les

facteurs naturels et anthropiques qui régissent la distribution de ces contaminants. Les analyses granulométriques indiquent un fort degré de floculation et de sédimentation et une absence de remise en suspension des sédiments de la baie Passamaquoddy, ce qui pourrait rendre la région impropre à la salmoniculture. Les analyses géochimiques des métaux montrent que les concentrations les plus fortes se retrouvent dans les sédiments fins présents en aval de l'embouchure de la St. Croix, et les plus faibles dans les sables vaseux de la portion extérieure du bras Letang et de l'estuaire de la St. Croix. La plupart des métaux sont en concentrations proches des concentrations naturelles, mais 20 % et 5 % respectivement des échantillons (n=19) dépassaient le seuil de contamination ( $>20$  et  $>40 \text{ mg}\cdot\text{kg}^{-1}$ ) pour As et Ni. La normalisation des données sur les métaux permet de penser qu'il y a des apports anthropiques de Cd et peut-être de Pb et de Zn par la St. Croix, mais les quantités n'ont encore jamais dépassé les seuils de contamination. Les analyses organiques indiquent que des résidus mesurables de PCB n'apparaissent que dans un seul échantillon. Des résidus mesurables d'HAP ( $67\text{-}3144 \text{ ng}\cdot\text{g}^{-1}$ ) ont été trouvés dans tous les échantillons analysés (n=13), et leur concentration varie avec la quantité de matériaux de taille supérieure à  $63 \mu\text{m}$ . Les patrons des résidus des contaminants examinés correspondent aux concentrations de résidus d'hydrocarbures provenant de la combustion et du trafic maritime et/ou de sources urbaines. Bien que ces données ne soient tirées que d'un échantillonnage ponctuel, il est possible que les concentrations de contaminants augmentent dans les sédiments. Cette accumulation de contaminants dans les sédiments avec le temps peut avoir un effet néfaste sur les activités comme l'aquaculture dans la baie Passamaquoddy, et, à un degré moindre, dans l'estuaire de la St. Croix.

## **Introduction**

One of the objectives of DFO Science Toxic Chemicals Green Plan Projects is to determine the sources and distribution of sedimentary materials and associated organic, and metallic contaminants (Loring et al, 1996) in Nova Scotian and New Brunswick coastal embayments. Sedimentary contaminants are of special interest both in terms of the protection and rehabilitation of estuaries and harbours, and in the development of aquaculture industries. In many coastal regions adjacent to industrial and urban areas, sediments are the largest repository for, and potential source of, organic and metallic contaminants in the marine environment.

Relatively little is known about the levels and distribution of contaminants in New Brunswick coastal areas especially those receiving urban and industrial waste and those being developed for salmon aquaculture. This report provides an assessment of the sedimentology and baseline levels of organic and metallic contaminants in bottom sediments from the St. Croix estuary and the adjacent Passamaquoddy Bay on the south-western coast of New Brunswick. It also provides some insight into the anthropogenic and natural factors that control the distribution of these contaminants.

### **St. Croix Estuary and Passamaquoddy Bay**

The St. Croix estuary and the adjacent Passamaquoddy Bay on the south-western coast of New Brunswick (N.B.) occupies an area of 172.3 km<sup>2</sup> with a maximum water depth of 67m (Fig. 1). Several large islands viz.: Deer and Campobello occur on the western approaches to the Bay. The St Croix River estuary is a long (16.7 km), narrow (0.1-2.4 km), shallow (water depths of 0.3 to 10 metres) inlet on the western side of the bay. It receives drainage via the St. Croix River which is 121 km long and forms part of the border between New Brunswick and Maine (Me.). The river drainage area (4224.8 km<sup>2</sup>) is mainly wooded with the vegetation developed on a thin mantle of glacial till underlain primarily by Ordovician-Silurian meta-sediments with some outcrops of granite and basalt. A narrow fertile floodplain borders the lower reaches of the river and supports mixed farming and light industry.

### **Potential Sources of Contamination**

Potential contributions of contaminants derive from agricultural activities and villages along the riverbanks, and urban and industrial wastes from the towns of St. Andrews and St. Stephen, N.B., and Calais, Me. Major industries in the past have been timber and cotton mills, shipbuilding operations, soap and axe factories, and candy manufacturing. At present, aquaculture activities primarily occur in Letang inlet on the eastern side of the Bay, and are being introduced elsewhere.

## **Materials and Methods**

### **Sampling**

The locations of the sampling stations (Stns.) are shown in Fig 1. Sediment samples (n=19) were collected with a Van Veen bottom grab from sites having water depths of 1 to 30 metres (m). Subsamples from the top 5 cm of the grab were transferred by plastic spoon to a plastic bag for metal analyses and those for organic analyses were placed in glass containers. Samples were frozen



immediately and returned to the laboratory for chemical and sedimentological analyses. In the laboratory, the samples for chemical analyses were initially stored in plastic vials, homogenised, and oven dried at 110°C. A portion of each dried sample was used for chemical and sedimentological analysis.

### Granulometric Analyses

The amount of material of size <63µm in diameter was determined gravimetrically after wet sieving the sample. The sediment textural classification used is that defined by Nota and Loring (1964) and is based on the proportions of sand (particles >63µm in diameter) and mud (particles <63µm in diameter) in each sample. Sediments containing >95 % weight of material having grain-size >63µm are referred to as **sands**. Those containing >95 % weight of material <63µm are referred to as **muds**. Sediments containing 5-30% by weight of sand or mud are designated **sandy or muddy** respectively. If the two components both represent more than 30% by weight, the finer grained component is indicated by a noun, the other by an adjective with the adverb **very** e.g. **very sandy mud**. For the purposes of this study, sediments containing >70% by weight of material <63µm has been used to determine the metal concentrations in the fine-grained sediments.

### Coulter Counter Analyses

Disaggregated inorganic grain-size (**DIGS**) spectral analyses were determined by electro-resistance particle sizing using the techniques described in Milligan and Kranck (1991). Sample analyses were carried out using a Coulter Multisizer IIE. Subsamples were digested in an excess of 35% H<sub>2</sub>O<sub>2</sub> and suspended in 1% NaCl before disaggregation with a sapphire-tipped sonic probe. Results were plotted as frequency distributions, or frequency spectra, of log<sub>10</sub> equivalent weight % of sediment, determined from the volume in logarithmically equal size classes using a specific gravity of 2.65 kg·m<sup>-3</sup>. The size distribution data has been normalized to the total mass over the size range analyzed. Size classes were 1/5 φ, with diameter doubling every 5 channels for the Multisizer analyses.

### Organic Chemical Analyses

Sediment samples were stored at -15 °C prior to analysis for organic contaminants. To estimate recovery, samples were spiked prior to analysis with <sup>13</sup>C chlorobiphenyls (ULTRA Scientific; Fisher Scientific Canada Ltd.) and deuterated polycyclic aromatic hydrocarbons (Supelco; Sigma-Aldrich Canada Ltd.). Residues of chlorobiphenyls (PCBs, King et al., 1993) and polynuclear aromatic hydrocarbons (PAHs, King et al., 1996) were extracted into hexane from saponified sediment and cleaned up using gel-permeation chromatography (GPC-autoprep, Model 1001) using 1:1 v/v dichloromethane:cyclohexane as solvent. GPC eluates were split into two fractions; one for PAH and the other for PCB analysis. PCB fractions received further cleanup by treatment with sulphuric acid. PCB and PAH fractions were analyzed by capillary gas chromatography (Hewlett Packard Series II, Model 5971B for PCB and Model 5989B for PAH gas chromatographs fitted with an HP-5MS column, 30 m x 0.25mm x 0.25µm). The detector was a Hewlett Packard mass selective detector utilizing selective ion monitoring, in electron impact mode. Operational blanks and one duplicate sample, randomly chosen, were analyzed with each batch of 20 samples. Recovery of <sup>13</sup>C CBs and deuterated PAHs was 88.4 ± 18.5 % (n = 103) and 90.1 ± 19.7 % (n = 64) respectively. Analyte concentrations (ng·g<sup>-1</sup>) are expressed on a dry weight basis (Table 1). PCB and PAH standards were obtained from ULTRA Scientific and Fisher Scientific Canada Ltd. All solvents were pesticide grade (distilled in glass; VWR/Canlab, Ontario Canada).

## **Inorganic Chemical Analyses**

Total Al, As, Ba, Be, B, Cd, Co, Cr, Cu, Fe, Li, Mn, Mo, Ni, Pb, Sb, Se, Sr, Tl, U, V and Zn were determined using Inductively-Coupled Plasma-Mass Spectrometry (ICP-MS). The results are shown in Table 2 except for Be, B, Mo Sb, and Se which were below their detection limits of  $<5\text{mg}\cdot\text{kg}^{-1}$  (B and Be) and  $<2\text{mg}\cdot\text{kg}^{-1}$  (Mo, Sb, and Se). ICP-MS metal determinations were carried out, under contract, by Philip Analytical Services, Halifax, N.S., following HF/HNO<sub>3</sub>/perchloric acid digestion of a 0.5 gm sample. For quality assurance purposes, NRCC certified reference materials were analyzed along with the samples. The relative accuracies for the determinations of elements were within the standard deviations of the certified values for the NRCC sediment reference materials MESS-1 and BCSS-1.

## **Metal Statistical Analyses**

The means, standard deviations and ranges of the concentration were determined for each element in the sediments from Passamaquoddy Bay and the adjacent St. Croix estuary. Correlation matrices (Bonferroni corrected) and factor analyses were used to clarify the relationships between the trace metals (except B, Be, Sb and Se) and textural and geochemical characteristics represented, in some cases, by proxy normalizing elements (Loring and Rantala, 1992). For the factor analyses, the variables (except Tl) are grouped on the basis of their correlation into factors that are associations of highly correlated variables. These factors are statistically the most dominant features of the data variability and are derived from correlation matrices. For this study, the principal factors (correlation matrices) are rotated to produce normal varimax orthogonal solutions.

## **Results and Discussion**

### **Sedimentary Environment**

Granulometric analyses indicate that although very sandy muds are the dominant sediment type, muds occupy Passamaquoddy Bay (Stns. 17-19), and muddy sands occur in Letang inlet (Stns. 3 and 4), and at several sites in the St. Croix estuary (Stns 8-11, and 15).

The disaggregated inorganic grain size (DIGS) of bottom sediments can be used to describe sedimentary conditions (Kranck et al., 1996a, b, Milligan and Loring, 1997). The size distribution of a sediment reflects the depositional conditions under which it was formed and is sensitive to changes in turbulence, particle concentration, and the cohesivity of the particles in suspension ( Milligan and Loring, 1997, Milligan and Hill, 1998). Kranck et al. (1996a, b) and Milligan and Loring (1997) presented a method whereby the DIGS of a bottom sediment can be used to determine its depositional history. Using the model described, the inorganic fraction of sediment can be separated into three components, each of which has a different mode of settling. The components are: 1) material settled in a flocculated form, defined by the straight-line portion of the curve starting at 0.7  $\mu\text{m}$  in the plots and termed the floc tails; 2) material deposited as single grains from suspension which forms the "one-round" modal peak with a slope of 2, occurring at around 10  $\mu\text{m}$ ; 3) material which has undergone further suspension sorting, usually as a result of high-energy events, as defined by the well sorted, "multi-round" peak in the coarsest size range. In any sediment size distribution, one or more of these components will occur. For a particular area, the slope of the flat floc tail is defined by the nature of the source material, and can be used to indicate the origins of the material making up the sediment (Kranck and Milligan, 1985). The modal diameter and degree of sorting

(number of rounds as defined by the steepness of the positive slope of the modal peak) in the bottom sediment is controlled by the energy to which the sediment is exposed.

The location of the sampling stations (n=19) is shown in Fig 1. Log-Log plots of the DIGS are shown in Fig 2. The source slope for the samples collected is similar to that found for the sediments in Letang Inlet (Milligan, 1994), with a mean value of 0.34. The lack of source slope variation suggests there is a common sediment supply for the Letang and Passamaquoddy regions. The size distributions for stations (5-16) along the main channel of the St. Croix River estuary are dominated by a well sorted modal peak in the range of 200-500  $\mu\text{m}$ . This peak, and the lack of floc settled material in the fine grained tail of the distribution, indicates that the sediments deposited in these regions are frequently subjected to high bottom stresses. Very little flocculated material is deposited in these locations. Samples (Stns. 3-4) collected from mid-channel in Letang Harbour, and off Campobello island (Stns. 1 and 2) show similar high-energy size distributions. In contrast, the three stations (17-19) located in Passamaquoddy Bay have modal diameters  $<10\mu\text{m}$  and consist of over 80% floc settled fine-grained material. These size distributions are very similar to those found in the more sheltered regions of Lime Kiln Bay and Bliss Harbour NB (Milligan, 1994). The lack of a coarse modal peak in these sediments, and the one collected off Todd's Point (Stn.5) in the upper St. Croix estuary, indicates that particle dynamics favour aggregation. Little or no resuspension occurs after deposition at these locations.

### Organic Contaminants

No measurable PCB residues were found in any of the samples except at Stn.12 (Fig. 1) At Stn 12, The congeners found were sporadic and few in number ( $\Sigma\text{PCB} <65 \text{ ng}\cdot\text{g}^{-1}$ ), and are not representative of any particular PCB mixture (i.e. Aroclor 1254). The location of this station, essentially opposite the mouth of the channel from St. Andrews Harbour suggests that these congeners might have originated from this area.

Measurable PAH residues ( $67\text{-}3144 \text{ ng}\cdot\text{g}^{-1}$ ) were found in all analysed samples (Table 1, and Fig. 3). Total PAHs vary in concentration with the amount of material  $<63\mu\text{m}$  (Fig. 4). with the highest concentrations occurring in the fine grained sediments. Seven stations, four on the St. Croix River (Stns. 5, 6, 12 and 13) and the three stations (17-19) in Passamaquoddy Bay had total measurable PAH  $> 950 \text{ ng}\cdot\text{g}^{-1}$  for each station, the remaining samples had total PAH residues ranging from 67 to  $478 \text{ ng}\cdot\text{g}^{-1}$  (Fig. 4). Station 5 (Todds Point) located at the merging of the Waweig river, the outflow from Oak Bay and the upper reaches of the St. Croix river had the highest total PAH ( $3144 \text{ ng}\cdot\text{g}^{-1}$ ) The towns of Calais, Me., and St. Stephen, N.B. situated on opposite sides of the river  $\sim 6 \text{ km}$  upstream, probably account for most of the PAH residues detected. Station 6, 1.8km downstream from Stn. 5, has slightly more than half the PAH content of Stn. 5 (Fig.4).

The Mill Cove (Stn. 12) sediment had high total PAH similar in composition to Station 5 (Todds Point), suggesting a similar source. Station 13, 1.8km downstream from Stn. 12, had one half the PAH residues found in the upstream station. This parallels the pattern observed for Stn. 5 (Todds Point) and Stn. 6. The three stations in Passamaquoddy Bay had elevated levels of PAH and the pattern of detected compounds was similar to all other stations. Station 19 had the highest residue levels (closest to St. Andrews) and Stn. 17 had the lowest (furthest from St. Andrews). Total PAH ranged from 960 to  $2200 \text{ ng}\cdot\text{g}^{-1}$  for these three stations with individual compounds accounting for between 10 and  $550 \text{ ng}\cdot\text{g}^{-1}$ .

All of these total PAH levels are within Canadian Ocean Dumping Guidelines, but according to the criteria set by the Norwegian State Pollution Control Authority for Norwegian coastal areas (Molev er et al, 1997), samples from Stns 8, 10, and 11 would be classified as slightly contaminated ( $\Sigma \text{PAH} <300$

ng·g<sup>-1</sup>), samples, those from Stns. 6, 7, 9, 12,13, 15, 17, and 18 as moderately contaminated ( $\Sigma$  PAH 300-2000 ng·g<sup>-1</sup>), and those from Stns 5 and 19 as markedly contaminated ( $\Sigma$  PAH 2000-6000 ng·g<sup>-1</sup>). Residue patterns for the contaminants surveyed are consistent with hydrocarbon residue levels from combustion and ship traffic/municipal sources (Pruell et al., 1990). The ratios between phenanthrene, fluoranthene, pyrene, benz[a]anthracene, chrysene and benzo[a]pyrene are similar for all samples and the lack of any methylated derivatives further support a 'single' source origin for these materials. Combustion source emissions favour particle transport (Wild and Jones, 1995), deposition and retention in sediments composed of fine silt or clay (McCaIn et al., 1992 and Means et al., 1980). Grain size analyses of the sediment samples indicates that most are taken from regions of high stress or, where minimal silt or clay size material (< 63  $\mu$ m) is deposited, These samples have low PAH residue levels and no PCBs (Fig. 3). Size analyses for Stns 5, 6, 12, 13, 17, 18 and 19 favour little stress and a higher percentage of mud in the sediment. The highest PAH residues are associated with these stations (Fig. 3). For all the samples studied the residue pattern and the ratios between detected PAHs are essentially the same, suggesting a similar source with little or no extraneous input. The spatial extent of sediment contaminants is very localised, influenced directly by grain size and tends to be associated with population centers.

Passamaquoddy Bay sediment has a high percentage of mud or fine grain particles due in part to its partial isolation from the Bay of Fundy. These particles favour retention of organic contaminants, not only PAH residues but also other organic materials. Contaminant input from population centers or aquatic activities would be retained in the sediment rather than removed through flushing action of tidal cycles, as was observed in the St. Croix River stations.

### **Metallic Contaminants**

Table 2 summarizes the mean concentrations of Al, As, Ba, Cd, Co, Cr, Cu, Fe, Li, Mn, Ni, Pb, Sr, Tl, U, V, and Zn in Passamaquoddy Bay and the St Croix estuary.

The concentrations of the metals at each sampling station are shown in Figs. 5-21. The range of total metal concentrations determined in the estuarine sediments are: As 9-27 mg·kg<sup>-1</sup>; Ba 290-420 mg·kg<sup>-1</sup>; Cd 0.03-0.18 mg·kg<sup>-1</sup>; Co 6-15 mg·kg<sup>-1</sup>; Cr 26-72 mg·kg<sup>-1</sup>; Cu 6-22 mg·kg<sup>-1</sup>; Ni 13-41 mg·kg<sup>-1</sup>; Pb 20-35 mg·kg<sup>-1</sup>; Sr 91-140 mg·kg<sup>-1</sup>; Tl 0.6-0.8 mg·kg<sup>-1</sup>; U 1.4-2.4 mg·kg<sup>-1</sup>; V 57-110 mg·kg<sup>-1</sup>; and Zn 48-100 mg·kg<sup>-1</sup> (Table 2). Most of the metals are at, or near, natural concentrations but, for As and Ni respectively, 20% and 5% of the samples exceeded the contamination threshold (>20 and >40 mg·kg<sup>-1</sup>). The highest concentrations of these metals occur in fine-grained sediments off Todd Point about 5 km below the mouth of the St Croix river (Stn. 5, Fig.1) and the lowest in the muddy sands in the outer part of Letang inlet (Stn 3 and 4), and in the St Croix estuary (Stns 8-11, and 15).

The metal concentrations in sediments from the mouth of the St. Croix river seaward along a 22 km long section (Stns. 5-16, Fig.1 ) are also summarized in Table 2. The range of total metal concentrations determined in the estuarine sediments are: As 9-27 mg·kg<sup>-1</sup>; Ba 290-420 mg·kg<sup>-1</sup>; Cd 0.03-0.18 mg·kg<sup>-1</sup>; Co 9-15 mg·kg<sup>-1</sup>; Cr 44-72 mg·kg<sup>-1</sup>; Cu 8-22 mg·kg<sup>-1</sup>; Ni 22-41 mg·kg<sup>-1</sup>; Pb 20-35 mg·kg<sup>-1</sup>; Sr 91-120 mg·kg<sup>-1</sup>; Tl 0.6-0.7 mg·kg<sup>-1</sup>; V 73-110 mg·kg<sup>-1</sup>, and Zn 55-99 mg·kg<sup>-1</sup>. Normalization of the trace metal data with Al and Li shows a marked decrease in Cd concentrations and a lesser decrease in Cu and Pb concentrations seaward. The other metals, however, remain relatively constant or vary inconsistently seaward from the mouth of the river.

Table 3 shows the average concentrations of As, Cd, Cr, Cu, Pb, and Zn in fine-grained sediments compared to those found in the estuaries of the La Have and St Mary's rivers in Nova Scotia (Loring et al, 1996). The data indicate that the St Croix estuary contains similar Cr and Cu concentrations, but more Pb and Zn, and less As, and Cd than the La Have estuary; in relation to the St Mary's estuary, the

concentrations of As and Cu are similar, but concentrations of Cr, Pb, and Zn are higher and Cd concentrations are lower.

The strong positive correlations of all the metals, excepting As, and Mn, with increasing mud (material <63µm) content of all the sediments (Table 4) as well as in those from the St Croix seaward transect (Table 5) confirm that the metal carriers are predominantly fine-grained. The strong positive correlations of all the metals (Tables 4 and 5), excepting As, Cd, and Mn with increasing Al and Li (except Ba) concentrations reflect their inclusion and/or association with the fine-grained aluminosilicate minerals such as the micas, illite, chlorite and kaolinite that comprise the bulk of the sedimentary material. The proportion of the metal variability explained by the granular variability of the aluminosilicates represented by Al decreases in the order: U> Zn> Ba> Co> Cr> Ni > Cu> V> Pb>> Cd>> As>> Mn >Ti. In addition, the strong covariances of Cr, Co, Cu, Ni, V, and Zn with Fe and with Li suggests that important carriers of these metals are the ferromagnesium primary and secondary minerals such as pyroxenes, hornblende and chlorite. In addition, As is significantly correlated with Mn, and Cd with Pb. These associations probably reflect the diagenetic association of As with Mn at the sediment-water interface and the small amounts of Cd with Pb in sulphides present in the sediments. Factor analyses based on the St. Croix correlation matrix (Table 5) show that three factors account for 89.7% of the total metal variance (Table 6). A common factor (1) has significant loadings of Al, Ba, Co, Cr, Fe, Ni, V, U, Zn, and material <63µm and accounts for 65.7% of the total problem variance. Factor 1 is essentially the sedimentation factor in which the accumulation of these metals is controlled by the deposition of fine grained silicate materials with which these metals are associated at sites determined by the physical, chemical and hydrodynamic conditions in the estuary. Factor 2 (14.4% of the total problem variance) with significant loadings on Cd, Pb and Zn suggests that these metals behave independently and are most likely derived from fine grained Cd, Pb and Zn sulfide particles and /or discrete metallic particles. The lack of strong correlations of these metals with Fe suggests that Fe sulphides are not the significant carriers of Cd and Pb. The seaward decline in Cd and, to a certain extent, Pb and Zn concentrations suggests an anthropogenic input of Cd and perhaps Pb and Zn from the St. Croix river system. Factor 3 (9.6% of the total variance ) with significant loadings on As and Mn suggests that As is associated with Mn oxides and grain coatings as a result of post depositional diagenetic processes at the sediment water interface.

The sediment samples obtained from the cross section of Passamaquoddy Bay, Letang inlet and off Nancy Head (Stns. 1 and 2) provide information on the levels of metals at these sites (Figs 5-21) but are insufficient in numbers to perform a significant factor analyses of the data. The metal data indicate that the levels at these sites are at or near natural background levels.

## Conclusions

The disaggregated inorganic grain size of the samples analyzed reflects the changing physical oceanography of the St. Croix-Passamaquoddy system. In regions of high current flow, well-sorted sand peaks resulting from saltation load dominate the sediment size distributions. In quieter areas, the high concentration of fine suspended particulate material readily aggregates and is deposited on the bottom as flocs. Bottom sediment in the central part of Passamaquoddy Bay consists of over 80% floc settled material, which accounts for the enrichment of contaminants at the three stations sampled. The similarity in the source slope and degree of flocculation between samples from this region and Letang Inlet suggests that they are derived from the same source and that aggregation dynamics are similar in the two areas. The high degree of floc settling and the lack of resuspension found in the samples collected from Passamaquoddy Bay suggests that this area would be unsuitable for salmon aquaculture.

The highest concentrations of the metals occur in fine-grained sediments about 5 km below the mouth of the St Croix river and the lowest in the muddy sands in outer part of Letang inlet and the St Croix estuary. Most of the metals are at, or near, natural concentrations but 20% and 5% of the samples exceeded the contamination threshold, ( $>20 \text{ mgAs/kg}$   $>40 \text{ mgNi/kg}$ ) levels for As and Ni concentrations respectively. Normalization of the metal data suggests anthropogenic inputs of Cd and perhaps Pb and Zn from the St. Croix river although the amounts have not yet exceeded contamination levels. Overall, the abundance and distribution of most of the metals, except As, are controlled by the accumulation of fine-grained primary and secondary iron-rich aluminosilicate minerals and materials in Passamaquoddy Bay. Arsenic, however, behaves independently and its distribution and accumulation can be related to post-depositional diagenetic effects of Fe-Mn cycling at, and near, the sediment water interface. Organic analyses indicate that measurable PCB residues occur in only one sample. Measurable PAH residues ( $67\text{-}3144 \text{ ng}\cdot\text{g}^{-1}$ ) occur in all analysed samples but are within the Canadian Ocean Dumping Guidelines. Residue patterns for the contaminants surveyed are consistent with hydrocarbon residue levels from combustion and ship traffic and/or municipal sources. Although these data represent a one time sampling, the possibility of increasing contaminant concentrations in the sediment exists. The build-up of contaminants in fine grained sediments over time may have a detrimental effect on activities such as aquaculture in Passamaquoddy Bay, and, to a lesser extent, in the St. Croix estuary.

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## **Table Captions :**

Table 1 PAH compounds in sediments from Passamaquoddy Bay and the St.Croix Estuary

Table 2 Metal Statistics for Passamaquoddy Bay and the St.Croix Estuary sediments ...

Table 3 Average concentrations of As, Cd, Cr, Cu, Pb, and Zn in the St Croix estuarine fine grained sediments<sup>1</sup> compared to those found those from the La Have and St Mary's estuaries<sup>2</sup>, Nova Scotia

Table 4 Correlation Matrix ( $p \leq 0.01$ ) for all samples ( $n=19$ ). x= not significant at  $p \leq 0.01$ )

Table 5 Correlation Matrix ( $p \leq 0.01$ ) for St. Croix long section ( $n=12$ ). x= not significant at  $p \leq 0.01$

Table 6 Varimax Metal Matrix for St Croix section



Table 1

## PAH compounds in sediments from Passamaquoddy Bay and the St.Croix Estuary

Station	5	6	7	8	9	10	11
Compound	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>
naphthalene	D	D	D	D	D	D	D
quinoline	ND	ND	ND	ND	ND	ND	ND
isoquinoline	ND	ND	ND	ND	ND	ND	ND
8-methylquinoline	ND	ND	ND	ND	ND	ND	ND
3-methylquinoline	ND	ND	ND	ND	ND	ND	ND
7-methylquinoline	ND	ND	ND	ND	ND	ND	ND
2,7-dimethylnaphthalene	D	D	D	D	D	D	ND
1,6-dimethylnaphthalene	D	D	D	D	D	ND	ND
2,3-dimethylnaphthalene/ 1,4-dimethylnaphthalene/ 1,2-dimethylnaphthalene.	D	D	D	D	D	ND	ND
1,5-dimethylnaphthalene	D	D	D	ND	D	ND	ND
acenaphthylene	D	D	D	ND	D	ND	ND
acenaphthene	D	D	D	D	D	D	ND
2,3,5-trimethylnaphthlene	D	D	ND	D	D	D	ND
5-nitroquinoline	ND	ND	ND	ND	ND	ND	ND
fluorene	21	17	ND	ND	ND	ND	ND
6-nitroquinoline	ND	ND	ND	ND	ND	ND	ND
9,10-dihydroanthracene	ND	ND	ND	ND	ND	ND	ND
1-methylfluorene	11	ND	ND	ND	ND	ND	ND
8-nitroquinoline	ND	ND	ND	ND	ND	ND	ND
phenanthrene	204	115	22	10	43	14	ND
anthracene	65	23	21	ND	12	ND	ND
acridine	ND	ND	ND	ND	ND	ND	ND
benzo[h]quinoline	ND	ND	ND	ND	ND	ND	ND
phenanthridine	ND	ND	ND	ND	ND	ND	ND
carbazole	ND	13	ND	ND	ND	ND	ND
2-methylanthracene	ND	ND	ND	ND	ND	ND	ND
1-methylanthracene	ND	ND	ND	ND	ND	ND	ND
9-methylanthracene	ND	ND	ND	ND	ND	ND	ND
4H-cyclopenta[def]phenanthrene	33	18	ND	ND	ND	ND	ND
3,6-dimethylphenanthrene	36	19	10	ND	ND	ND	ND
fluoranthene	689	310	58	30	92	36	13
pyrene	632	275	48	24	73	28	13
benz[a]anthracene	291	135	25	14	37	15	ND
chrysene	327	183	34	17	49	20	ND
benzo[a]fluorene	99	48	11	ND	13	ND	ND
benzo[b]fluorene	65	21	ND	ND	ND	ND	ND
benzo[b+j]fluoranthene/ benzo[k]fluoranthene.	355	271	47	40	66	20	21
benzo[a]pyrene	169	125	21	19	28	12	10
indeno[123,cd]perylene	116	106	25	15	29		10
benzo[ghi]perylene	ND	101	19	ND	24	ND	ND
dibenz[a,c]anthracene/ dibenz[a,h]anthracene.	30	29	17	ND	14	ND	ND
SumPAH	3144	1809	359	170	478	146	67

Table 1 (cont'd)

Station	12	13	15	17	18	19
Compound	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>	ng·g <sup>-1</sup>
naphthalene	D	D	D	D	D	D
quinoline	D	ND	ND	ND	ND	ND
isoquinoline	D	ND	ND	ND	ND	ND
8-methylquinoline	ND	ND	ND	ND	ND	ND
3-methylquinoline	ND	ND	ND	ND	ND	ND
7-methylquinoline	ND	ND	ND	ND	ND	ND
2,7-dimethylnaphthalene	D	D	D	D	D	D
1,6-dimethylnaphthalene	ND	D	D	D	ND	D
2,3-dimethylnaphthalene/ 1,4-dimethylnaphthalene/ 1,2-dimethylnaphthalene.	ND	D	D	D	ND	D
1,5-dimethylnaphthalene	ND	D	D	D	ND	D
acenaphthylene	D	D	D	D	D	D
acenaphthene	D	D	D	D	D	D
2,3,5-trimethylnaphthlene	D	D	D	D	D	D
5-nitroquinoline	ND	ND	ND	ND	ND	ND
fluorene	22	10	ND	ND	11	14
6-nitroquinoline	ND	ND	ND	ND	ND	ND
9,10-dihydroanthracene	ND	ND	ND	ND	ND	ND
1-methylfluorene	ND	ND	ND	ND	ND	ND
8-nitroquinoline	ND	ND	ND	ND	ND	ND
phenanthrene	166	70	26	59	71	184
anthracene	47	16	9	16	19	48
acridine	ND	ND	ND	ND	ND	ND
benzo[h]quinoline	ND	ND	ND	ND	ND	ND
phenanthridine	ND	ND	ND	ND	ND	ND
carbazole	ND	10	ND	12	15	ND
2-methylanthracene	ND	ND	ND	ND	ND	ND
1-methylanthracene	ND	ND	ND	ND	ND	ND
9-methylanthracene	ND	ND	ND	ND	ND	ND
4H-cyclopenta[def]phenanthrene	24	12	ND	13	15	18
3,6-dimethylphenanthrene	30	13	9	13	16	ND
fluoranthene	506	206	67	165	205	553
pyrene	423	159	51	130	142	391
benz[a]anthracene	205	77	25	61	68	191
chrysene	220	102	33	88	84	244
benzo[a]fluorene	64	26	12	24	26	63
benzo[b]fluorene	33	15	ND	14	16	40
benzo[b+j]fluoranthene/ benzo[k]fluoranthene.	257	185	45	147	180	203
benzo[a]pyrene		65	20	57	69	77
indeno[123,cd]perylene	106	64	19	72	79	75
benzo[ghi]perylene	93	66	ND	65	63	87
dibenz[a,c]anthracene/ dibenz[a,h]anthracene.	22	18	11	25	29	31
SumPAH	2219	1114	327	961	1108	2220

## Abbreviations:

D Detected - not quantified due to low recovery

ND Not detected

/. co-eluting compounds

Table 2

Metal Statistics for Passamaquoddy Bay and the St.Croix Estuary sediments. Concentrations in  $\text{mg}\cdot\text{kg}^{-1}$

except percent (%) per dry weight for Al, Fe, and mud (material  $<63\mu\text{m}$  in diameter):

All samples n=19

	Mean	SD	Range		Mean	SD	Range
Al%	6.17	0.52	5.1-6.9	Mn	589	146	450-910
As	14.71	4.4	9.4-27	Ni	28	7.5	13-41
Ba	363	39.6	290-420	Pb	28	4.7	20-35
Cr	57	14.2	26-72	Sr	106	14.5	91-140
Co	11	2.7	6-15	Tl	0.6	0.06	0.6-0.8
Cu	14	4.9	6-22	U	1.8	0.32	1.4-2.4
Cd	0.07	0.03	0.03-0.18	V	93	16.7	57-110
Fe%	3.31	0.60	2-4	Zn	78	18.1	48-100
Li	45	8.0	27-55	Mud%	50	34.5	6-96

Sr Croix Estuary Section n=12

	Mean	SD	Range		Mean	SD	Range
Al%	5.99	0.49	5.1-6.6	Mn	598	140	450-860
As	15.4	1.4	9-27	Ni	28	6	22-41
Ba	345	39	290-490	Pb	27	5	20-35
Cr	56	10	44-72	Sr	100	8	91-120
Co	11	2	9-15	Tl	0.6	0.03	0.6-0.7
Cu	14	5	8-22	U	1.75	0.29	1.4-2.2
Cd	0.07	0.04	0.03-0.18	V	92	12	73-110
Fe%	3.29	0.43	2.70-4.00	Zn	75	15	55-99
Li	45	5	36-52	Mud%	42.6	31.3	6-88

Table 3

Average concentrations of As, Cd, Cr, Cu, Pb, and Zn in the St Croix estuarine fine grained sediments<sup>1</sup> compared to those found those from the La Have and St Mary's estuaries<sup>2</sup>, Nova Scotia

	n	As	Cd	Cr	Cu	Pb	Zn
SC AVG.	7	15	0.09	69	18	33	97
SD		6	0.04	4	1	2	4
LA AVG.	20	27	0.33	84	39	58	118
SD		6	0.10	12	11	18	25
SM AVG	5	19	0.38	44	14	21	72
SD		7	0.13	4	3	5	17

<sup>1</sup>For sediments containing >70% material <63µm in diameter

<sup>2</sup>Metal data from Loring et al, 1996

n = number of samples; AVG = average; SD = standard deviation

SC= St Croix estuary; LA = LaHave Estuary; SM= St.Mary's Estuary

Table 4 Correlation Matrix ( $p \leq 0.01$ ) for all samples ( $n=19$ ). x= not significant at  $p \leq 0.01$

	Al	As	Ba	Cr	Co	Cu	Cd	Fe	Pb	Mn	Ni	Sr	Tl	U	V	Zn	Li	Mud
Al																		
As	x	1																
Ba	0.82	x	1															
Cr	0.79	x	0.60	1														
Co	0.79	x	0.70	0.94	1													
Cu	0.75	x	0.68	0.90	0.94	1												
Cd	0.30	x	x	x	x	x	1											
Fe	0.76	x	x	0.96	0.94	0.89	x	1										
Pb	0.65	x	x	0.58	x	0.57	0.62	x	1									
Mn	x	0.78	x	x	x	x	x	x	x	1								
Ni	0.76	x	0.64	0.97	0.96	0.95	x	0.95	x	x	1							
Sr	x	x	x	x	x	x	x	x	x	x	x	1						
Tl	x	x	x	x	x	x	x	x	x	x	x	0.56	1					
U	0.87	x	0.79	0.77	0.81	0.83	x	0.72	0.78	x	0.77	x	x	1				
V	0.71	x	x	0.94	0.92	0.85	x	0.95	x	x	0.93	x	x	0.67	1			
Zn	0.84	x	0.62	0.92	0.92	0.93	0.56	0.92	0.76	x	0.90	x	x	0.89	0.86	1		
Li	0.68	x	x	0.91	0.87	0.84	x	0.91	0.62	x	0.88	x	x	0.74	0.92	0.90	1	
Mud	0.84	x	0.72	0.85	0.88	0.91	0.56	0.82	0.78	x	0.85	x	x	0.95	0.77	0.96	0.81	1

Table 5 Correlation Matrix ( $p \leq 0.01$ ) for St. Croix long section ( $n=12$ ). x= not significant at  $p \leq 0.01$ )

	Al	As	Ba	Cr	Co	Cu	Cd	Fe	Pb	Mn	Ni	Sr	Tl	U	V	Zn	Li	Mud
Al	1																	
As	x	1																
Ba	0.76 x		1															
Cr	0.92 x		0.91	1														
Co	0.8 x		0.91	0.90	1													
Cu	0.74 x		0.83	0.86	0.94	1												
Cd	x	x	x	x	x	x	1											
Fe	0.88 x		0.77	0.89	0.9	0.83 x		1										
Pb	x	x	x	x	x	x	0.75 x		1									
Mn	x	0.81 x	x	x	x	x	x	x		1								
Ni	0.81 x		0.92	0.93	0.98	0.94 x		0.88 x	x		1							
Sr	x	x	x	x	x	x	x	x	x	x		1						
Tl	x	x	x	x	0.72 x	x		0.78 x	x		0.74 x		1					
U	0.77 x		0.73	0.83	0.83	0.88 x		0.75	0.73 x		0.81 x	x		1				
V	0.72 x		0.75	0.79	0.81	0.74 x		0.83 x	x		0.82 x	x	x		1			
Zn	0.84 x		x	0.84	0.82	0.89 x		0.83	0.76 x		0.79 x	x	x	0.94 x		1		
Li	x	x	x	x	x	0.72 x		0.71 x	x	x	x	x	x	0.78	0.72	0.78	1	
Mud	0.75 x		0.72	0.79	0.82	0.90	0.70	0.72	0.73 x		0.79 x	x	x	0.95 x		0.96	0.70	1

Table 6 Varimax Metal Matrix for St Croix section

Factor	1	2	3	4
% variance	65.7	14.4	9.6	2.8
Metal				
Al	0.77	-	-	0.48
As	-	-	0.92	-
Ba	0.93	-	-	-
Cd	-	0.91	-	-
Cr	0.87	-	-	-
Co	0.97	-	-	-
Cu	0.87	0.91	-	-
Fe	0.86	-	-	-
Li	0.54	-	-	-
Pb	-	0.89	-	-
Mn	-	-	0.96	-
Ni	0.97	-	-	-
V	0.81	-	-	-
U	0.69	0.63	-	-
Zn	0.68	0.68	-	-
<63µm	0.68	0.68	-	-

## Figure Captions

- Figure 1: Sample location map for Passamoquoddy Bay
- Figure 2: Disaggregated inorganic grain size (DIGS) distributions
- Figure 3:  $\Sigma$ PAHs in surficial sediments of Passamoquoddy Bay
- Figure 4: Variation of  $\Sigma$ PAHs with material  $<63\mu\text{m}$  in surficial sediments of Passamoquoddy Bay
- Figure 5: Aluminium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 6: Arsenic concentrations in surficial sediments of Passamoquoddy Bay
- Figure 7: Barium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 8: Cadmium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 9: Chromium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 10: Cobalt concentrations in surficial sediments of Passamoquoddy Bay
- Figure 11: Copper concentrations in surficial sediments of Passamoquoddy Bay
- Figure 12: Iron concentrations in surficial sediments of Passamoquoddy Bay
- Figure 13: Lead concentrations in surficial sediments of Passamoquoddy Bay
- Figure 14: Lithium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 15: Manganese concentrations in surficial sediments of Passamoquoddy Bay
- Figure 16: Nickel concentrations in surficial sediments of Passamoquoddy Bay
- Figure 17: Strontium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 18: Thallium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 19: Uranium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 20: Vanadium concentrations in surficial sediments of Passamoquoddy Bay
- Figure 21: Zinc concentrations in surficial sediments of Passamoquoddy Bay



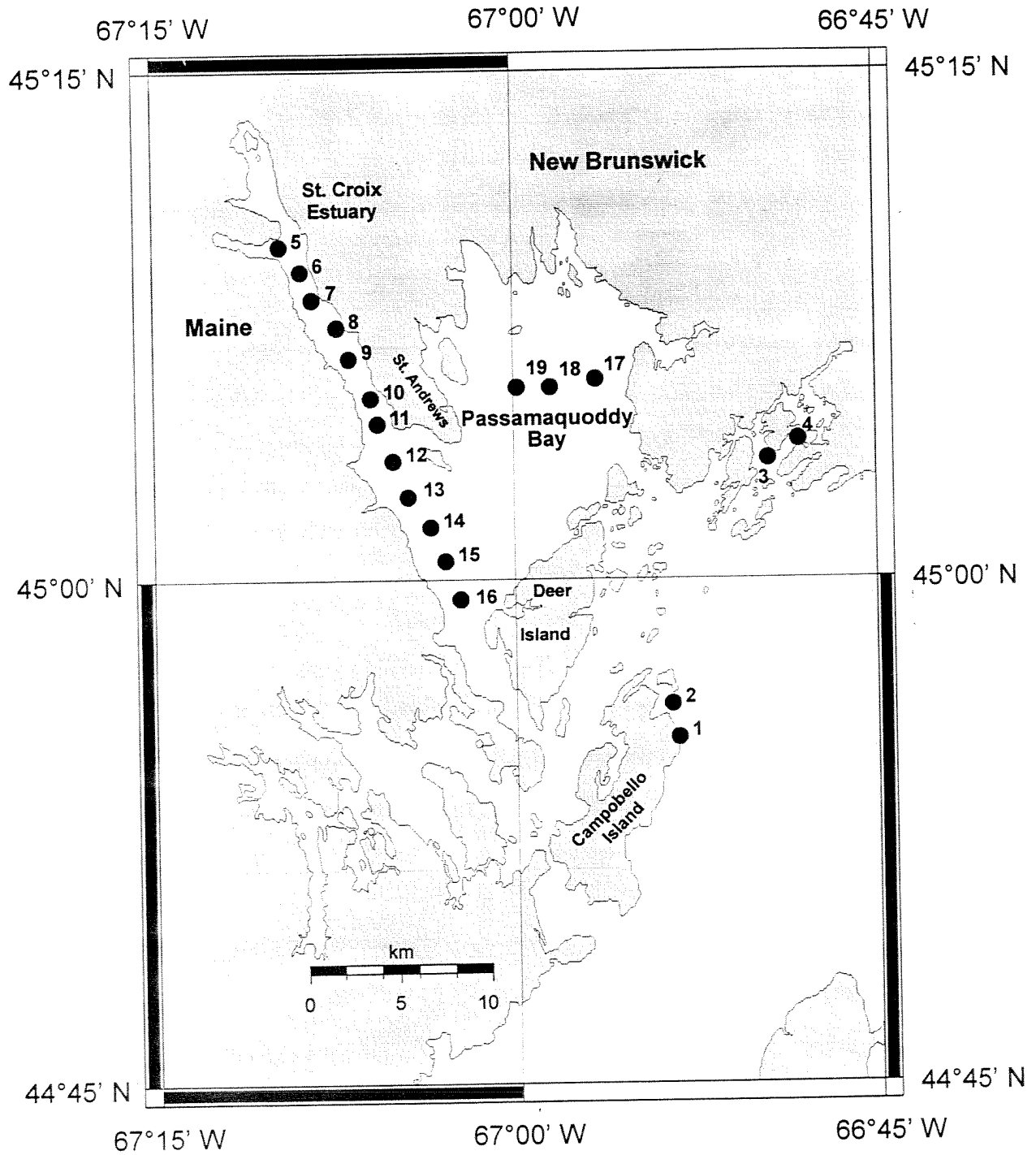


Figure 1: Sample location map for Passamaquoddy Bay.

# Passamaquoddy Bay Area, 1996

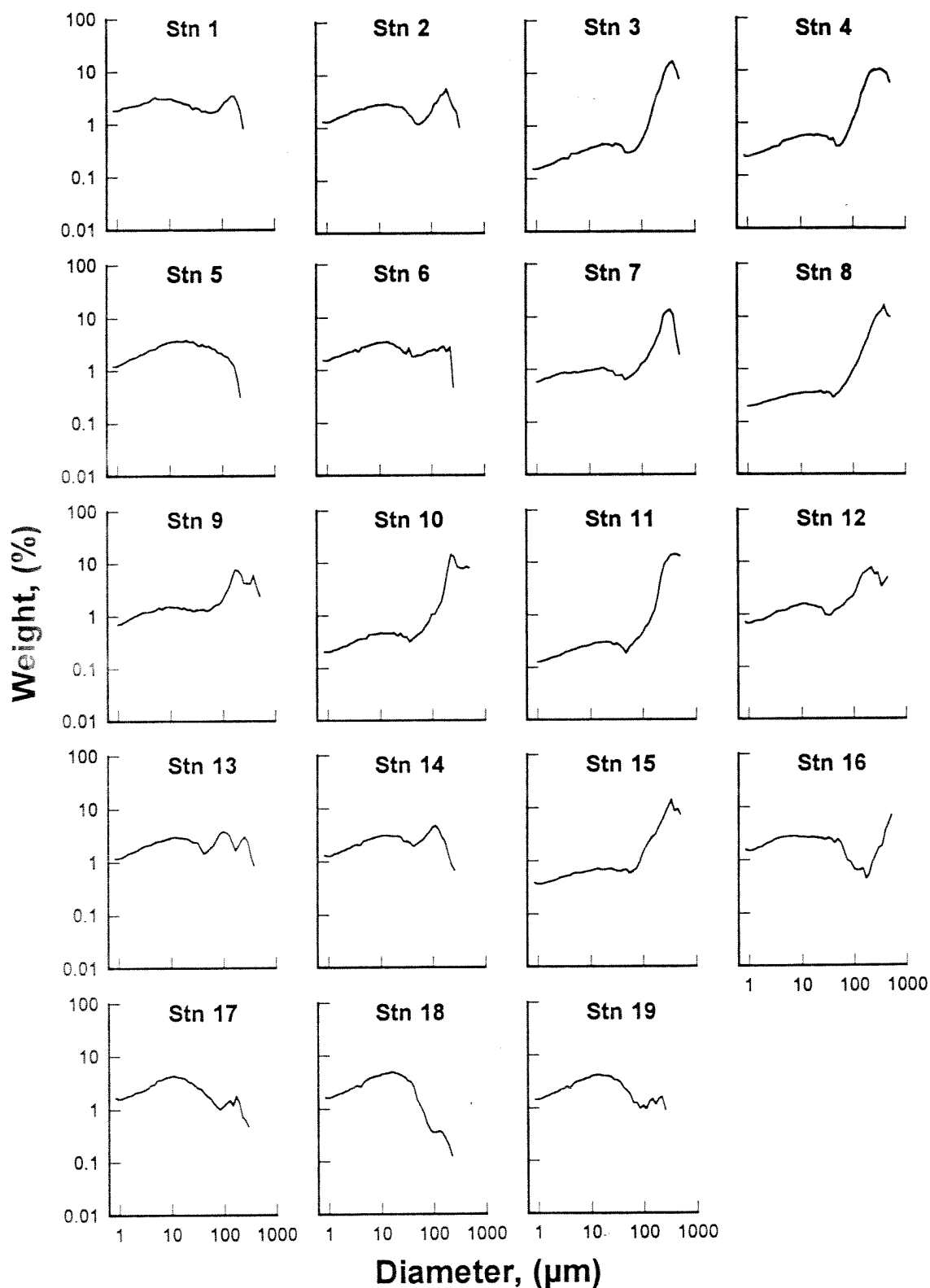


Figure 2: Disaggregated inorganic grain size (DIGS) distributions.

# Passamaquoddy Bay

$\Sigma$  PAH ng/g

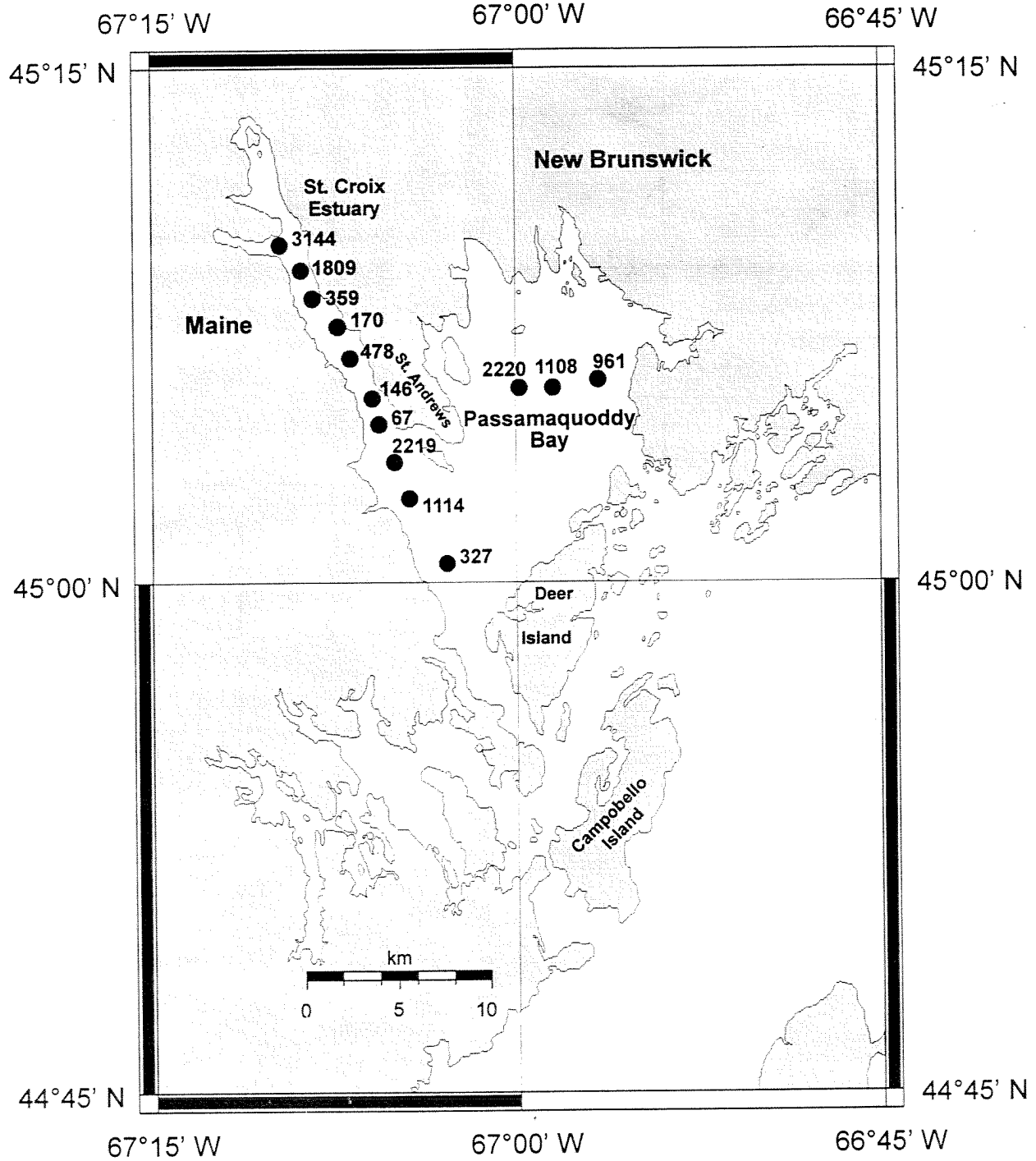


Figure 3:  $\Sigma$ PAHs in surficial sediments of Passamaquoddy Bay.

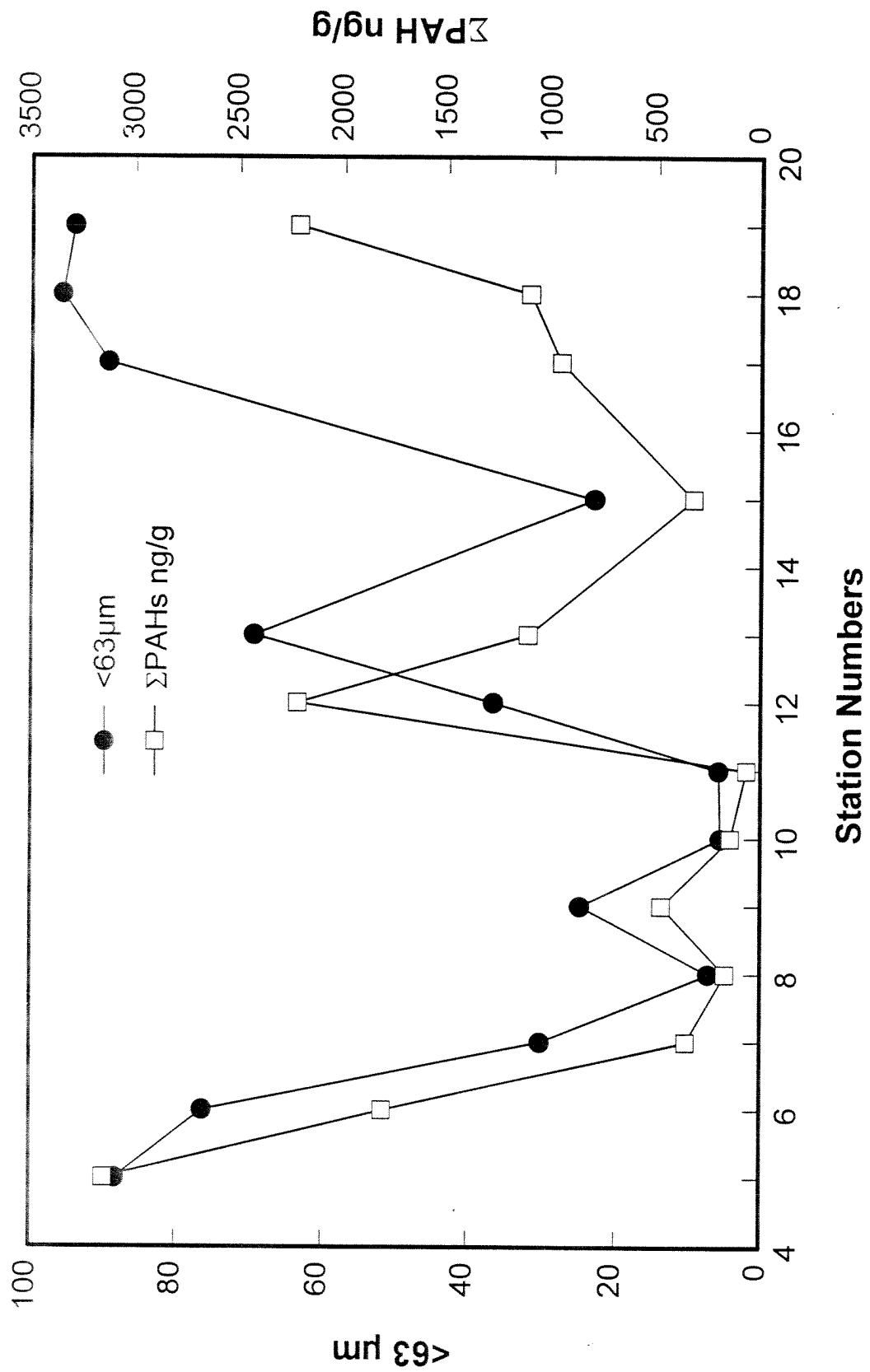


Figure 4: Variation of  $\Sigma$ PAHs with material <63  $\mu$ m in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay

Al %

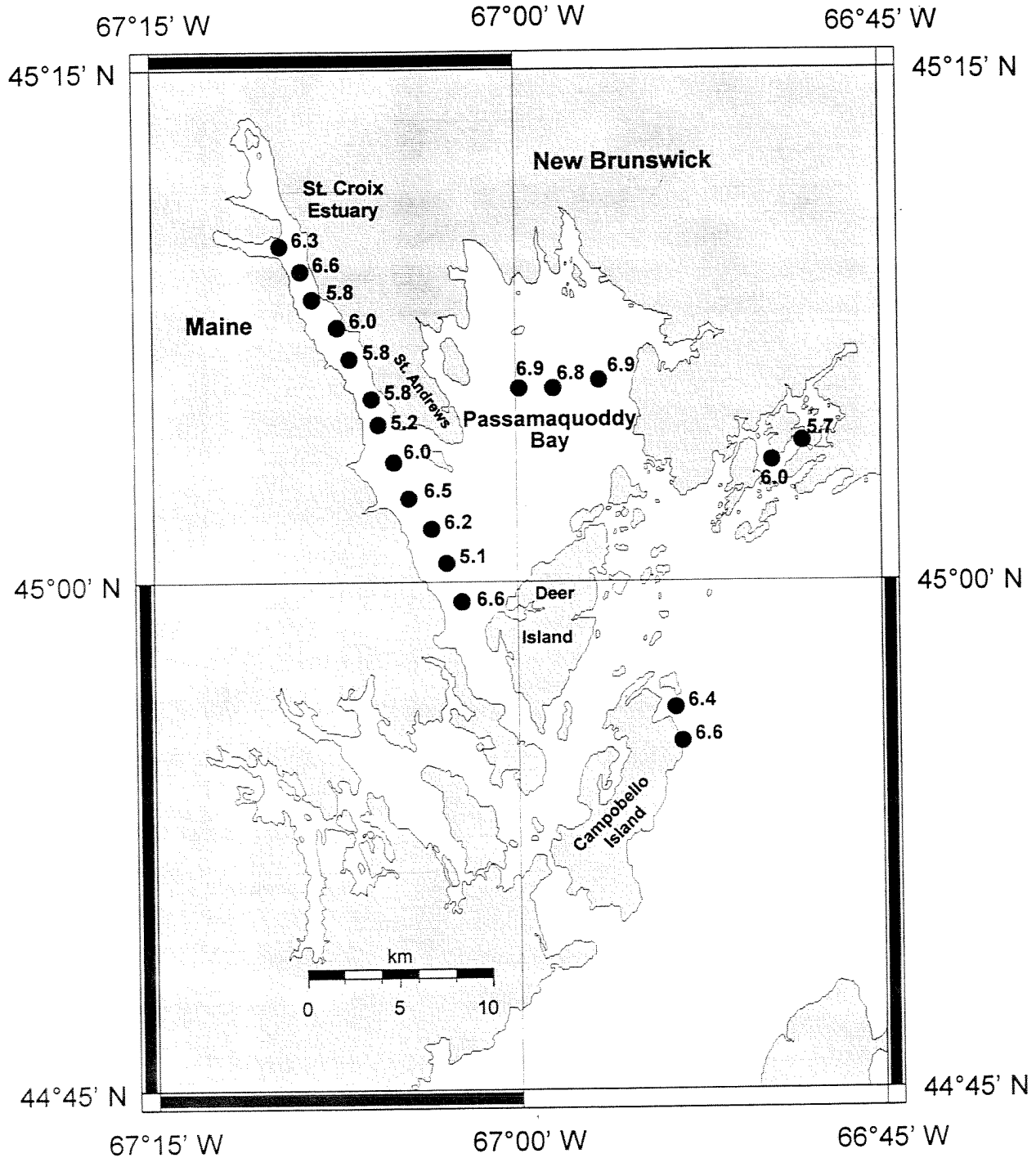


Figure 5: Aluminium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay As mg/kg

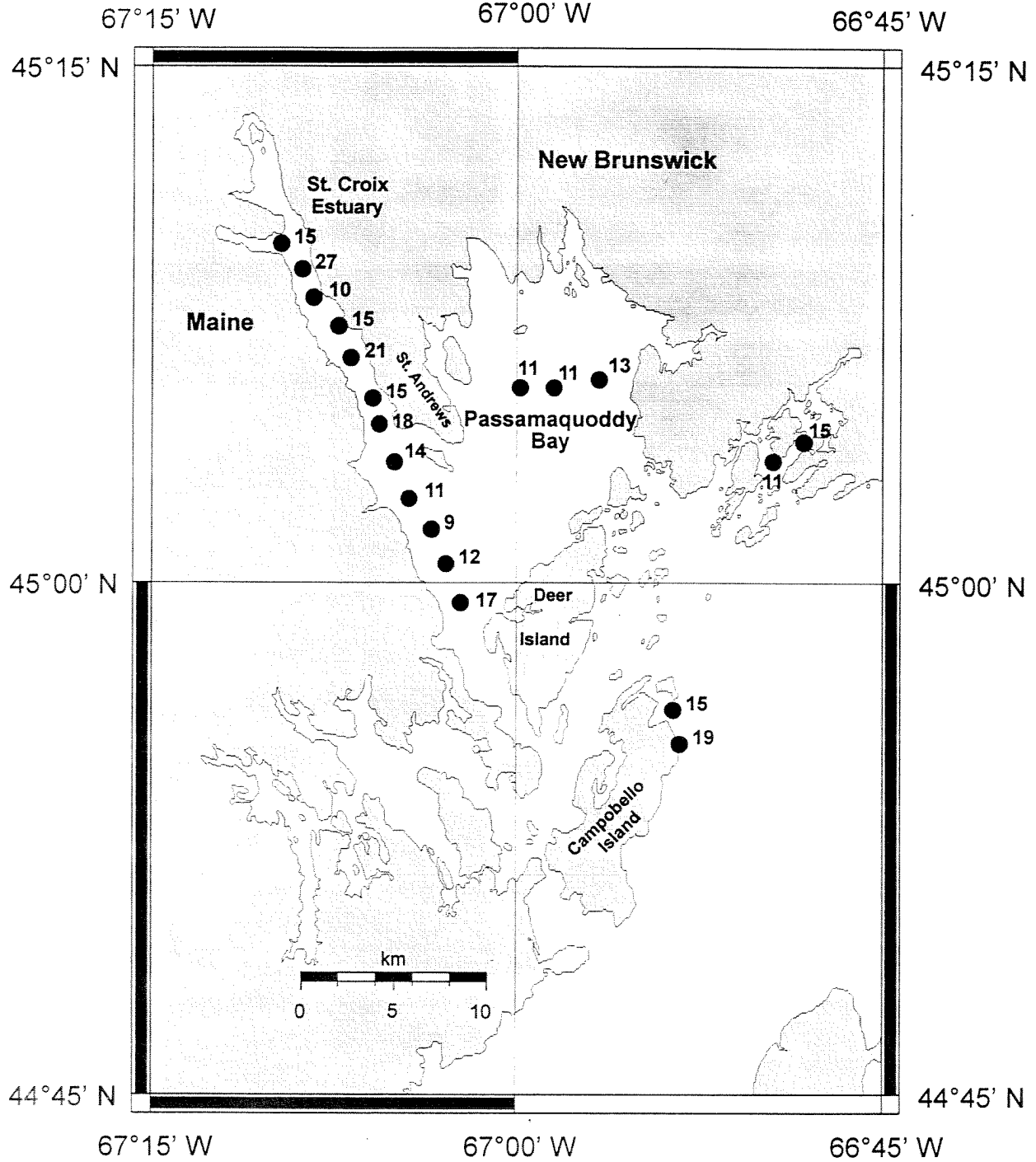


Figure 6: Arsenic concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Ba mg/kg

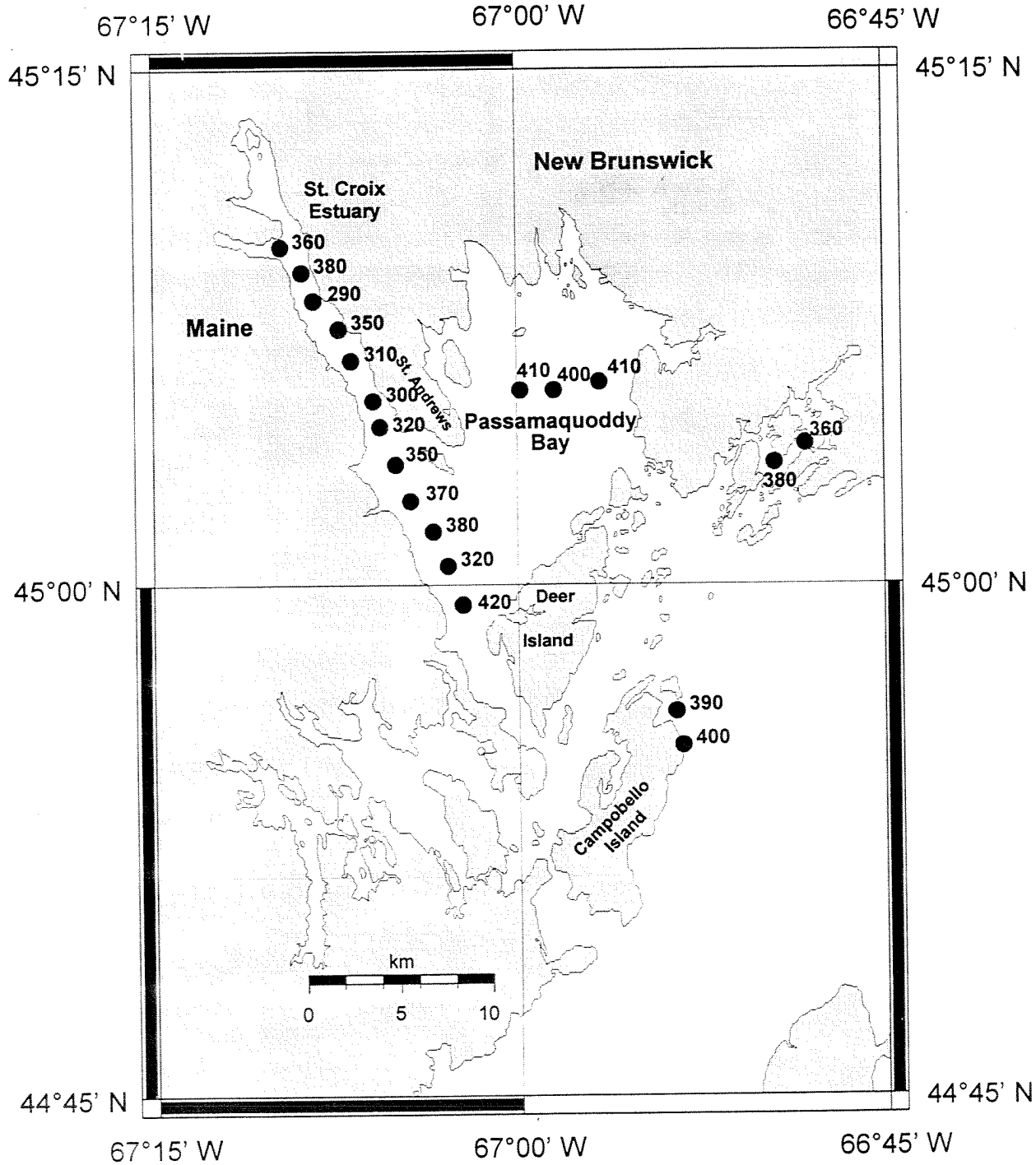


Figure 7: Barium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Cd mg/kg

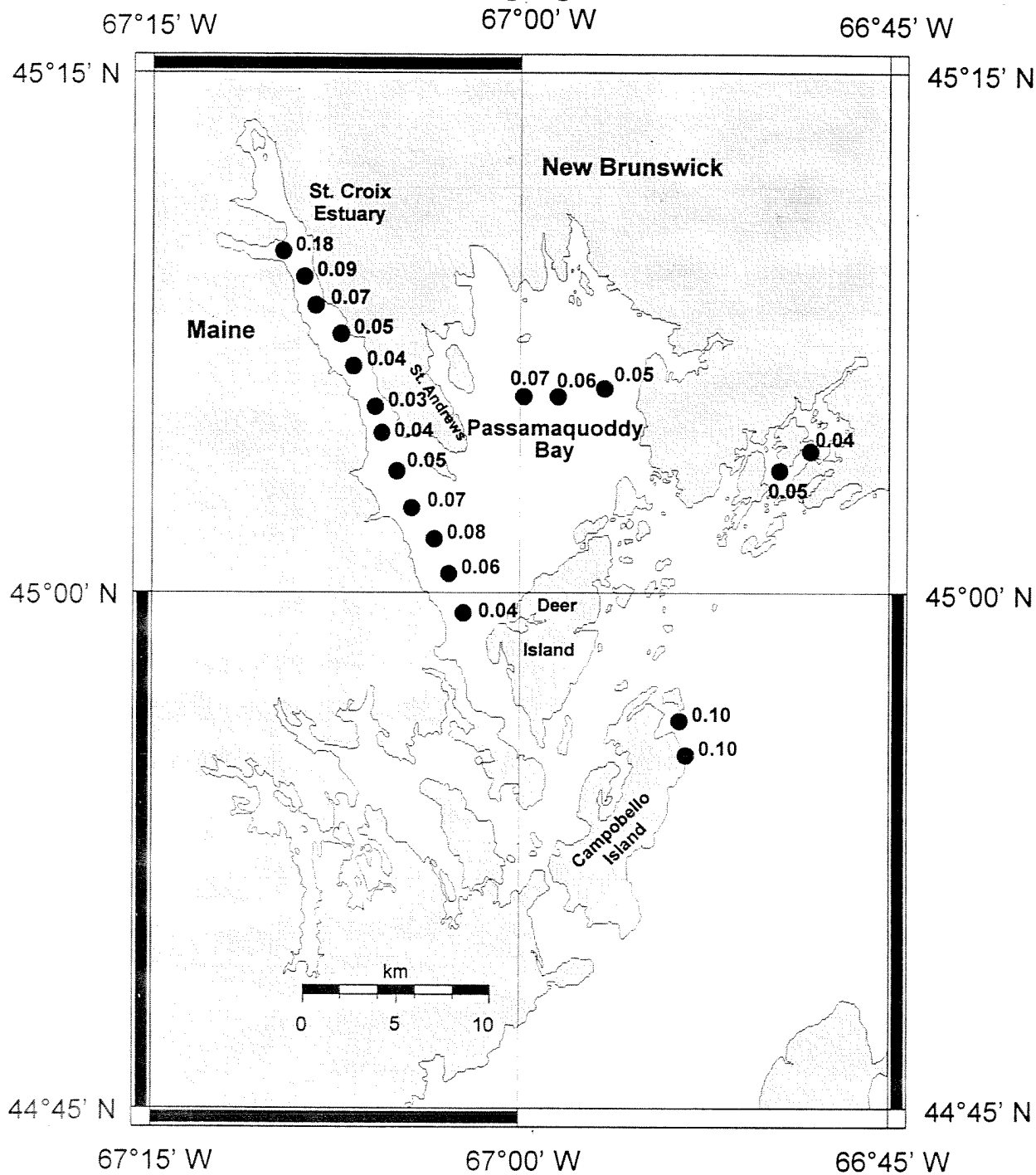


Figure 8: Cadmium concentrations in surficial sediments of Passamaquoddy Bay.



# Passamaquoddy Bay Cr mg/kg

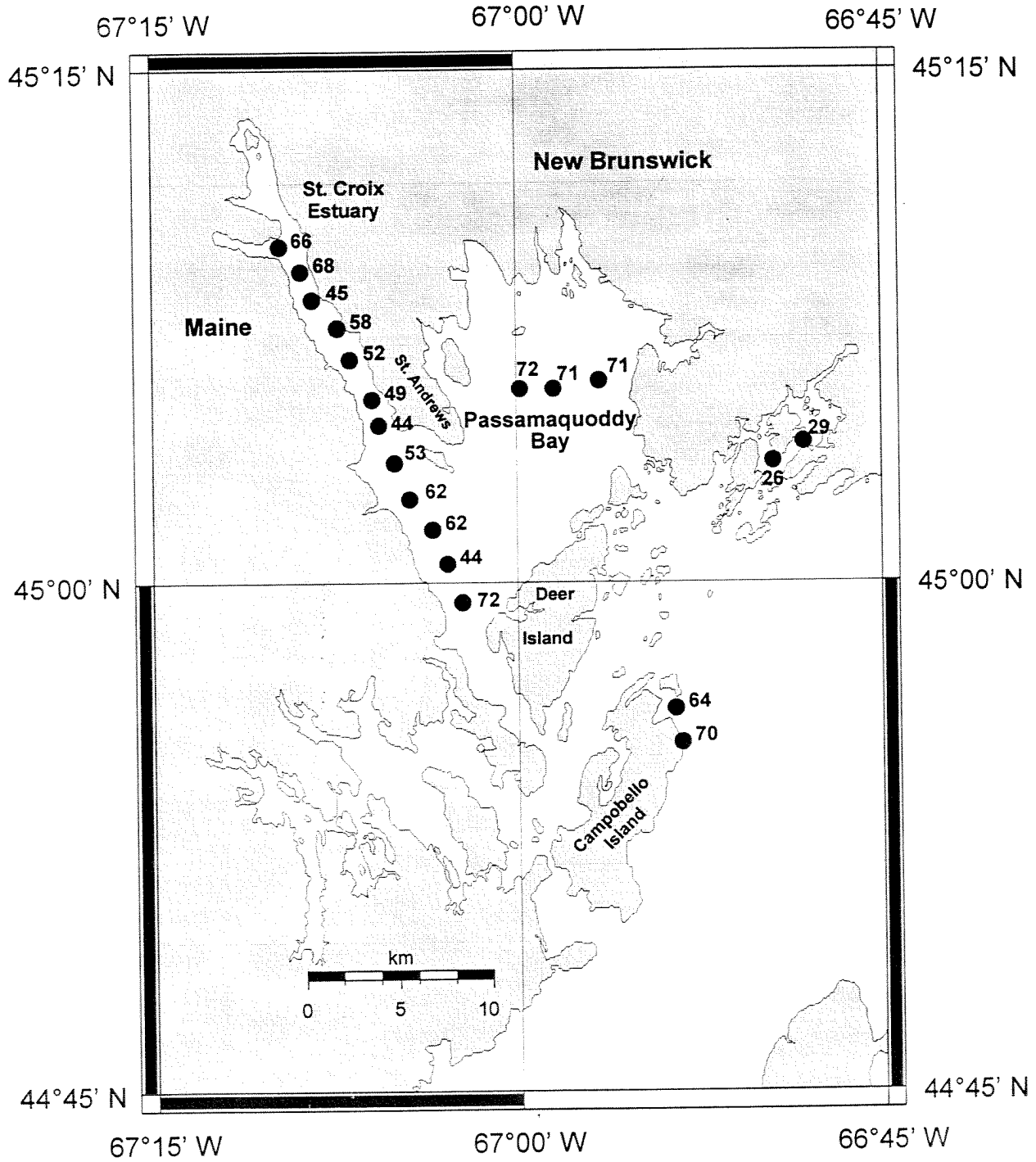


Figure 9: Chromium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Co mg/kg

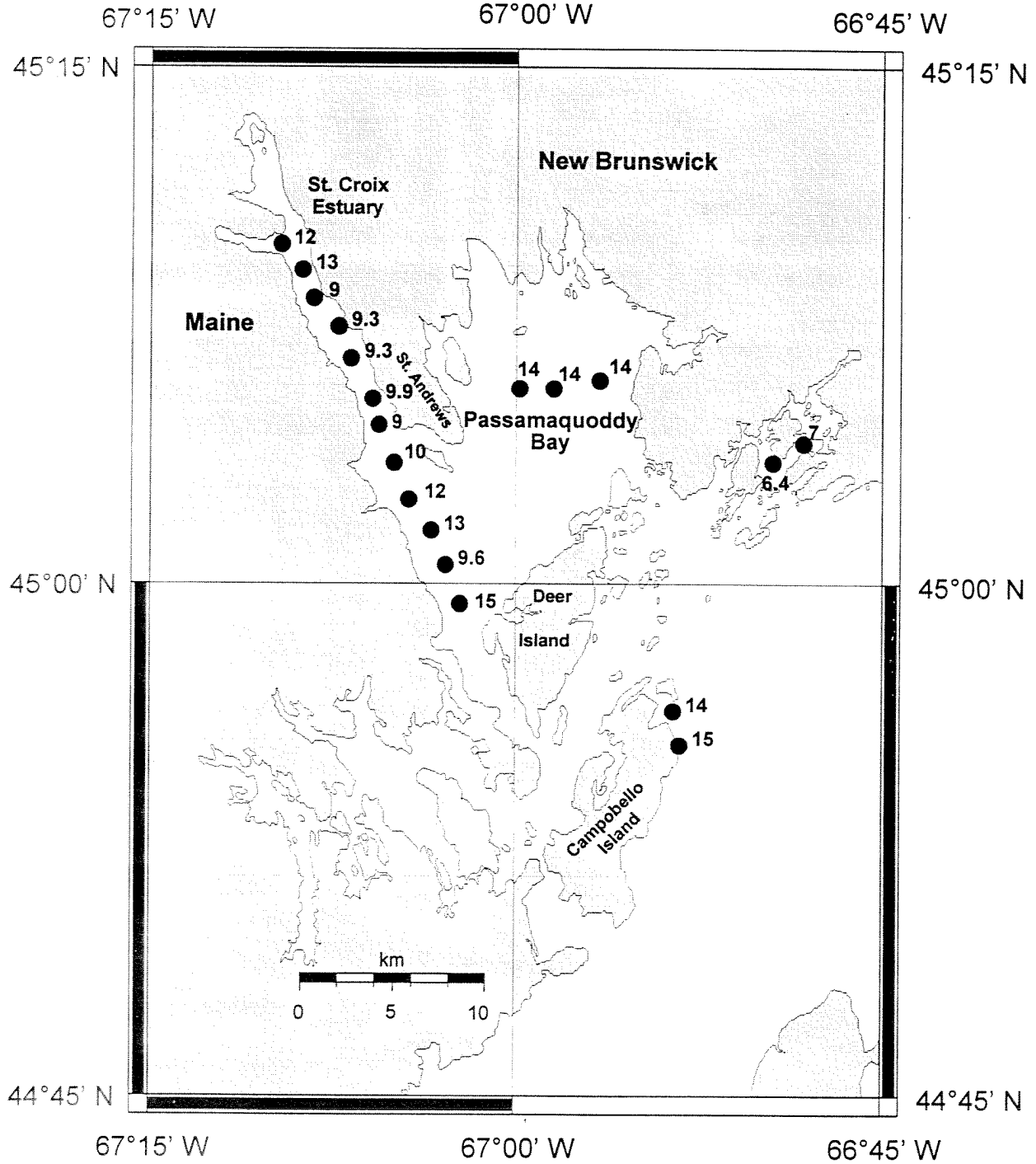


Figure 10: Cobalt concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Cu mg/kg

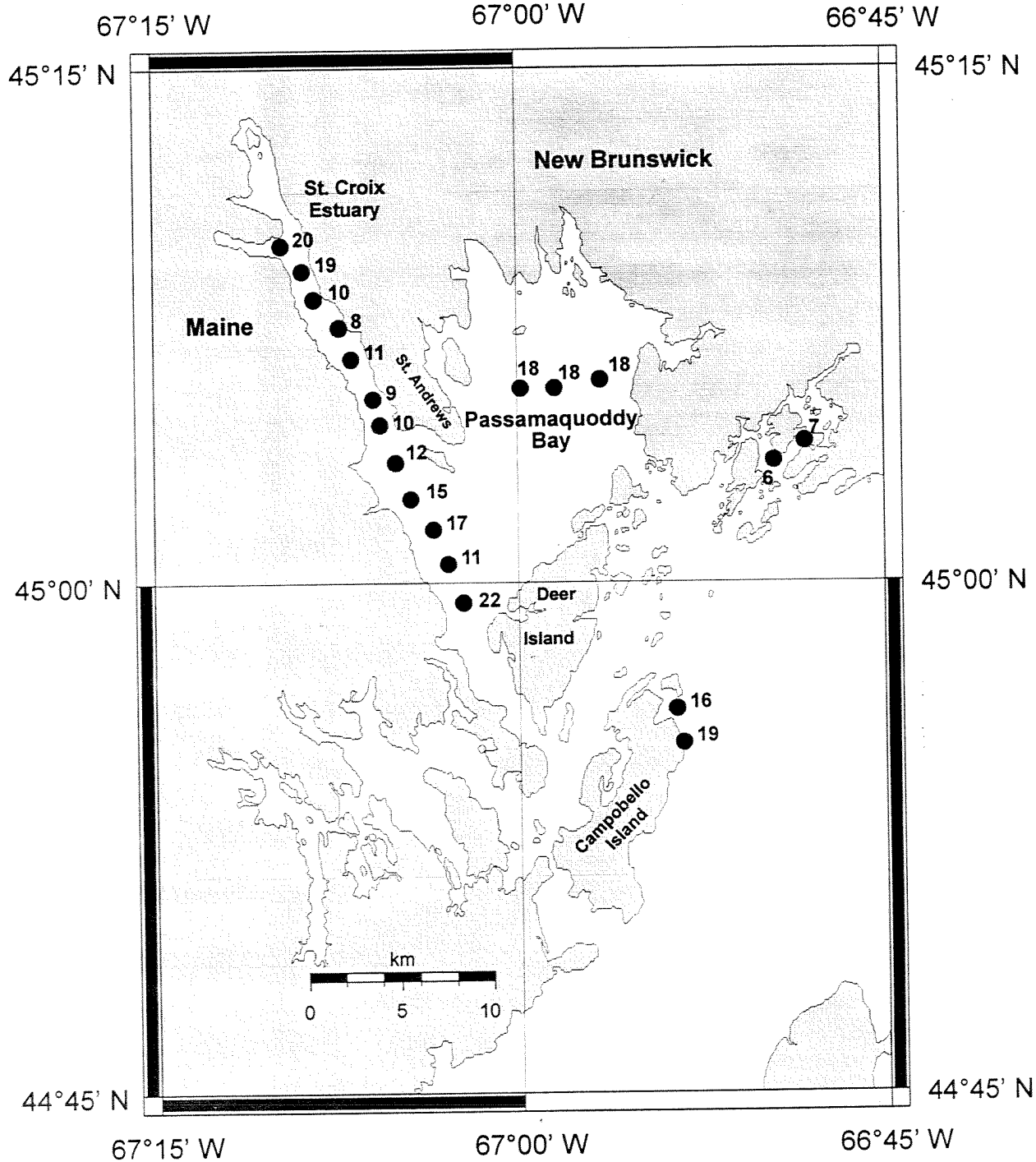


Figure 11: Copper concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Fe %

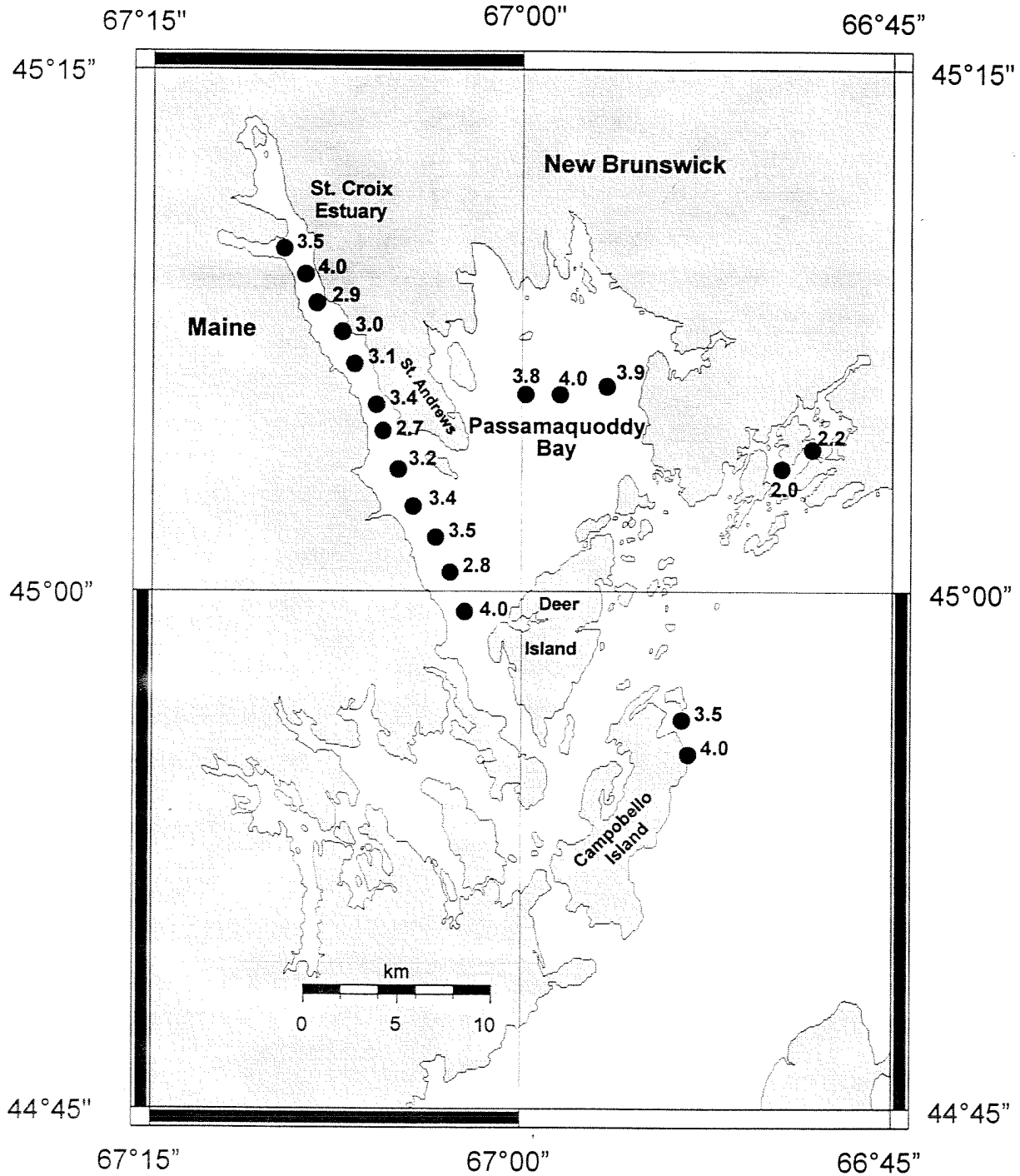


Figure 12: Iron concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Pb mg/kg

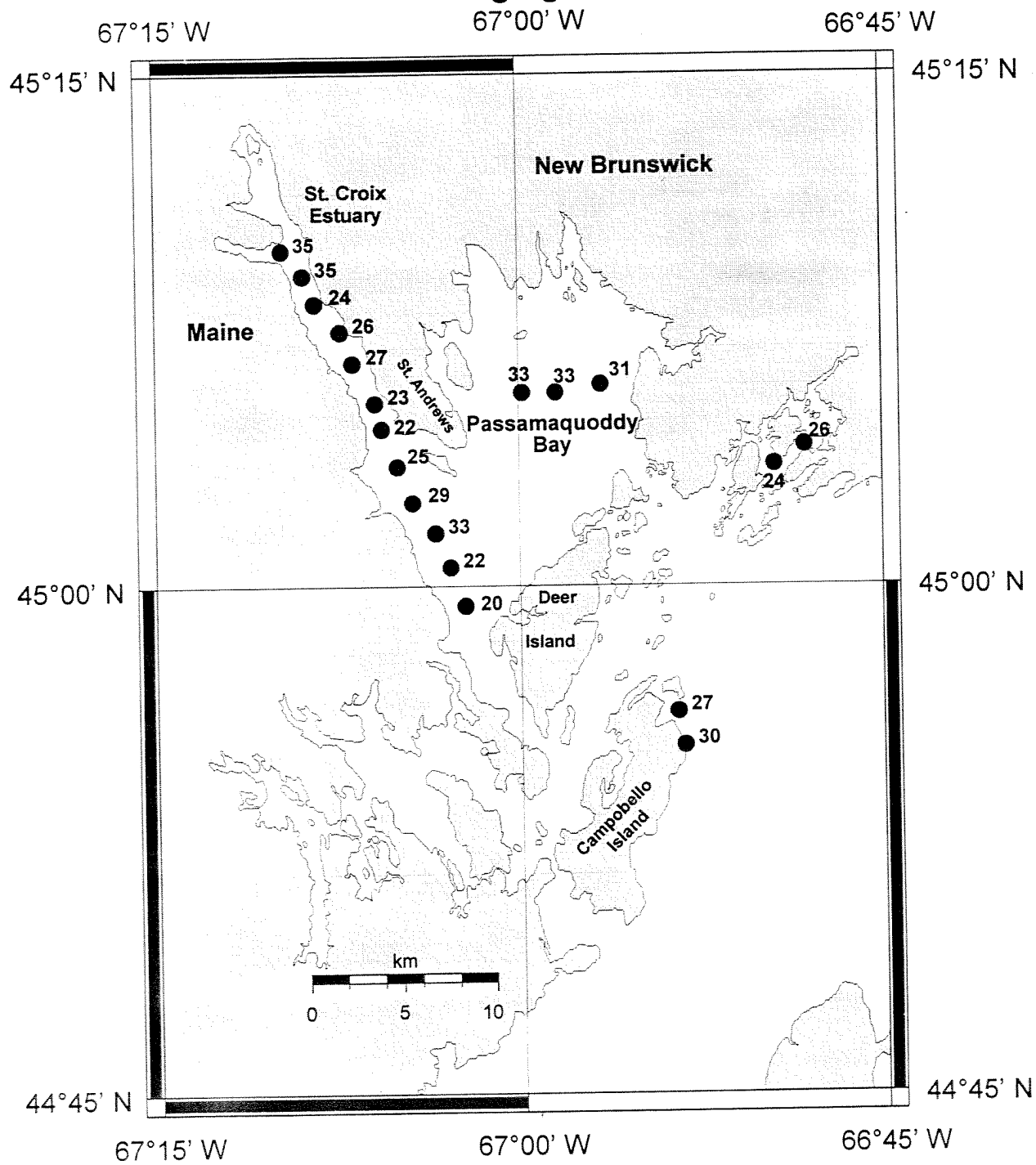


Figure 13: Lead concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Li mg/kg

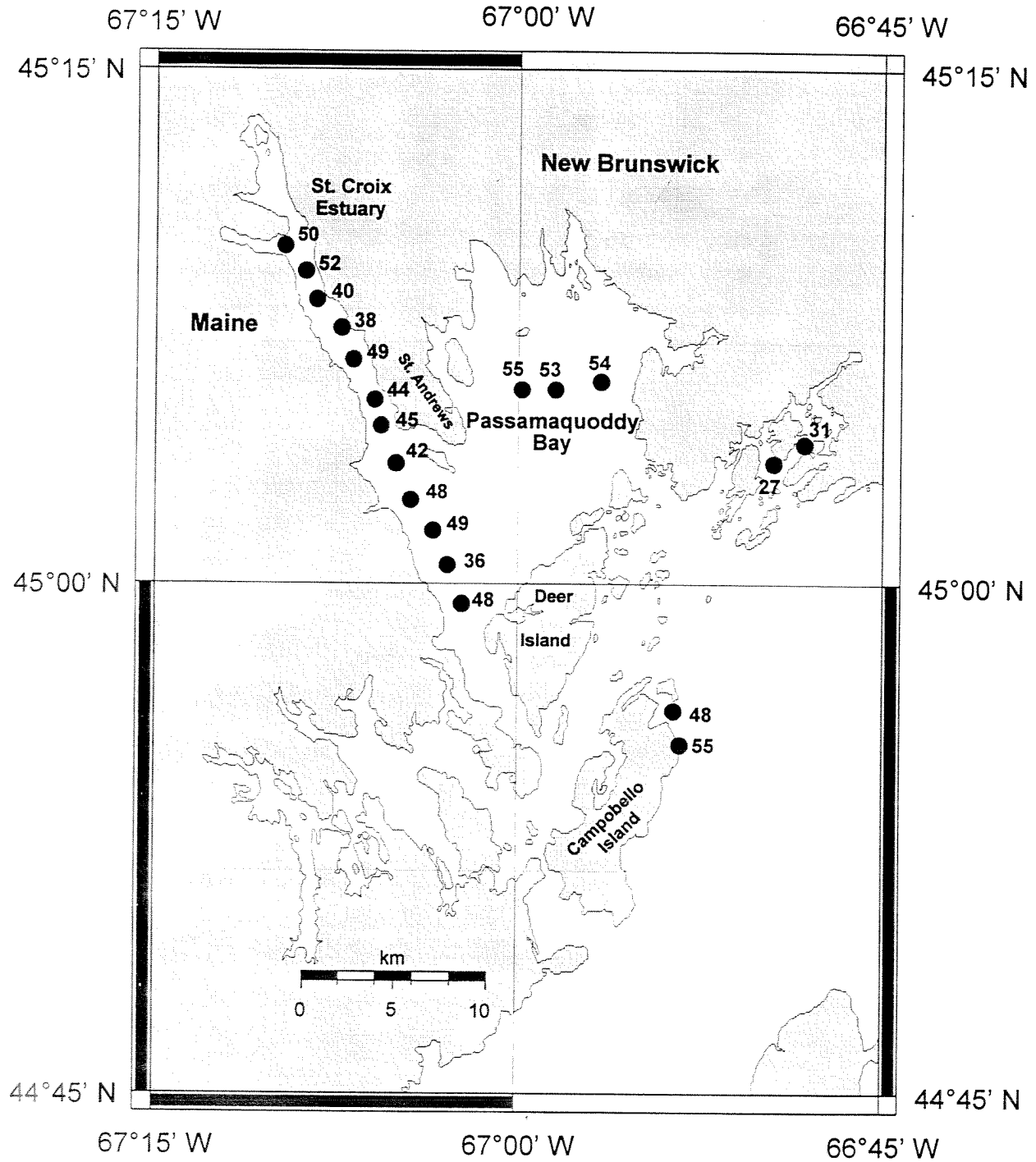


Figure 14: Lithium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Mn mg/kg

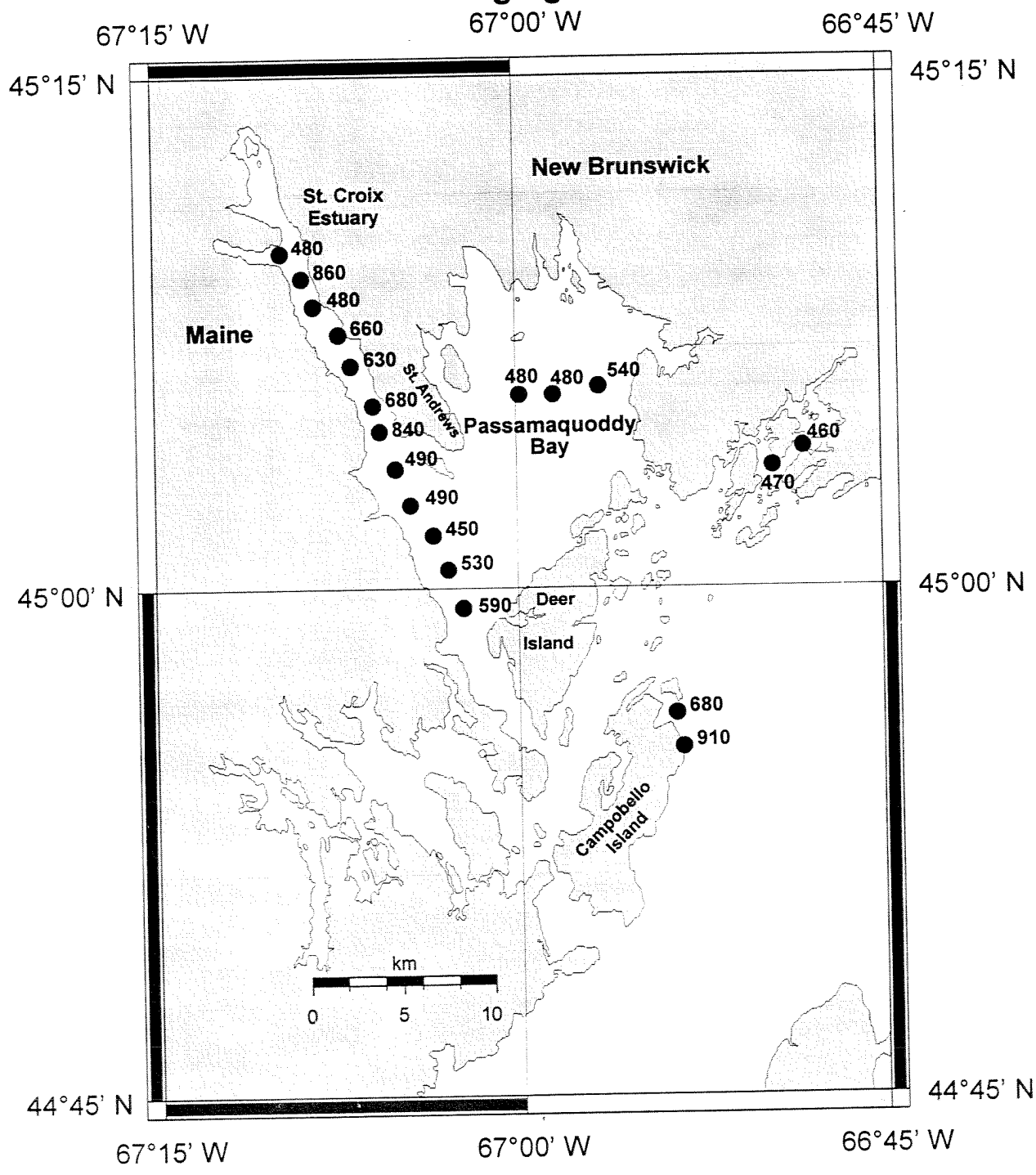


Figure 15: Manganese concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Ni mg/kg

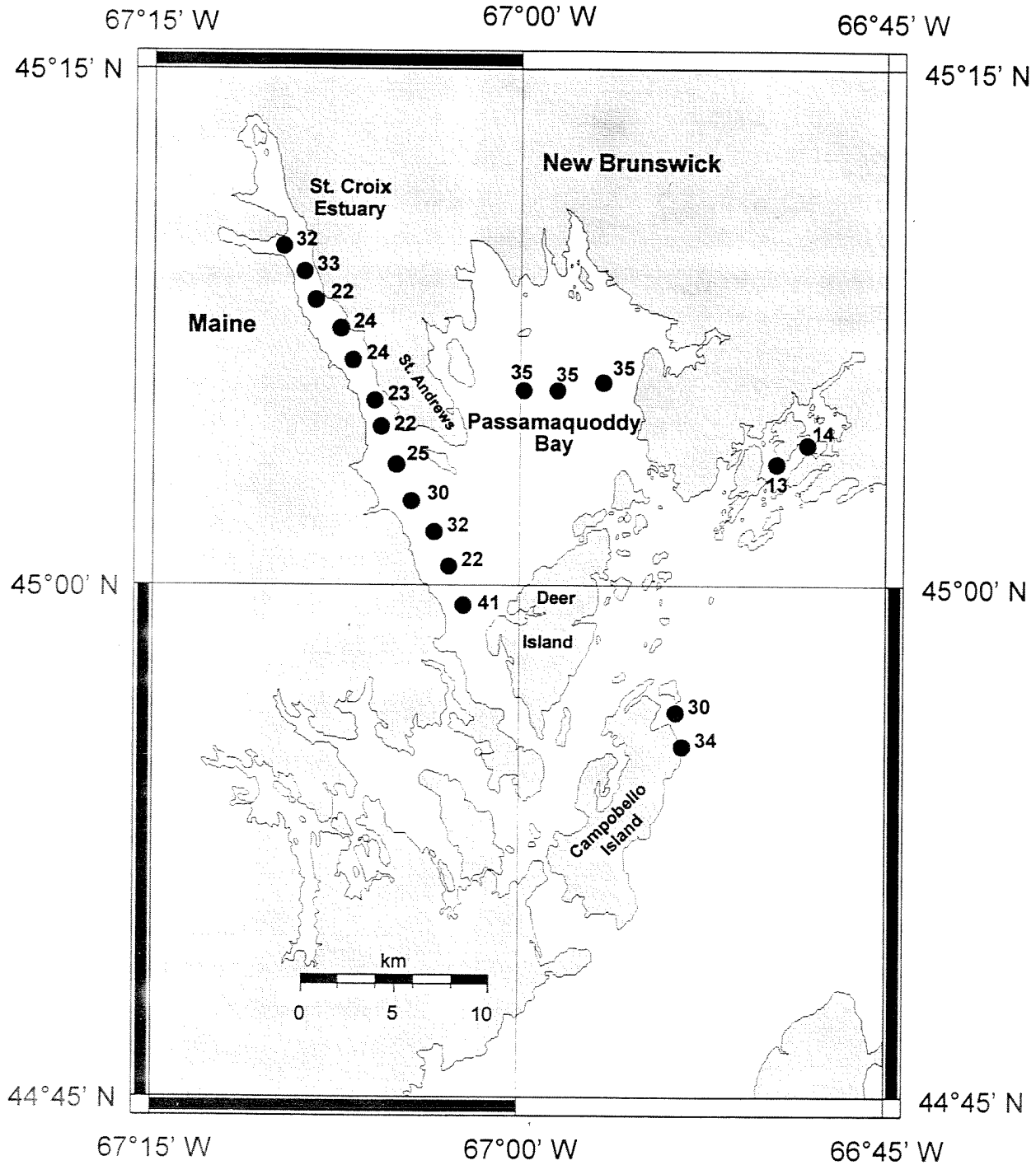


Figure 16: Nickel concentrations in surficial sediments of Passamaquoddy Bay.



# Passamaquoddy Bay Sr mg/kg

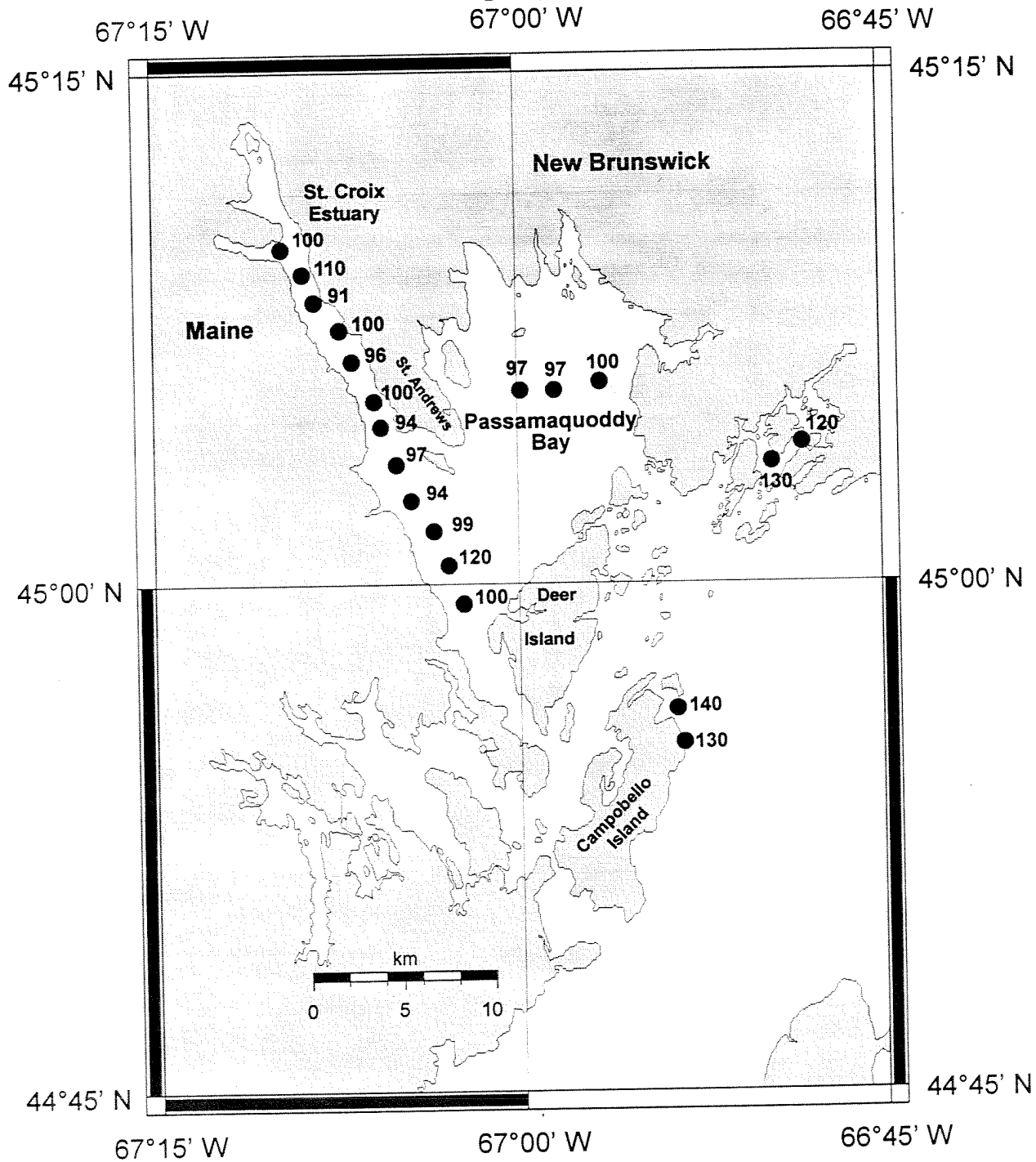


Figure 17: Strontium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Tl mg/kg

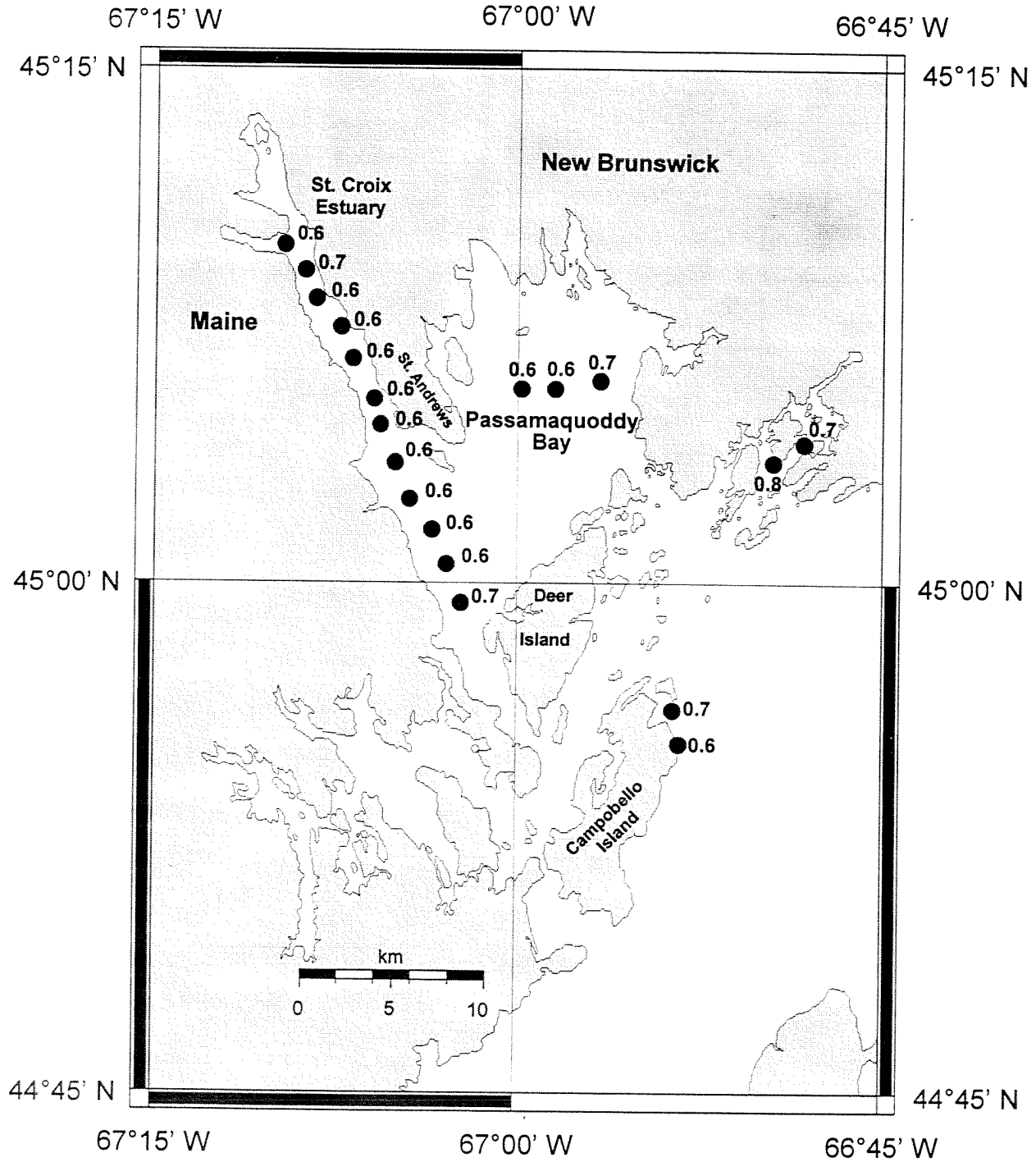


Figure 18: Thallium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay U mg/kg

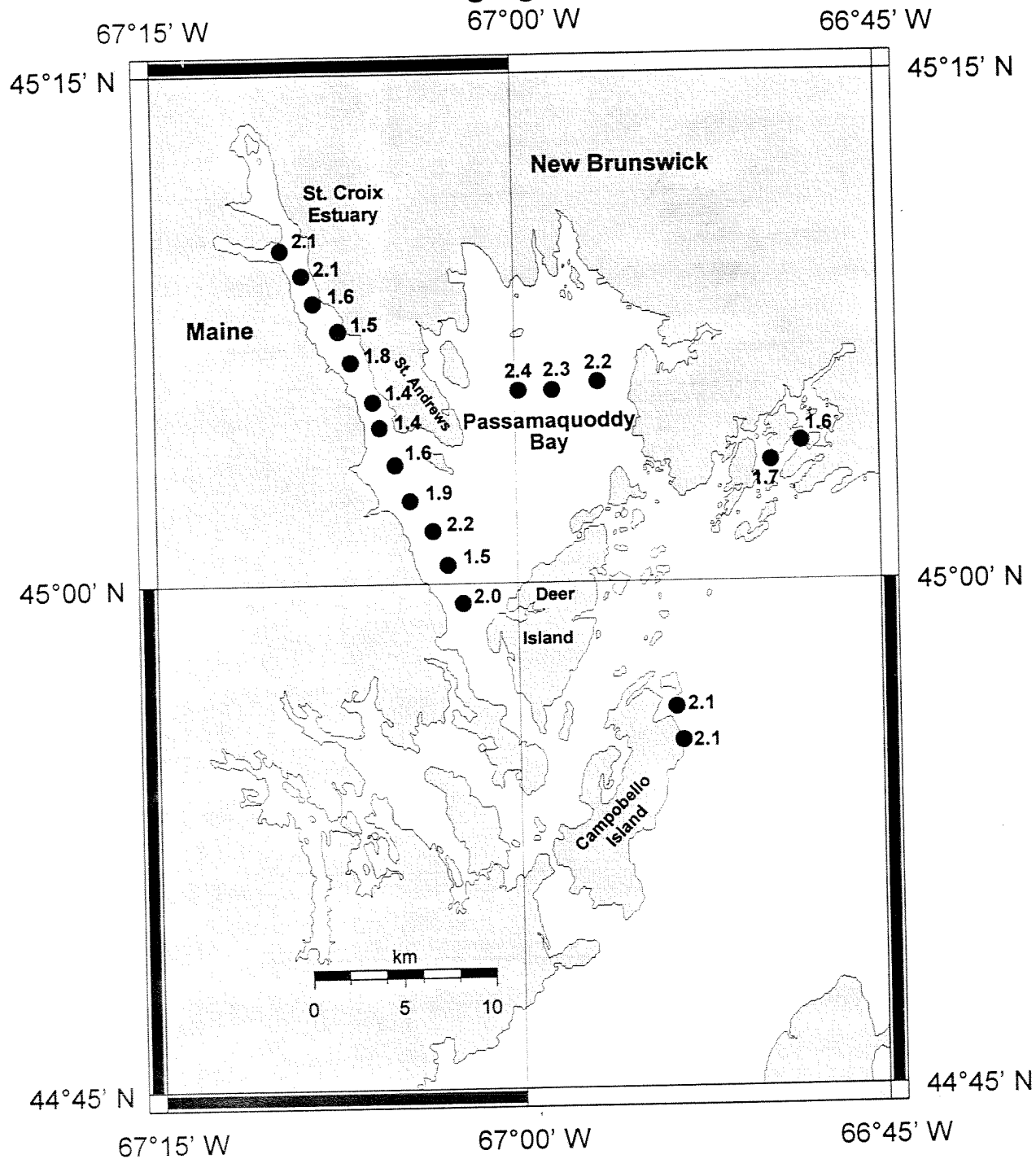


Figure 19: Uranium concentrations in surficial sediments of Passamaquoddy Bay.

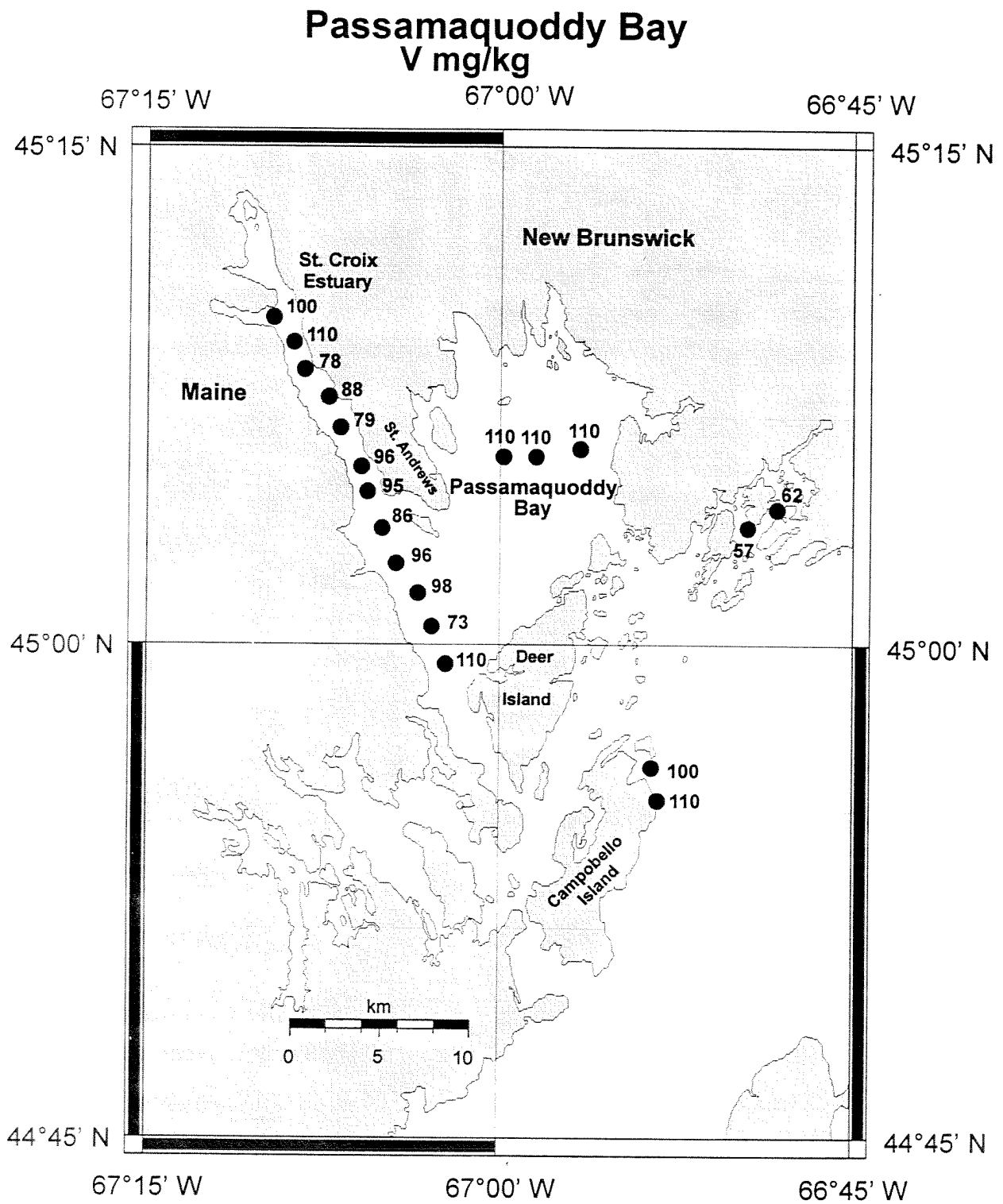


Figure 20: Vanadium concentrations in surficial sediments of Passamaquoddy Bay.

# Passamaquoddy Bay Zn mg/kg

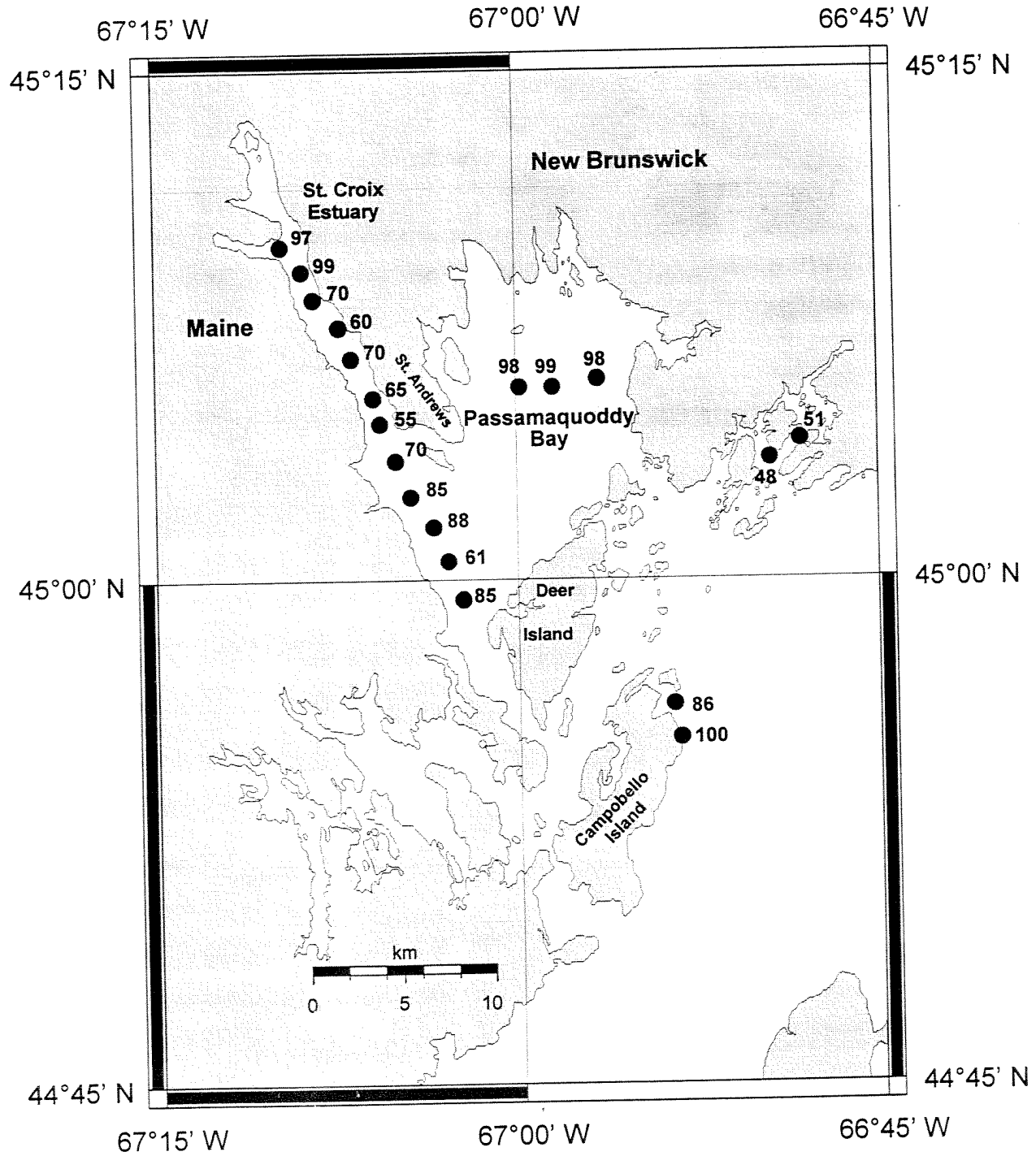


Figure 21: Zinc concentrations in surficial sediments of Passamaquoddy Bay.